Chapter I

Installing the Hardware Protection Key

Activating the Hardware Key and Personalizing the Program

Changing Hardware or Versions of the MacOS Installation

The application MacTempas and its associated files are installed by double clicking on the installer package. After authorizing the installer with the administrator password, the installer will install MacTempas into a directory in your applications folder. The driver for the hardware key will also be installed.

MacTempas uses a hardware copy protection key which must be installed on your computer. If you already have installed a key for use with CrystalKit, you do not need a second key to run MacTempas and you can proceed to the next paragraph describing how to activate the key for running MacTempas. Just plug the USB key into an open USB slot on your computer, keyboard or display.

When MacTempas is run for the first time it will put up its installation screen. Enter your name and affiliation as appropriate together with the installation code for the hardware key. T

If you have just changed your computer or installed a new clean version of the MacOS, you must ensure that the driver for the USB key is installed. Run the installation program for Mactempas once more to install the driver. Without the driver in place, the program will not recognize the hardware key and MacTempas will run in demonstration mode.

Ch. Installation - p.1

MacTempas User Manual

Ch. Installation - p.2

Chapter 1

Introduction to Image Simulation

The best High Resolution Transmission Electron Microscopes (HRTEM) have a resolution approaching 1 Å which sometimes leads to the erroneous conclusion that using an electron microscope, all atoms in a structure can be resolved. However, it is not the inter-atomic distances that matter, but rather the projected distances between atoms seen from the direction of the incident electron. In order to obtain interpretable results, it is necessary to orient the specimen such that atomic columns are separated by distances that are of the order of the resolution of the microscope or larger. This is a condition that very often is difficult to satisfy and often limits the use of the HRTEM to studies of crystals only in low order zone-axis orientations. The HRTEM image is a complex function of the interaction between the high energy electrons (typically 200keV - 1MeV) with the electrostatic potential in the specimen and the magnetic fields of the image forming lenses in the microscope. Although images obtained from simple mono-atomic crystals often show white dots separated by spacings that correspond to spacings between atomic columns, these white dots fall on or between atomic columns depending on the thickness of the specimen and the focus setting of the objective lens (O'Keefe et al., 1989). Fortunately, in many cases it is only necessary to see the general pattern of image intensities to gain the desired knowledge. However, in general, the image can be best thought of as a complex interference pattern which has the symmetry of the projected atomic configuration, but otherwise has no one-to-one correspondence to atomic positions in the specimen. It is because of this lack of directly interpretable images that the need for image simulation arose. Image simulation grew out of an attempt to explain why electron microscope images of complex oxides sometimes showed black dots in patterns corresponding to the patterns of heavy metal sites in complex oxides, and yet other images sometimes showed white dots in the same patterns (Allpress et al.,1972). This first application was therefore to characterize the experimental images, that is to relate the image character (the patterns of light and dark dots) to known features in the structure.

Most simulations today are carried out for similar reasons, or even as a means of structure determination. Given a number of possible models for the structure under investigation, images are simulated from these models and compared with experimental images obtained on a high-resolution electron microscope. In this way, some of the postulated models can be ruled out until only one remains. If all possible models have been examined, then the remaining model is the correct one for the structure. For this process to produce a correct result, the investigator must ensure that all possible models have been examined, and compared with experimental images over a wide range of crystal thickness and microscope defocus. It is also a good idea to match simulations and experimental images for more than one orientation.

The simulation programs can also be used to study the imaging process itself. By simulating images for imaginary electron microscopes, we can look for ways in which to improve the performance of present-day instruments, or even find that the performance of an existing electron microscope can be improved significantly by minor changes in some instrumental parameter. Alternatively, based on imaging requirements revealed by test simulations, we can adjust the electron microscope to produce suitable images of some particular specimen, or even of some particular feature in a particular specimen. Describing the Transmission Electron Microscope In order to simulate an electron microscope image, we need firstly to be able to describe the electron microscope in such a way that we can model the manner in which it produces the image. As a first step, we can consider the usual geometrical optics depiction of the transmission electron microscope (TEM).

Figure 1 shows such a diagram of a TEM operated in two distinct modes, set up for microscopy (a), and for diffraction (b). In microscopy mode we see that the TEM consists of an electron source producing a beam of electrons that are focused by a condenser lens onto the specimen; electrons passing through the specimen are focused by the objective lens to form an image called the first intermediate image (I1); this first intermediate image forms the "object" for the next lens, the intermediate lens, which produces a magnified image of it called the second intermediate image (I2); in turn, this second intermediate image becomes the "object" for the projector lens; the projector lens forms the greatly-magnified final image on the viewing screen of the microscope. In microscopy mode, electrons that emerge from the same point on the specimen exit surface are brought together at the same point in the final image.

At the focal plane of the objective lens, we see that electrons are brought together that have left the specimen at different points but at the same angle. The diffraction pattern that is formed at the focal plane of the objective lens can be viewed on the viewing screen of the TEM by weakening the intermediate lens to place the microscope in diffraction mode (b).



Figure 1. Geometrical optics representation of the TEM in imaging mode (a), and diffraction mode (b)

Ch. 1 Introduction to Image Simulation - p.6

Simplifying the Description of the Microscope

Consideration of the description of the electron microscope in figure 1 shows that the projector lens and the intermediate lens (or lenses) merely magnify the original image (I1) formed by the objective lens. For the purposes of image simulation we can reduce the TEM to three essential components; (1) an electron beam that passes through (2) a specimen, and then through (3) an objective lens (fig. 2).

Our next step in describing the electron microscope for image simulation is to move from the geometrical optics description of the TEM to a description based on wave optics. In this description of the microscope we examine the amplitude of the electron wavefield on various planes within the TEM, and attempt to determine how the wavefield at the viewing screen comes to contain an image of our specimen.

By treating the electrons as waves, and considering our simplified electron microscope (Figure 2), we see that there are three planes in the TEM at which we need to be able to compute the (complex) amplitude of the electron wavefield.

(1)The image plane:

Working backwards, we start at our desired information, the electron wavefield at the image plane; this wavefield is derived from the wavefield at the focal plane of the objective lens by applying the effects of the objective aperture and the phase changes introduced by the objective lens.

(2)The focal plane of the objective lens:

In turn, the electron wavefield at the focal plane of the lens is derived from the wavefield at the exit surface of the specimen by a simple Fourier transformation.

(3)The specimen exit surface:

In order to know the exit-surface wavefield, we must know with which physical property of the specimen the wave interacts, and describe that physical property of our particular specimen.

The Reduced Electron Microscope

Electron Microscope

Image Calculation



Fig. 2. The simplified TEM (left) and the calculations required for the image simulation (right). The three principal planes are marked.

Ch. 1 Introduction to Image Simulation - p.8

	Cowle electro called throug electro functio propag on the ous for the Sch	y and Moodie (1957) showed that the interaction of an on beam with a specimen could be described by the so- multislice approximation, in which electrons propagate h the specimen and scatter from the crystal potential, the on scattering is described by the so-called phase-grating on, a complex function of the potential, and the electron gation is computed with a propagation function dependent electron wavelength. Since then there have been numer- rmulations of the multislice approximation derived from hrödinger equation.
Simulating TEM Images	The pr compu micros images	oblem of simulating images thus becomes a problem of ting the electron wavefields (wavefunction) at three scope planes. Currently the best way to produce simulated s is to divide the overall calculation into three parts:
	(1)	Model the specimen structure to find its potential in the direction of the electron beam.
	(2)	Produce the exit-surface wavefield by considering the interaction of the incident electron wave on the specimen potential.
	(3)	Compute the image-plane wavefield by imposing the effects of the objective lens on the specimen exit surface wave.
	Each o ever, b everyth encour	of these steps will be covered in the next sections. How- because of space constraints, it is impossible to cover hing in great depth. For detailed derivation, the reader is raged to read the many excellent texts on the subject.

MacTempas User Manual

Ch. 1 Introduction to Image Simulation - p.10

Chapter 2

Modeling the Specimen

Theory of Image Simulation

The specimen is a three dimensional objects consisting of a huge number of atoms. From a modeling point of view, it is necessary to reduce the number of parameters to a more manageable number. For crystalline materials described by a repeat of perfect unit cells this is easily accomplished. The unit cell in this case is defined by the lattice parameters A, B and C where A and **B** are in the plane the specimen perpendicular to the electron beam and C is in the main direction of the incoming electrons. A,B and C are related to the normal lattice vectors **a**,**b**, and c depending on the orientation of the specimen. The specimen is thus reduced to M number of unit cells, where M*C is equal to the thickness of the sample, giving in the end a 2D image which covers the area given by A and B. In the case of a defect structure which no longer can be modeled as a small repeating structure, it is necessary to limit the extent of the calculation by defining a supercell which contains the defect. The resulting image obtained from the calculation will contain artifacts which arise from limiting the structure at arbitrary boundaries and care must be taken to ensure that the image gives a faithful representation of the area of interest. The entire electrostatic potential of the specimen is now defined by one unit cell with axes **a**,**b**, and **c**, angles *alpha*, *beta* and gamma, and N atoms with coordinates x,y,z. For simplicity, we use the nomenclature of the crystallographic unit cell even though we are referring to the transformed unit cell (A,B,C) as described above.

The electrostatic potential in the crystal can be written

$$\phi(\mathbf{r}) = d^3 \mathbf{r}' \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}$$

where (**r**), the charge density is:

$$\rho(\mathbf{r}) = \bigcap_{\substack{all \\ atoms \ i}} \rho_i(\mathbf{r} - \mathbf{r}_i)$$
2

with the sum extending over all atoms i at positions \mathbf{r}_i , each giving rise to a charge density

$$\rho_i(\mathbf{r}) = Z_i e \delta(\mathbf{r}) - e |\psi_i(\mathbf{r})|^2$$
3

where Z_i : atomic number, e: electronic charge, $\psi(\mathbf{r})$: the quantum mechanical many electron wavefunction for the atom. The potential (**r**) is described by its Fourier transform (u) through the relationship

$$\varphi(\mathbf{r}) = (\mathbf{u})e^{-2\pi i \mathbf{u} \cdot \mathbf{r}} d\mathbf{u} = (\mathbf{H})e^{-2\pi i \mathbf{H} \cdot \mathbf{r}}$$

since because of the periodicity of the unit cell, (**u**) is nonzero only when $\mathbf{u} = \mathbf{H} = h\mathbf{a}^* + k\mathbf{b}^* + l\mathbf{c}^*$, **H** being a reciprocal lattice vector.

The potential **(H)** is given as a sum over all atoms in the unit cell

$$(\mathbf{H}) = \int_{\substack{all\\atoms \ i}} f_i^{el}(\mathbf{H}) e^{2\pi i \mathbf{u} \mathbf{r}_i} = \frac{e}{4\pi^2 \varepsilon_0} \frac{Z_i - f_i^x(|\mathbf{H}|/2)}{\mathbf{H}^2} e^{2\pi i \mathbf{u} \mathbf{r}_i}$$

where the electron scattering factors f_i^{el} and the x-ray scattering factors f_i^x have been calculated from relativistic electron wavefunctions and parameterized. They can be found in various tables (Doyle and Turner, 1968) and are in use by most image simulation programs such as SHRLI (O'Keefe at al., 1978), NCEMSS (Kilaas, 1987) and EMS (Stadelman,). Taking into account any deviation from full occupancy at a particular site and the thermal vibration of the atom, the Fourier

coefficients of the crystal potential from one unit cell is calculated as:

$$(\mathbf{H}) = \int_{i}^{el} (\mathbf{H}) Occ(\mathbf{r}_{i}) \exp[-\mathbf{B}_{i} \mathbf{H}^{2}] e^{2\pi i \mathbf{H} \mathbf{r}_{i}} \qquad 6$$

B: Debye Waller factor; $Occ(\mathbf{r}_i)$: The occupancy at position \mathbf{r}_i

The interaction between an electron of energy E and the crystal potential (\mathbf{r}) is given by the Schrödinger equation

$$\left[-\frac{h^2}{8\pi^2 m}\right]^2 - e\phi(\mathbf{r}) \quad (\mathbf{r}) = E \quad (\mathbf{r})$$

where m is the relativistic electron mass and h is Planck's constant.

Before entering the specimen, the electron is treated as a plane wave with incident wavevector ${\bf k}_0,\,k_0$ =2 / , so that the incident electron wave is written

$$_{0}(\mathbf{r}) = \exp\{i(\omega t - 2\pi \mathbf{k}_{0} \mathbf{r})\}$$
8

It is useful to define the quantity $V(\mathbf{r})$ which will loosely be referred to as the potential as:

$$V(\mathbf{r}) = \frac{8\pi^2 me}{h^2} \phi(\mathbf{r})$$

Ch. 2 Theory of Image Simulation - p.13

Simulating the Interaction Between the Electrons and the Specimen The Schrödinger equation above cannot be solved directly without making various approximations. Depending on how the problem is formulated, one can derive the most common solutions to the electron wavefield at a position T within the specimen.

The Weak Phase Object Approximation

In the Phase Object Approximation (POA) (Cowley and Iijima, 1972), the phase of the electron wavefunction after traversing a specimen of thickness T is given as

$$(x, y, z = T)$$
 $(x, y, z = 0) \exp[-i\sigma V_p(x, y)T]$ 10

with

$$\sigma = 2\pi m e \lambda \ 1 + \frac{eE}{mc} \ / h^2$$
 11

where V(x,y) is the average potential per unit length. The specimen is considered thin enough so that electrons only scatter once and are subject only to an average projected potential. In the weak phase object approximation, the exponent is considered much less than one, so that the electron wavefunction emerging from the specimen is:

$$\psi(x, y, z = T) \quad \psi(x, y, z = 0)(1 - i\sigma V_p(x, y)T)$$
 12

The WPOA only applies to very thin specimens of the order of a few tenths of Å, depending on the atomic number of the atoms in the structure (Gibson, 1994). The FT of the wavefunction gives the amplitude and phase of scattered electrons and in the WPOA one has:

$$(\mathbf{u}) = \delta(\mathbf{u}) - i\sigma V_p(\mathbf{u})T$$
 13

where u is a spatial frequency.

Again, for periodic crystals, $V_p(\mathbf{u})$ are non-zero only for frequencies $\mathbf{u}=\mathbf{H}$ where \mathbf{H} is a reciprocal lattice vector in the crystal.

We will now use V to mean V_p . Thus for single electron scattering and when the Fourier coefficients V(**H**) are real (true for all centro-symmetric zone axis), the WPOA illustrates clearly that: i) Upon scattering, the electron undergoes a -90° phase shift. ii) The amplitude of a scattered electron is proportional to the Fourier coefficient of the crystal potential.

The Bloch Wave Approximation

In the BWA the electron wavefunction of an electron with wavevector \mathbf{k} is written as a linear combination of Bloch waves b(\mathbf{k} , \mathbf{r}) with coefficients (Howie, 1963). Each Bloch wave is itself expanded into a linear combinations of plane waves which reflect the periodicity of the crystal potential.

$$\psi(\mathbf{r}) = \sum_{j} \varepsilon^{(j)} b^{(j)}(\mathbf{k}, \mathbf{r}) = \sum_{j} \varepsilon^{(j)} c_{\mathbf{g}}^{(j)} \exp[-2\pi i (\mathbf{k}_{0}^{(j)} + \mathbf{g}) \mathbf{r}] \quad 14$$

The formulation above gives rise to a set of linear equations expressed as

$$[k_0^2 - (\mathbf{k}^{(j)} + \mathbf{H})^2]c_{\mathbf{H}}^{(j)} + V(\mathbf{H}')c_{\mathbf{H}-\mathbf{H}'}^{(j)} = 0$$
 15

which needs to be solved. Detailed derivation of the Bloch wave approximation can be found elsewhere.

Characteristics of the Bloch wave formulation are:

- Requires explicit specification of which reflections g are included in the calculation.
- Easy to include reflections outside the zero order Laue zone.
- Very good for perfect crystals, not suited for calculating images from defects.
- The solution is valid for a particular thickness of the specimen.

- Allows rapid calculation of convergent beam electron diffraction patterns.
- Includes dynamical scattering.

The Multislice Approximation

The multislice formulation (Goodman and Moodie, 1974 & Self et al., 1983) i s by far, the most commonly used method of calculating the electron wavefield emerging from the specimen. Although it does not as easily include scattering outside the zero order Laue zone as the BWA, the multislice formulation is more versatile for use with structures containing any kind of defects, either they be point-defects, stacking faults, interfacial structures, etc. The multislice solution gives the approximate solution to the electron wavefunction at a depth z+dz in the crystal from the wavefunction at z. In the multislice approximation one has:

$$\psi(x, y, z + dz) \quad \exp[-i\sigma dz \quad {}^{2}_{x,y}] \quad \exp[-i\sigma \quad {}^{z+dz}_{z}V(x, y, z')dz']\psi(x, y, z)$$
16

Thus starting with the wavefunction at z=0, one can iteratively calculate the wavefunction at a thickness n*dz, by applying the multislice solution slice by slice, taking the output of one calculation as the input for the next. Equation 16 is solved in a two step process.

The potential due to the atoms in a slice dz is projected onto the plane t=z, giving rise to a scattered wavefield

$$\psi_1(x, y, z + dz) = \exp\left[-i\sigma \sum_{z}^{z+dz} V(x, y, z') dz'\right] \psi(x, y, z) \quad q(x, y)\psi(x, y, z)$$
17

The function q(x,y) is referred to as the phasegrating.

Subsequently, the wavefield is propagated in vacuum to the plane t=z+dz, according to

$$\psi(x, y, z + dz) = \exp[-i\sigma dz \frac{2}{x, y}] \psi_1(x, y, z)$$
 18

The last equation represents a convolution in real space and is solved more efficiently in Fourier space (Ishizuka and Uyeda, 1977), where the equation transforms to

$$(\mathbf{H}, z + dz) = \exp[-i\pi\lambda dz\mathbf{H}^2] \quad {}_{1}(\mathbf{H}, z) \quad p(\mathbf{H}, dz) \quad {}_{1}(\mathbf{H}, z) \quad 19$$

where (\mathbf{H}, \mathbf{z}) are the Fourier coefficients of (x, y, z). $p(\mathbf{H}, dz)$ is called the propagator.

The multislice formulation is a repeated use of the last two equations and will give the wavefield at any arbitrary thickness T of the specimen. If the slice-thickness is chosen as the repeat distance of the crystal in the direction of the electron beam, only the zero order Laue reflections are included in the calculation as the unit cell content is projected along the direction of the electron beam. Three dimensional information which involves including higher order Laue reflections can be included by reducing the slice thickness (Kilaas et al, 1987).

Sampling Criteria

Any numeric calculation must be performed for a limited set of data points (x,y) or reciprocal spatial frequencies u. Working with periodically repeated structures; if the lateral dimensions of the unit cell is a and b, which we for simplicity make orthogonal so that the axes are associated with an orthogonal x,y coordinate system, then for a given sampling interval dx=dy, we have

$$N = \frac{a}{dx} \quad ; \qquad M = \frac{b}{dy} \qquad 20$$

defining the calculation to a grid of N*M points. The sampling interval automatically restricts the calculation in reciprocal space as well. The maximum reciprocal lattice vector for orthogonal axes is given as

$$\mathbf{H}_{\max}^{2} = \left| h_{\max} \mathbf{a}^{*} + k_{\max} \mathbf{b}^{*} \right|^{2} = \frac{N}{2a}^{2} + \frac{M}{2b}^{2} \qquad 21$$

Because most implementations of the multislice formulation makes use of Fourier transforms, the calculation grid N and M is adjusted so that both are powers of 2. This is because Fourier transform algorithms can be performed much faster for powers of 2 rather than arbitrary dimensions. This results in uneven sampling intervals dx,dy when a b. In order to not impose an arbitrary symmetry on the calculation, a circular aperture is imposed on the propagator. In practice, this aperture is set to 1/2of the minimum of (h_{max} , k_{max}) as defined above in order to avoid possible aliasing effects associated with digital Fourier transforms. The sampling must be chosen such that the calculation includes all (or sufficiently enough) scattering that takes place in the specimen.

After the electron wavefield emerge from the specimen, it is The Image Forsubjected to the varies magnetic field of the lenses that form the mation imaging and magnification part of the microscope. Of these lenses, only the first lens, the objective lens, is considered in the image formation calculation. Since the angle with which the electron forms with the optic axis of the lens varies inversely with the magnification, only the aberrations of the objective lens are important. The remaining lenses serve to just magnify the image formed by the objective lens. The effects of the lens which normally are included in the calculation are spherical aberration, chromatic aberration and lens defocus. Two-fold and three-fold astigmatism, including axial coma, are considered correctable by the operator although they can be included in the equations.

Without any aberrations, no instabilities and with the specimen

in the focal plane of the objective lens, the image observed in the electron microscope would be am magnified version of

 $I(x, y) = |\psi(x, y, z = exitplane \ of \ specimen)|^{2} = \psi_{e}(x, y)\psi_{e}(x, y)$ 22

Objective Lens Defocus

Consider an electron traveling from the plane defined by the exit surface of the specimen to the plane given as the plane of focus for the objective lens. This distance is referred to as the objective lens defocus f.



The electron traveling along the optic axis will have a path length of f while an electron that has been scattered an angle =H, will travel a distance f/\cos . This can be expressed as a phase difference

$$\frac{2\pi}{\lambda} \left(f / \cos \alpha - f \right) \pi \lambda f \mathbf{H}^2$$
 23

Spherical Aberration

Electrons crossing the optic axis with an angle a at the focal

plane of the objective lens should form parallel paths emerging from the lens.



However, the spherical aberration of the lens causes a phase shift relative to the path of the unscattered electron (=0) which is written as (Scherzer, 1949):

$$2\pi/\lambda \quad \frac{1}{4}C_x\alpha^4 = \frac{1}{2}\pi C_s\lambda^3\mathbf{H}^4 \qquad 24$$

If there were no other effects to consider, the image would be obtained as follows:

- Calculate the wavefield emerging from the specimen according to one of the approximations.
- Fourier transform the wavefield which gives the amplitude and phase of scattered electrons.
- Add the phase shift introduced by the lens defocus and the spherical aberration to the Fourier coefficients.
- Inverse Fourier transform to find the modified wavefunction.
- Calculate the image as the modulus square of the wave-field.

However, there are two more effects that are usually considered. Variations in electron energy and direction.

Chromatic Aberration / Temporal Incoherence

Electrons do not all have exactly the same energy for various

reasons. They emerge from the filament with a spread in energy and the electron microscope accelerating voltage varies over the time of exposure. The chromatic aberration in the objective lens will cause electrons of different energies to focus at different planes. Effectively this can be thought if as rather than having a given defocus f_0 , one has a spread in defocus values centered around f_0 . The value f_0 is what is normally referred to as f as indicating defocus. The images associated with different defocus values add to make the final image. Assuming a Gaussian spread in defocus of the form

$$D(f - f_0) = \exp[-\frac{(f - f_0)^2}{2}]$$
 25

gives:

$$I = |(f - f_0)|^2 D(f - f_0) df \qquad (\mathbf{H}) \qquad (\mathbf{H}) \exp[-1/2(\pi\lambda \mathbf{H}^2)^2]$$
26

This states that each Fourier term (diffracted beam) is damped according to the equation above].

Beam Divergence / Spatial Incoherence

The electron beam is not an entirely parallel beam of electrons, but form rather a cone of an angle . This implies that electrons instead of forming a point in the diffraction pattern form a disk with a radius related to the spread in directions. As for a variation in energy, the images formed for different incoming angles are summed up by integrating over the probability function for the incoming direction. It turns out that this also leads to another damping of the diffracted beam (Frank, 1973) so that:

$$I(\mathbf{r}) = \left| \psi(\mathbf{r}, \alpha) \right|^2 D(\alpha) d\alpha \qquad (\mathbf{H}) \qquad (\mathbf{H}) \exp\left[\pi \alpha \lambda (C_s \mathbf{H}^2 \lambda^2 + f) \right]^2 \qquad 27$$

The Final Image

Equation 26 and equation 27 are only valid when the intensities of the scattered beams are much smaller than the intensity of the central beam. Thus the image results from scattered beams interfering with the central beam, but not with each other. This is referred to as linear imaging. Although the formulation is slightly more complicated in the general case, the expressions above give sufficient insight into the image formation. Image simulation programs do however include the more general formulation which include non-linear imaging terms (O'Keefe, 1979). Each Fourier component is damped by the spread in energy and direction and the image is formed by adding this to the recipe in section 4.2

The Contrast Transfer Function CTF

When reading about HRTEM, it is impossible not to encounter the expression "Contrast Transfer Function". Loosely speaking, the CTF of the microscope refers to the degree with which Fourier components of the electron wavefunction (spatial frequencies) are transferred by the microscope and contribute to the Fourier transform of the image. Although the CTF only holds for thin specimen and linear imaging, it is often generalized and wrongly applied to all conditions. However, the CTF does provide insight into the nature of HRTEM images. In order to derive the expression for the CTF, we start by calculating the image intensity as given by the Weak Phase Object approximation. In the WPOA:

$$(x, y, z = T) \quad 1 - i\sigma V_p(x, y)T \qquad 28$$

and

$$(\mathbf{H}) = \delta(\mathbf{H}) - i\sigma V_{n}(\mathbf{H})T$$
²⁹

Applying the phase shift due to the spherical aberration and the

objective lens defocus which we will call (**H**), we get that the FT of the wavefunction is (for simplicity $V = V_p$):

$$(\mathbf{H}) = \delta (\mathbf{H}) - i\sigma V(\mathbf{H})e^{i\chi(\mathbf{H})}A(\mathbf{H})$$
 30

where $A(\mathbf{H})$ is the damping terms arising from partial coherence.

The FT of the intensity is now given as

$$I(\mathbf{H}) = FT(\psi \ \psi \) = (\mathbf{H}' \) * (\mathbf{H} - \mathbf{H}')$$

$$= \int_{\mathbf{H}} (\delta(\mathbf{H}') - i\sigma A(\mathbf{H}')V(\mathbf{H}')e^{i\chi(\mathbf{H}')}) \left(\delta(\mathbf{H} - \mathbf{H}') - i\sigma A(\mathbf{H} - \mathbf{H}')V(\mathbf{H} - \mathbf{H}')e^{i\chi(\mathbf{H} - \mathbf{H}')}\right)$$

$$= \int_{\mathbf{H}} \delta(\mathbf{H}') + 2\sigma A(\mathbf{H})V(\mathbf{H})\sin\chi(\mathbf{H})$$

\mathbf{a}	1
3	L
-	_

The last result is very useful and it leads to the frequently used concept of the Contrast Transfer Function (CTF). The CTF is defined as $A(\mathbf{H}) \sin(\mathbf{H})$ The equation above states that each reflection \mathbf{H} contributes to the image intensity spectrum with a weight that is proportional to the CTF. Figure 3. shows a plot of a CTF including sin and the damping curves. When sin $(\mathbf{H}) = -1$ for a large range of frequencies \mathbf{H} , which is the condition referred to as Scherzer defocus[11], the image can be thought of as:

$$I(x, y) \quad 1 - 2\sigma U(x, y) \tag{32}$$

where U(x,y) is a potential related to the original crystal potential, but keeping only the Fourier coefficients related to frequencies transferred by the microscope. The equation above shows the often used rule of thumb. For thin specimens, under Scherzer imaging conditions, atoms are black.



Figure 3. Plot of the Contrast Transfer Function for a 200kV microscope with the parameters indicated.

References

References

Allpress J.G. et al (1972) n-beam Lattice Images. I. Experimental and Computed Images from W₄Nb₂₆O₇₇, Acta Cryst. A 28, 528-536 Cowley J.M. and Iijima S. (1972) Electron microscope image contrast for thin crystals, Z. Naturforschung 27a, 445-451 Doyle P.A. and Turner P.S. (1968) Relativistic Hartree-Fock Xray and Electron Scattering Factors, Acta Cryst.. A 24, 390-397 Frank J (1973) The envelope of electron microscope transfer functions for partially coherent illumination, Optik 38, 519-536 Gibson J.M. (1994) Breakdown of the weak-phase object

approximation in amorphous objects and measurement of highresolution electron optical parameters, Ultramicroscopy 56, 26-32

Goodman P, Moodie A.F. (1974) Numerical evaluation of Nbeam wave functions in electron scattering by the multislice method, Acta Cryst. A30, 322-324

Howie A. (1963) Inelastic scattering of electrons by crystals, Proc. Roy. Soc. A271, 268-275

Ishizuka K. and Uyeda N. (1977) A new theoretical and practical approach to the multislice method, Acta Cryst. A 33, 740

Kilaas R. et al. (1987) On the inclusion of upper Laue layers in computational methods in High Resolution Transmission Electron Microscopy, Ultramicroscopy 21, 47-62

Kilaas R. (1987) Interactive software for simulation of high resolution TEM images, Proc. 22nd MAS, R. H. Geiss (ed.), Kona, Hawaii, 293-300.

Kilaas R. (1987) Interactive simulation of high resolution electron micrographs. In 45th Ann. Proc. EMSA, G.W. Bailey (ed.), Baltimore, Maryland, 66-69.

O'Keefe MA, Buseck PR, Iijima S (1978) Computed crystal structure images for high resolution electron microscopy. Nature 274, 322-324.

O'Keefe M.A. (1979) Resolution-damping functions in non-linear images, Proc. of EMSA 37, 556-557

O'Keefe M.A. et al. (1989) Simulated Image Maps for use in Experimental High-Resolution Electron Microscopy, Mat. Res. Soc. Symp. Proc. 159, 453-458

Scherzer O. (1949) The Theoretical Resolution Limit of the Electron Microscope, Journal of Applied Physics 20, 20-29

Self P.G.et al. (1983) Practical computation of amplitudes and phases in electron diffraction, Ultramicroscopy 11, 35

MacTempas User Manual

Chapter **?**

The Three Simulation Steps

Calculate Tables Quanti

Full Calculation Projected Potential Exit Wavefunction(s) Image(s) Image-Plane Wavefunctions(s)

Weak Phase Object Images... Integrated Diffraction Pattern... CBED Pattern...

Introduction to MacTempas

Since the simulation process can be subdivided into independent calculations involving the structure, the scattering process and the imaging process, MacTempas allows one to invoke these independent calculations separately through the "Calculate" menu.

Full Calculation

This command will start the calculation from the required starting point and proceed to calculate finale images.

Projected Potential

generates the crystal potential that produces electron scattering from the structural data, unit cell dimensions, symmetries, and atom positions, occupancies, and temperature factors.

Exit Wavefunctions(s)

generates the electron wavefield at the specimen exit surface; it uses projected potential combined with information about the accelerating voltage of the electron microscope, and the specimen thickness and tilt. The computation algorithm is the multislice approximation.

Image(s)

generates the image intensity at the microscope image plane; the effects of the objective lens phase changes and resolutionlimiting aberrations are included via parameters like defocus, spherical aberration, incident beam convergence, spread of defocus, and the position and size of the objective aperture.

Image Plane Wavefunctions(s)- generates the electron wavefunction at the imaging plane in the microscope. This is equiva-

	lent to the application of the Contrast Transfer Function to the Fourier transform of the electron wavefunction at the exit sur- face of the specimen followed by an inverse Fourier transform. The calculation of the image plane wavefunction is used for comparing with the electron wavefunction found by the use of electron holography.
	Thus " <i>Projected Potential</i> " calculation considers only the spec- imen structure, " <i>Exit Wavefunctions(s)</i> " calculation treats the interaction of the specimen with the electron wave, and the " <i>Image(s)</i> " calculation simulates how the wave leaving the specimen interacts with the lens system of the electron micro- scope. Once a simulation has been made, any additional simula- tion will usually not require a full re-calculation; any change in microscope parameters will not affect the results of the " <i>Pro- jected Potential</i> " and " <i>Exit Wavefunctions(s)</i> " calculations, and only Image(s) will need to be re-run; any change in microscope voltage or in specimen thickness and tilt will not affect the out- put of " <i>Projected Potential</i> ", but " <i>Exit Wavefunctions(s)</i> " and " <i>Image(s)</i> " will need to be re-run. Of course, any change in the specimen structure will require the re-running of all three sub- programs.
Generated Files	MacTempas generates and stores various files in the course of a simulation. The 6 possible data files are:
	(1) < structurename>.at stores all the structure and micro- scope information needed to run the simulation. This information is derived from user input and the supplied data files. In particular, the string " structurename " is a unique name for the structure, input by the user when creating the structure file. This is an editable file of type

'TEXT'.

(2) <structurename>.pout is the result of running the projected potential routine from the information stored in <structurename>.at; it contains the specimen potential in the direction of the electron beam. This is a BINARY file of type Real 4. The first 80 bytes consists of record information and the data starts at byte 80. The first line of data contains the data for the bottom line of the "image" since the coordinate system for MacTempas is at the lower left corner of the image/unit cell. Thus if the data is imported into a program for viewing, the image will appear flipped.

- (3) <structurename>.mout is the result of running the multislice routine using the data in <structurename>.pout with those in <structurename>.at; it contains the exit-surface wavefunction at one or more selected specimen thicknesses. This is also a BINARY file with the same structure as <structurename>.pout, except for the fact that the data is complex, pairs of numbers (real and imaginary). The data starts at byte 80 and the file can contain more than one exit wavefunction.
- (4) <structurename>.iout is the result of running the image formation routine to apply the effects of the microscope parameters in the <structurename>.at file to the exit-surface wave; it contains one or more images ready to be displayed. This again is a BINARY file with data starting at byte 80 and the file can contain more than one image. Data is Real 4
- (5) **<structurename>.hout** is the result of calculating the image plane electron wavefunction(s) instead of calculating the simulated images. The data is complex, pairs of numbers (real and imaginary). The data starts at byte 80 and the file can contain more than one image plane exit wavefunction.
- (6) **<structurename>.aout** contains the complex amplitudes of several diffracted beams at one-slice increments in specimen thickness. The beams are specified by the

user, and can be plotted as a function of specimen thickness.

In addition, two "print" files are produced (but rarely printed) just in case additional information about a computation is required by the user. These files are:

- (7) <structurename>.p_prnt contains information about the way in which the "*Projected Potential*" subprogram processed the <structurename>.at data to produce the specimen potential.
- (8) <structurename>.m_prnt contains information about the way in which the "*Exit Wavefunctions(s)*" subprogram processed the <structurename>.pout data with the <structurename>.at to produce the exit-surface wave; that is, it contains information from the multislice computation.

Chapter 4

Running MacTempas

The first step in running a simulation is generating the structure input file. This is done through **New Structure File...** in the FILE menu. This generates the input dialog window with values for a default cubic structure. Use this template to modify the date to fit your structure..

Simulation Estameters

Generating an Input Structure

Crystal Parameters	Specimen Parameters
A (Å) 4.0000 Alpha (deg.) 91.00 B (Å) 4.0000 teta (deg.) 31.00 C (Å) 4.0000 Gamma (deg.) 91.00 Spacegroup # det. Tabled 225 # of Atoms in Basis 1 28000 # of Symm. 0pt. 192 28000 # of atoms in UCell 4 36000	Direction 3-locker 0 0 1 0 0 1 Mumber of Sloce per online 1 1 1 1 Graze (A-L) 2.00 1 2.00 1 1 Graze (A-L) 2.00 1.00 0 1.01 1 1.01 1.03
F of different alores 1	Type of Absorption New
Microscope And Lens Parameters Microscope Name <u>4000Ex</u> Valuage (N/) 400 Cs (mm) L00 Convergence angle (mrad) 0.55 Spread of defocus (A) IID Defocus (beg, inc, end) (A) -000 D	Anternation (k) / Crime angle [1] More angle [1] Two faild 0 Three faild 0 Come 1 0
Obj. Issna apert, rad. (A-1); 0.70 Barr, of Obj. Lans Apr. 0.00 0.10 0.10 0.00 Cent. of Obj. Lans Apr. 0.00 0.10 <td>Bigmus of Ga,00 0.00 0.00 Angle with x-axis [*] 0.0</td>	Bigmus of Ga,00 0.00 0.00 Angle with x-axis [*] 0.0
Over-ride calculation status Mark for re-takelation	Cott Neverfunction Image

a, b, c,

alpha, beta, gamma

These are the unit cell dimensions in Ångstrøm units, and the unit cell angles in degrees. MacTempas will automatically set the angles depending on the spacegroup, if possible. The program will also automatically set lattice parameters depending on the spacegroup. Thus if the user chooses a cubic system, **b** and **c** are set equal to **a**

Space group#

MacTempas generates symmetry operators for the any one of the 230 space groups when selecting the number or the symbol of the space group (as listed in the International Tables for Crystallography). By clicking on the pop up menu "Space Group" one can choose one of the 230 spacegroups by first selecting the type of crystal-structure, i.e. hexagonal or cubic. The user can choose one of the spacegroups by clicking on the symbol for the spacegroup or by entering the number for the spacegroup.

Simulation Farameter	873								
Crystal Parameters Spectroscope A (A) 4.0000 Alpha (deg.) 91.00 B (A) 4.0000 Beta (deg.) 91.00 B (A) 4.0000 Beta (deg.) 91.00 C (A) 4.0000 Gamma (deg.) 91.00 Spacegroup P det. Tableto 225 Other/Im P of Atoms in Basis 1 Stare Tragata P of Symm. Ope 192 Stare Tragata	cimen Paran sens faceout lo 0 ber of Sices M. W. al. ba al. ba	1 0 percell red 100 ses ()	3-4 0 5et 0.00	0 1 t 200 L00 k 0.00					
e of different stores 1 Tage Nicroscope and Less Parameters Nicroscope Name 400000 N Voltage (NV) 400 Ca (mm) L00	Aotom	PE3 PH3 Nam Path Nam	123 123 124 124 124 124	115 743 743 743 743 743 74	1253 100 100 100 100 100 100	2.j 142 Min Pals	Pal) P4j() Pila Pala	Pell NAS Stree Tender	Paul PA,S PA
Convergence wright (mrwal) 0.55 Spenal of defecus (A) 0.0 Defecus (beg,inc,eed) (A) 0.00 Db, inne apert, rad (A-1) 0.00 Served (A) 0.00 Came, of Obj, Lans April 0.00 Came, of Obj, Lans April 0.00 Came, of the Dyric Asis 0.00	Three S Corea Michae Sigma o Angle w	⊡Secon F (a,b) Ith x-asi	0,00 0	Spore	Group Ni	imber i İn	tern Tab	kat [2	08
Over-ride calculation status Nark for re-talculation Projected Potential Nark as calculated Projected Potential	Exit Wavefur Exit Wavefur	notian Notian		le le]				

The input also allows for choosing the second setting for a specific spacegroup if one exists. If no space group is required, one should use the space group P1 (1), in which case the only symmetry operator is x,y,z. Additional symmetry operators can be entered by opening the dialog displaying the symmetry operators.

Show (Basis atoms)

Simulation	Paramet	873.					
Crystel Parameters				Atoms in the	Motif/Basis		
A (Å) 38140 Alpha (dep.) 90.00		Name	x-coord.	y-coord.	z-coord.	dw-fact.	Occ.
C [A] 30.5200 Gamma [deg.] 90.00	1	Ca	0.000000	0.000000	0.000000	3.600000	1.00000
Spacegroup # Ort. Tablesi 130	2	ŝr	0.0000000	0.000000	0.109700	3.600000	1.00000
# of Atoms in Basis 9 (these)	3	Di	D.DODDOD	0.000000	0.302200	3.600000	D.87000
# of Symm. Ops. 32 (How)	4	84	0.000000	0.000000	0.2681.00	3.600000	0.13000
# of atoms in UCell 38 (See	5	CV	0.0000000	0.000000	0.445600	3.600000	1.00000
# of different access 5	б	0	0.500000	0.000000	0.446000	3.4000000	1.00000
	7	0	0.0000000	0.000000	0.375000	5.6000000	1.00000
Microscope and Lens Parameters	8	0	0.000000	0.000000	0.205000	3.6000000	1.00000
	0	0	0.500000	0.000000	0.250000	3.600000	0.06500
Vacage [kV] 300 Li (mm) 0.0			1		1	1	1
Spread of defocus (Å)			1		-	1	1
Defecus dealing each Mi -2400 -100					-		
Obi, lans apert, rad, (A-1) 1.25		1	1		18		
b b Mag. Ar			1		16		1
Cent. of Obj. Lens Aprt. 0.00 0.00 0.00 1			1	1			
Cent, of the Optic Asis 0.00 0.00 0.00 1			1	- 1	1		1
Over-ride calculation status		-	1				
Mark for re-calculation 📃 Projected Potenti		222	1 Ands all				
Mark in admittance I Development Bernard		600	(Delete			(Cancel)	OK

Use this button to bring up the dialog window that enables the input of the atoms in the basis.

Number of Atoms in the Basis

This value is the number of independent atom positions in the basis or asymmetric unit of the cell. When operated on by the symmetry operators, the basis generates all the atom positions within the cell. This value is never modified by the user since the program always recalculates this number depending on the data entered.

Show (Symmetry Operators)

The symmetry operators are automatically created by specifying the spacegroup. By clicking on this button, a window dis-

Simulation Pa	rameters		
Crystal Parameters	Specimen Parameters		
B(A) 3.8140 Beta (deg.) 90.00	Symmetry	y operator	5
C (A) 50.5200 Gamma (deg.) 50.00		100 7	
e of Atoms in Basis 9 (Show)	1 ×9,2	16	-9+1/2,-x+1/2,-2+1/2
A of Symme One S2	2 x+1/2,y+1/2,z+1/2	17	-x,-y,-z
e of stoms in Fall 38	3 -X, -Y,2	18	-x+1/2,-y+1/2,-2+1/2
a of different storms	4 -x+1/2,-y+1/2,2+1/2	19	X,Y,-2
e or omerent atoms 3	5 -γ.x.z	20	x+1/2,y+1/2,-z+1/2
Microscope and Lens Parameters	6 -y+1/2,x+1/2,2+1/2	21	ý,-X,-2
Microscope Name CM300	7 y,-x,z	22	y+1/2;-x+1/2,-2+1/2
Voltage (kVI 300 Cs Inter) 0.50	8 y+1/2,-x+1/2,z+1/2	23	-7,x,-z
Convergence angle (mrad) 0.20	9 -x,y,-z	24	-y+1/2,x+1/2,-z+1/2
Spread of defocus (Å) 30	11 -x+1/2,y+1/2,-z+1/2	25	x,-y,z
Defocus ibeg inc. endi IAI -2400 -100 -	10 ×72	26	x+1/2,-y+1/2,2+1/2
Obi, lens apert, rad. [A-1]: 1.25	12 x+1/2,-y+1/2,-z+1/2	27	-x.y.t
h k Mug. Ang	13 Y.X2	28	-x+1/2,y+1/2,z+1/2
Cent. of Obj. Lens Aprt. 0.00 0.00 0.00 0.0	14 y+1/2×+1/2,-z+1/2	29	-7%2
Cent. of the Optic Axis 0.00 0.00 0.00 0.0	15 -72,-2	30	-y+1/2,-x+1/2,z+1/2
Over-ride calculation status			
Mark for re-calculation Projected Potential	Page #		
Mark as calculated Projected Potential	(<<) Delete)	1	Cancel OK

playing the symmetry operators are shown.

Show (Atoms in Unit Cell)

The atoms in the unit cell are automatically created by the operation of the symmetry operators on the atoms in the basis. The number of atoms is given and by clicking on the button "Show",

201	· ac at the			_				
stal Parameters	Spe	cimen Paramet	ers	2001 - I				
Alpha Idea 1 90.00	Zene	e-axis [uvwtti]		-Index				
1 2 81 40 Bass [days] 90.00	11	0 11 10	0	0 1 0	10.00 0.00			
1 20 5000 George (upp)				woms in th	e unit Cell			_
(j 30.5200 Gamina (deg.) 90.00	10.11	100000000	20000	0.000	12-11-11-1		1202	
cegroup # (Int. Tables) 139		Type Name	X-coord.	Y-coord.	Z-coord.	DM-L	oca	
Atoms in Basis 9 Show	1	1 00.	0.00000	D. DOG DG	0.00000.0	5 600	1.000	
Symm. Ops. 32 Show	2	I Ca	0.50000	0.50000	0.50000	3.600	1:000	
atoms in UCell 38	3	2 Sr	0.00000	0.00000	0.10970	3,600	1.000	
different stores N	4	2 Sr	0 50000	0.50000	0.60970	3.600	1.000	
dimerens atoma i a	5	2 . 97	0.00000	0.00000	0.89030	3.600	1.000	
and an	6	2 Sr	0.90000	0.500.00	0.39030	3.600	1.000	
roscope and Lens Parameters	7	5 BI	0.00000	D.D00D0	0.30220	3.600	0.870	
rescope Name CM300	6	3 BI	0.90000	0.500.00	0.90220	3.600	0.870	
ane kMI 200 Ce Immi 0.5	9	S 84	0.00000	0.00000	0.65780	5.600	0.870	
age way soo us (mini) ole	10	3 Di	0.50000	0.50000	0.19780	3.600	0.870	
vergence angle [mrad] 0.20	11	3 BI	0.00000.0	0.00000	0.26810	3.600	0.130	
ead of defocus (A) 30	12	3 Di	0.50000	0.50000	0.76810	3.600	0.130	
ocus (bea.inc.end) (Å) -2400 -100	13	5 Di	d dabab	D. DOG DG	0.75190	3.600	0.150	
lans source and (A-1) 1.25	14	a ba	0.90000	0.500.00	0.28190	5.600	0.130	
sens apert, rau, perg. 1.23	15	4 CE	0.00000	D. DOG DO	0.4-860	5.600	1.000	
h k Mag. A	16	4 68	0.90000	0.50000	0.94560	3.600	1,000	
t. of Obj. Lens Aprt. 0.00 0.00 0.00 0	17	4 08	0.00000	0.00000	0.55440	3.000	1 000	1
t. of the Optic Axis 0.00 0.00 0.00 0	10		0.50000	0.00000	0.45600	3.000	1 000	<
er-ride calculation status	20	5 0	0.00000	0.50000	0.94600	\$ 600	1.000	
k for re-calculation 📃 Projected Potenti	- horizon		3120335	1000000				
k as calculated Potenti	J 🖸	Exit Wavefuncti	on 🗆 Im	age				

a window displaying the atoms in the unit cell appears.

Number of different atoms

This value is the number of different types of atoms in the specimen structure; difference is due to a different atomic number or a different Debye-Waller factor. The correct value is calculated by MacTempas and displayed.

Zone Axis

Specimen orientation in relative real space axes units.

Number of slices per unit cell

For unit cells with large repeat distances in the beam direction, moderate values of G_{max} may allow the Ewald sphere to approach the so-called pseudo upper-layer line that the multi-

slice allows at the reciprocal of the chosen slice thickness. In this case MacTempas will sub-divide the slice into two or more subslices. How this is done depends upon the potential setting chosen in the Option menu.

Gmax

The maximum value (in reciprocal Ångstrøm units) of any scattering vector to be included in the multislice diffraction calculation. This value imposes an "aperture" on the diffracted beams included in the dynamic scattering process. It should be large enough to ensure that all significant beam interactions are included. The default value is 2.0. MacTempas will compute phase-grating coefficients out to twice G_{max} in order to avoid aliasing in the multislice calculations.

Specimen Thickness

The thickness of the specimen foil is entered as a beginning thickness, an ending thickness and an incremental thickness. All numbers are in Ångstrøm units. A series of thicknesses represented by the upper and lower bounds and a thickness step; e.g. 100 50 250 will cause MacTempas to store the exit wavefield at specimen thicknesses of 100Å to 250Å in steps of 50Å (a total of four thicknesses).

Store Ampl./Phases - Set...

Clicking this button allows a number of diffracted beams to be selected for plotting of their intensity and phase variation as a
Specim Zone-axi	ten Param	eters	3	Inde		1				
Zone-axi	is [uve0)]		3	Inde	()					
0			1.	and the second second						
And the second sec	and the state	0 0		1 1	0					
Number	r of Slices	per cell								
Gmax [/	A-1]			- 4	2.00					
Thick. (beg.inc.en	d) 30	0		Sto	re am	plitude	s and p	phases	for
Store A	mpl./Phase	es 🤇	Set		h	k	1	h	k	1
Cent. of	f Laus Cin	cle h	0.00		0	Q	D			
Eq. Tilt	[mrad] & a	ingle "	0.00		2	D	-2		8-3	-
Type of	Absorptio	n [N	DF	1	D	-1			
		100				1	1996			
1	Attignatis	m [Å] / C	DIT18							
		Mag	4	/8g //h					0	ear
	Two fold	0	1	0.0			-		-	_
-	Three fold	0	- 1				Ca	ncel	\in	ж
	fame.	1000		20						
2700	Coma	1000	11	YA						
E	. Mechanica	Vibratio	• (A) _	_						
<u></u>	Sigma of	(a,b)	0.00	0.0	0					
-	Angle wit	h x-axi	11	0.0	-					
	unite m	1.022233	22.5	2000		_				
				_		-				
Exi	t Wavefund	tion	🕀 Ima	ge						
Exi	t Wavefund	tion	🖂 Ima	ge						
	Thick. I Store A Cent. o Eq. Tilt Type of 700	Thick. (beg.inc.en Store Ampl./Phase Cent. of Law Cin Eq. Tilt [mrad] & a Type of Absorptio Antigratis Two fold Three fold Coma Mechanica Sigma of Angle wit Exit Wavefunc	Thick. (beguinc,end) 30 Store Ampl./Phases Cent. of Laue Circle h Eq. Tilt [mrad] & angle " Type of Absorption Autignatism [Å] / C Medhanical Mission Sigma of (a,b) Angle with x-axis Exit Wavefunction Exit Wavefunction	Thick. (beg.inc.end) 30 0 Store Ampl./Phases Set Cent. of Laue Circle h 0.00 Eq. Tilt [mrad] & angle * 0.00 Type of Absorption N Autgmatism [Å] / Coma Mag. * Two fold 0 0 0 Three fold 0 0 Coma 1000 0 Mechanical Vibration [Å] Sigma of (a,b) 0.00 Angle with x-axis [*] Exit Wavefunction Imm	Thick. (beg.inc.end) 30 0 Store Ampl./Phases Set Cent. of Law Circle h 0.00 Eq. Tilt [mrad] & angle 0.00 Type of Absorption Non Antigratism (Å] / Cema Assg. Wag. Two fold 0 0.0 Three fold 0 0.0 Coma 1000 0.0 Mechanical Vibration (Å] Sigma of (a,b) 0.00 0.0 Angle with x-axis (°) 0.0 Exit Wavefunction Image	Thick. (beg,linc,end) 30 0 Sto Store Ampl./Phases Sen h Cent. of Law Circle h 0.00 0 Eq. Tilt [mrad] & angle * 0.00 2 Type of Absorption Non 1 Antigratism (Å] / Cema Asg w/h Two fold 0 0.0 Three fold 0 0.0 Coma 1000 0.0 Mechanical Vibration (Å) Sigma of (a,b) 0.00 Angle with x-axis (*) 0.0 Exit Wavefunction Image Exit Wavefunction Image	Thick. (beg.inc.end) 30 0 Store am Store Ampl./Phases Set h k Cent. of Law Circle h 0.00 0 0 Eq. Tilt [mrad] & angle * 0.00 2 0 Type of Absorption Non 1 0 Adignatism (Å] / Cema Asg Wag. W/h Two fold 0 0.0 Three fold 0 0.0 Coma 1000 0.0 Mechanical Vibration (Å) Sigma of (a,b) 0.00 0.00 Angle with x-axis (*) 0.0	Mag. Mag. With Three fold 0 0 0 Antigmatism [A] / Coma Assignatism [A] / Coma Assignatism [A] / Coma Assignatism [A] / Coma Mag. With 0 0 0 Two fold 0 0 0 0 Three fold 0 0.00 0.00 Ca Mag. With 0.00 0.00 Ca Three fold 0 0.00 0.00 Ca Magle with x-axis (") 0.00 0.00 0.00 Ca Exit Wavefunction Image Exit Wavefunction Image	Thick. (beg.inc.end) 30 0 Store amplitudes and p Store Ampl./Phases See h k i h Cent. of Laus Circle h 0.00 2 D -2 Type of Absorption Non 1 D -1 Astignatism (Å) / Coma Astignatism (Å) / Coma A	Thick. (beguinc,end) 30 0 Store amplitudes and phases Store Ampl./Phases Set h k i h k Cent. of Laus Circle h 0.00 0 0 0 0 0 Eq. Tilt (mrad) & angle * 0.00 2 0 -2 -2 -2 Type of Absorption Non 1 0 -1 -1 -1 Astignatism (Å) / Coma Asg W/h Ch Ch Cancel Ch Two fold 0 0.0 0.0 0.0 Ch Cancel Ch Two fold 0 0.0 0.0 0.0 Ch Ch Cancel Ch Two fold 0 0.0 0.0 0.0 Ch Ch Cancel Ch Ch Two fold 0 0.00 0.0 0.0 Ch Ch

function of specimen thickness. The reflections to be tracked

are determined by entering the hkl values for the reflection. Only 10 reflections can be tracked this way.

Center of the Laue Circle

Specimen tilt is specified by entering the center of the Laue circle in units of the h and k indices of the projected two-dimensional reciprocal-space unit cell. The new indices and their relationship to the original reciprocal cell is found in the data file <**structurename**>.**p_prnt**

Type of Absorption

Absorption can be included in the program by introducing an

MacTempas User Manual

Simulation	Parameters
Crystal Parameters A (Å) 3.8140 Alpha (deg.) 90.00 8 (Å) 3.8140 Beta (deg.) 90.00 C (Å) 30.5200 Gamma (deg.) 90.00 Spacegroup # (Int. Tables) 139 # of Atoms in Basis 9 # of Symm. Ops. 32 Show \$ bios	Specimen Parameters Zene-auts (acout)) 3-Index 0 1 0 0 1 0 Number of Sices per cell 1 1 0 0 1 0 Number of Sices per cell 1 2.00 1 0 30 30 Thick. (beg, inc, end) 30 0 30 30 30 30 Store Ampl./Phases Set No 0.00 1 0.00 1
# of atoms in UCell 3.8 Show	Eq. Tilt [mrad] & angle * 0.00 0.0 Type of Absorption None Absorption
Microscope and Lens Parameters Microscope Name CM300 Voltage (kV) 300 Cs [mm] 0.60 Convergence angle [mrad] 0.20 Spread of defocus [Å] 30 Defocus (beg,inc,end) [Å] -2400 -100 Obj. Iens apert. rad. [Å-1]: 1.25	Attis Absorption Parameters No absorption Two 1 Two 1 Two 1 Three Coma Other Parameters No absorption Two 2 Coma Other Parameters No absorption Other Parameters Other Parameters Net
h k Mag. An Cent. of Obj. Lens Aprt. 0.00	Ingle* Sign Cancel OK
Mark for re-calculation Projected Potentia Mark as calculated Projected Potentia	al Exit Wavefunction Image al Exit Wavefunction Image (Cancel)

imaginary projected potential.

Microscope

The type of electron microscope used to generate the imaging parameters. Predefined microscopes are shown in the popup menu together with one undefined microscope. If a predefined microscope is used, MacTempas provides values for Cs, the spherical aberration coefficient of the objective lens (in mm.); DEL, the halfwidth of a Gaussian spread of focus due to chromatic aberration (in Ångstrøm units); TH., the semi-angle of incident beam convergence (in milliradian). If the type of microscope is unknown to MacTempas, the above values must be entered separately (We will see later how a new microscope

Cryst	al Paramete	ers			1.1	Specin	nen Paran	neters .		1.201
A SĂI	3.8140	Alpha b	den 1	90.00	1 3	Zarie-an	ds. Juree (t).			-index
# CÅ1	3.8140	Reta Ide	a.1	90.00			1 1	0 0		V 1 0
CIA:	30.5200	Gamma	Idea	95.00		Numbe	r of Slices	bet cel	1	1
Casro	arrain # diet	Tabler)	(asge)	20100	1	Cmax [[A-1]	111999	-	2.00
A mi A	group e tra			29	1	TRICK.	(beg,exc,e	nd) 30	0	1.50
1014	coms in eas	0 3		HOW.		Store A	impl./Phas	es (Set	_) No
# 01 5	ymm. Ops.		4000EX	500	12	Cent. a	I Laue Ci	icle t	0.00	A 0.0
# of a	toms in UCe	9	ABT	- 12		Eq. Tilt	[mrad] &	angle *	0.00	0.0
# of d	ifferent ator	mi	ARM			Type of	f Absorpta	on [Norie
			200CX						2	
Micro	scope and	Lens	2000FX	_			Antiomati	um (A) 2	Согла	decent at
Micro	scope Name		CM300	-			00005000		100004	Argin
Voltag	pe (kVI 3	00	Unknown	0.	60			Mag		ei hariz.
Corive	ergence angl	e [mrad]	0.20				Two fold	0		80
Sprea	s of defocus	(Å)	30				Three fold	0		0.0
Defoc	us ibeg, inc.	end) [Å]	-2400	-100	-27	00	Coma	1000		0.0
Obj. F	ens apert, ra	Id. [A-1]	1.25				Wechanic	il Vibrati	on IAI .	
			h k	Mag.	Angle*		flamad	in hi	0.00	0.00
Cent.	of Obj. Lens	Apit.	0.00 0.00	0.00	0.0		signa or	(4,1)	0.00	0.00
Cent.	of the Optic	Axis	0.00 0.00	0.00	0.0		Angre w	он м-ак	(e.f.)	0.0
Over	-ride calcul	ation sta	tus							
Mark	for re-calcul	lation [Projecta	i Paten	sial .	Exi	it Wavefun	ction	🖸 în	iage
	an calculates	4 5	Projecter	d Poten	tial	Ex	t Wavefun	ction		age

may be made known to MacTempas).

Constitution Processing

Voltage

The electron microscope accelerating voltage in kilovolts.

Objective Lens Defocus

The defocus of the objective lens is entered in Ångstrøm units with a negative value representing underfocus (weakening of the lens current). As for the speciment thickness parameter, the input is a range specified by the upper and lower bounds and an increment.

Cs, Spherical Aberration

The spherical aberration of the objective lens in mm.

Convergence Angle

This is the spread in angle for the cone of incoming electrons depending on the condenser lens aperture. The angle is given in mrad.

Spread of Defocus

This is the effective spread in defocus which results from the distribution of energies of the imaging electrons and the chromatic aberration of the objective lens. The unit is Å.

Aperture Radius

The radius of the objective aperture is specified in $Å^{-1}$

Center of objective Aperture

The center of the objective lens aperture is defined in units of h and k of the two dimensional reciprocal space unit cell, as for the Laue circle center.

Center of the Optic Axis

The center of the optic axis of the electron microscope is specified in terms of the h and k indices of the two-dimensional reciprocal-space unit cell, just as for the Laue circle center and the aperture center.

Two-fold astigmatism

The two fold astigmatism of the objective lens and the angle with the x-axis. The magnitude is given in Å.

Three-fold astigmatism

The two fold astigmatism of the objective lens and the angle with the x-axis. The magnitude is given in Å.

Coma

The coma of the objective lens and the angle with the x-axis. The magnitude is given in Å.

Mechanical Vibration

This simulates the effect of a slight vibration of the microscope. One finds that often the simulated images show details that are not present in the experimental data regardless of other imaging conditions. This may be due to image degradation caused by microscope vibration or other effects not included and thus one can introduce a slight mechanical vibration in an attempt to create more realistic simulated images. It is possible to specify an anisotropic vibration by introducing the amplitude in two perpendicular directions with the diagonal of the ellipse at an angle with the a axis (as in the unit cell viewed in the zone axis orientation). **MacTempas User Manual**

Ch. 4 Running MacTempas - p.42

Chapter 5

Windows

This chapter explains the windows of Mactempas, the information presented in each and how one interacts with the contents of the windows.

Status	

IDLE

Status Window This window shows the current status of the program indicating the number of phasegrating coefficients calculated, the current slice number being calculated, the current image being calculated etc.



Atom Window This window shows which atoms are present in the structure, the color the atom will be drawn in (if colored atoms are set) and the relative sizes of the atoms to be drawn. To change the color of an atom, choose the Color Picker tool from the Tools Window, click on a color in the Color Window and deposit that color on an atom by clicking on the colored circle representing the atom. The color of the atom will be set to the new color.

Ch. 5 Windows - p.43

To change the atomic radius, double-click on the chemical sym-



bol. A dialog window will pop up and a new value for the atomic radius can be entered (units in Å).

Image Control-Window



This window is used primarily to control the appearance of images. The black and white values of the current selected image is shown and can be changed by typing in new values. The contrast and brightness can be changed by using the appropriate sliders. An image can be shown on a logarithmic scale which is the default for images in frquency space (reciprocal space). The line in the graph represents how input image values are mapped to output display values. Thus an image can be pseudo colored by choosing a color from the color bar with the color picker tool selected and "depositing" this color in the vertical gray scale bar showing the display vlaues. The histogram of the current image is shown and black and white values can be chosen by clicking and dragging to select a region of the histogram. To invert the display, click in the "Invert" button. Similarily the image is reset to the original values through the "Reset" button. This window is also used to set the color of a particular atom species and the color of lines and text. To choose a color, the Color Picker Tool must have been chosen.

Ch. 5 Windows - p.44

Tools Window

The following tools are currently defined:

Pointer

Used for general moving around objects in the display window. If an object is selected and the "Option" key is held down while dragging an object, a copy is made of the object.

Text Tool

Clicking on this tool turns the cursor into an i-beam cursor which can be used to select an insertion point for text. To set the insertion point for text to be typed in the image window click the mouse at the desired point. The Font, Size and style of the text is determined from the menu bar. The text will be drawn in the current foreground color and can be left, canter or right justified.

Magnifying Glass

When selected the cursor turns into a magnifying glass which can be used to zoom in on a selected part of the display. Each time the mouse is clicked in the image window, the image is zoomed by a factor of two. By holding down the Option key while clicking, the image will be zoomed out by a factor of 1/2 for every click. Double-clicking the magnifying glass returns the image to normal. Currently no other tools work in zoomed mode.

Line Tool

This tool is used to draw lines on the display. If the Shift key is down, only vertical or horizontal lines will be drawn.

Selection Tool

This tool is used to select a portion of the screen for several possible operations such as copying, cutting, histogram computation etc. To select an area, click at a point in the display and



drag the cursor while the mouse button is pressed.

Trace Tool

This tool is used to get a line trace for the line drawn with the Trace Tool being the current tool.

Color Picker Tool

This tool when selected, allows the user to pick a color from the Color Window and color atoms, selecting fore-/back-ground colors and pseudo-color atoms. The selection of color is described under Color Window above

Hand Tool

Use this tool to move images around in the image window.

Ruler Tool

Use this tool to measure distances in an image. An image can be calibrated from the menu command under Process after a line is drawn using the ruler tool..

Rotate Tool

This tool is used to rotate drawings of crystal structures. In order for it to be active, a structure must have been selected first.

Masking Tools

The last 5 tools are masking tool normally used in reciprocal space, but they can be used in real space as well. The masks are a) **Spot mask**. A reflection and its conjugate is selected.

b) Lattice mask. A mask defined by two lattice vectors.

c) **Band Pass mask**. This mask is defined by an inner and an outer circle.

d) Wedge mask. Defined by two lines.

e) **Line mask**. Defined by a line and a single lattice vector. All these masks can be transparent or opaque, meaning they work on the region within or outside of the mask. The mask parameters can be edited by double clicking on the mask or selecting the mask and choosing "Edit Mask" from the "Process" menu. The number of lattice spacings for the vector(s) for the lattice mask and line mask can also be changed by clicking in the end point of the vector with the "Option" key down. Each click increments the number of lattice spacings to the endpoint by one. Holding down the "Shift" key and the "Option" key decreases the number of lattice spacings by one.

Info Window



Display Window



This window shows the current position of the cursor within the image window and the intensity of the underlying pixel. When dragging a rectangle, the dimensions of the rectangle are shown. Line lengths and angles are also displayed. Image statistics is displayed in this window when invoked through the "Statistics" in the "Process" menu.

Use this window to define which part of the calculation to display. The choices are:

Projected Potential - Essentially the output of the projected potential routine. There is a one to one correspondence between the points in the projected potential and those in the image if displayed under equivalent conditions.

Exit Wavefunction - This is the output of the multislice component of the program and shows the distribution of electrons as they emerge from the bottom of the specimen, or at a predefined depth in the specimen. By holding down the **Option** key when selecting the button, one can select to display either the magni-

Ch. 5 Windows - p.47



tude squared (default), the complex amplitude or the complex

phase of the electron wavefunction at the exit surface of the specimen.

Diffraction Pattern - Select this option to display the diffraction pattern for one of the selected specimen thicknesses. This is a dynamical diffraction pattern including multiple scattering in the specimen.

Image - When selected, one of the calculated images becomes the source of the operations defined by clicking in the Operand Window. By holding down the **Option** key when selecting the button, one can select to display either the image intensity



(magnitude squared, default), or if the image plane wavefunction(s) has been calculated, the complex amplitude or the complex phase of the electron wavefunction at the image.

FFT

Use this to perform a Fourier Transform on the selected source. Operating on the Projected Potential will yield the structure factors, operating on the Exit Wavefield will yield the diffraction pattern and operating on the image will give the Power spectrum of the image.

#Unit cells

Use this to specify the number of unit cells that should be displayed. The input determines the number of cells in the a-direction and b-direction.

Zoom

Use this selection to Zoom the object to either magnify the object or to reduce the object. A zoom factor greater than 1. magnifies and a zoom factor less than 1. reduces the object.

Display

Before the result of operating on a selected source is displayed in the image window, **Display** must have been clicked. Choosing the source and operations only selects the functions to be performed. When **Display** gets activated, the functions get executed.

->File

This will allow for output of the numeric values of images, amplitudes and phases to a file. Options allow for writing the data in ascii format or binary format. Images can also be written

Sare As.	untitled	
Format:	✓ Binary	-
CrystalKitX	Grayscale TIFF Floating Point TIFF MAL Input File ASCII	•
		• ••

as TIFF files in this fashion.

Cancel:

Use this button in case the wrong sequence of commands was chosen or anything else was entered wrong. This cancels the set functions. Chapter 6

Menus

Many of the functions in MacTempas are run from one of the MacTempas menus, including the multislice calculation. In addition, most options are set from one of the menus. This is a list of the currently available menus and a description of their function.

File	Edit	Options	Con	nmands	
Nev	w Norn	nal Structu	re	ЖN	
Nev	w Laye	red Structu	ire	☆%N	
Op	en Stru	icture File.	••	жо	
Clo	se			96 W	
Sav	e Struc	cture		ЖS	
Sav	e Strue	cture As		ፚ ሄ ያ	
Op	en Ima			公米O	
Sav	e Imag	je		~ %S	
Sav	e Imag	je As	14	之心粥S	
Imp	oort Pie	ct File			
Sav	e Winc	low			
Pag	je Setu	p		۵жР	
Prir	nt			ЖP	

This menu contains the following commands:

File Menu

Ch. 6 Menus - p.51

New Normal Structure...

Create a new structure file. A name is prompted for before input is made. Enter a unique structure name, the program will append the extension .at. Make sure that you do not add an extension of the type .at in which case MacTempas will not properly deal with the file later on. Also make sure the filename does not have a period in it. This opens a new structure with

	uctures 📑 💽 —
CrystałKOX 2014 Carbon Ubrary 2 Example Structures	95 bcscb.at copper.at Imp.at
	()++
New Folder	Add to Favorites

default values for all the parameters. Change the input to reflect

Crystal Parameters A (K) 4.0000 Alpite (Breg.) 30.01 B (K) 4.0000 Beta (Breg.) 30.01 C (K) 4.0000 Beta (Breg.) 30.01 C (K) 4.0000 Beta (Breg.) 30.01 Semanne (Breg.) 30.01 52.02 # of Atoms in Race 1 Mem. # of Seman. Ops. 152 Mem. # of Seman. Ops. 152 Mem. # of Seman stem. 1 Telse.)	Specifierer Nariameters P-Holes Date sets (seed) 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 1 1 Number of States per cell 1 1 1 2.00 1
Microscope and Lans Parameters Microscope Raine 40000 Vehape (Kr) 400 Co (mm) Los Generating (micail) 057 Peyrot Re Seried of debots (A) 50 0 0 - 0 0 - 0 0 - 0 0 - 0 0 - 0 0 - 0 0 - 0 0 - 0 0 -	Type of Absorption Hurs ac Integration (A) / Cana Integration (A) / Cana ac Mag. Integration (A) / Cana ac Teo hold T 0.0 Certer Three hold T 0.0 Certer Teo hold T 0.0 Certer Teo hold T 0.0
Story central learn 6 8 May Aug Cent. of Oil Least Aut. 8 00 000 000 000 Cent. of the Opric Aug	Signa of is to 0.00 0.00 Angle with a serie [] 0.0
Over-ride calculation status Mark for re-calculation 🔛 Projected Potential Mark as calculated 🔄 Projected Potential	Carl Novelaction Charge

the structure that you want to create

New Layered Structure...

Create a new layered simulation file. A name is prompted for before input is made. Enter a unique structure name, the program will append the extension .lay. A layered structure is characterized by being made up of a sequence of pre-calculated projected potantials. Thus a layered simulation file does not contain atomic positions. The structure information in the input dialog is replaced by .

A [Å]	4.0000	Al	pha (deg.)	90.00
[Å] 8	4.0000	Be	ta [deg.]	90.00
CIAI	Not Applic.	G	imma (deg.)	90.00
# of	phase grating	95	Stacking se	quence

Only A and B and Gamma have meaning for a layered "structure". The buttons "Define PGratings" and "Define Stacking" are used to choose the different projected potentials and to define their sequence to make up the entire specimen.

Open Structure File...

Open an existing structure or a leyered file. The standard Macintosh file open dialog is presented and only files of the type 'TEXT' with the extension ".at" or "lay" are displayed as selectable. The name of the display window will change to reflect the name of the current structure.

Close

Close the file, image or window currently selected

Save Structure

Save the current data for the structure file in use. The current data will be written to the file, overwriting any old data.

Save Structure As...

Save the current structural information. Do not use a name with

an extension if the file being saved is a structure file for later use by MacTempas.

Open Image...

Open an image. Supported images are currently tiff files and binary files. RGB tiff files and compressed tiff files are not supported. Binary files can be of integer or float types with different length and byte order.

File	Format	
angth: 2097152	1 Official	
Data Type Roal = Bytes	Get Size By Country	
Size	@ Linury	
Nichtiv 734 Avrigate	Officiat: 0	
eigte: 724 90	🔄 Swap Data Bytes	
Simulation Sampling	1	
Width: 0 Height: 0	Angle: 90.00	

Save Image...

Saves the content of the image window into a file. MAL input

Same As:	45948.17		
Format Where CrystalCox	✓ Binary Grayscale T Floating Poi MAL Input F ASCU	IFF nt TIFF Tile	•
Died Carbo	Aben	an observe	_
Courtple St	uctures P	ing ut SF rescue	
e			
0	Www.Polder) (Add to Favorities	3

files are binary files used by the mal or TrueImage program for exit wave reconstruction from a through focal series.



Save Image As...

Similar to "Save Image".

Import PICT File...

Import a PICT file and display it in the MacTempas image window.

Save Window...

Saves the content of the image window as a PICT file..

Page Setup...

Set the options for the page to be printed.

Print...

Print the front window..

Edit Menu

Undo

Ch. 6 Menus - p.55

Edit	Options	Comma
Undo	6	ЖZ
Redo		0%Z
Cut		жx
Сору	6	ЖC
Paste	(36V
Clear	è	
Selec	t All	ЖA
Obje	ct Info	
Arran	ige Objec	t 🕨

Undo / Redo the last operation. These operations do not cur-

rently work in MacTempas.

Cut

Cut the selected Object or the Selection made by the selection tool.

Copy

Copy the selection or the selected object.

Paste

Paste the content of the paste buffer into the display window. The source for the paste can be an image cut out from another application or through the cut/copy commands of MacTempas. If the object is an image, the image will be pasted into the display window if it is currently selected or into a separate image window if not.

Clear

Clears the selection made by the selection tool

Select All

Select all objects in the display window or an entire image.

Object Info

Shows the clipboard and the content of the clipboard.

Arrange Object

Shows the clipboard and the content of the clipboard.



Ch. 6 Menus - p.57

Options Menu

Options	Commands	Parameters
Live mi	croscope Cont	rol
✓ Automa	tic Erase	ЖE
Atom O	verlay	ЖL
Montag	e	☆೫M
Intensit	y Scaling	
Magnifi	cation	
CTF Sca	ding	
Diffr. pa	atterns	ЖD
Min. Le	ns Intensity	
Slice Me	ethod	
Show M	icroscopes	
✓ Use Ele	ctron Paramet	er Fit
Edit Sca	tt. Fact. Para	meters
Treat as	s monolayer	

Live Microscope Control...

When a calculated image is selected, this command can be invoked to bring up an interactive window for changing the calculation parameters for this image. Changes in the parameters

	— Spe	cime	n Thickn	ess —	-	
Specimen Thi	ckness (/	Ingst	rom]	200.00	2	
	3	Speci	men Tilt	-		
			0	Theta (m	rac	0.00
::=:: <	100		0	Phi [Deg.	1	0.00
		-	hD	0.00	k0	0.00
Diff.Patt.	Geome	try	_			
Distance Plate In	1 0.00	Objec	tive Lens			
Defocus (Å)	-200	-	1	S		۱ı –
Colored (A)	0.600	-			× 1	(()))
C Cs (mm)	0.600	-		1	11	ψr.
Div (mrad)	0.200				~	
Dal [A]	30.0		imic	16)4 3	- ini	CTF) 4
Abberrations I	Mag (Å)	A	ngle (*)	Vibratio	on (A)	
77	0	0	D	🔾 Sign	na-a	0.00
Two fold	1. A . A			Constant in		0.00
Two fold	a	0	D	O Sign	na-b	0.00

are reflected live aslong as the calculation time is reasonable..

Automatic Erase

Toggles whether you are prompted for the position of the upper left corner of the image to be displayed.

Atom Overlay

If set, the atom positions will be drawn in as circles on top of images. The circles are scaled to the atomic radius and the color is the color set for that atom species. If the Option key is held down while the image is "drawn", only the circles are drawn (no image).

Montage...

Brings up a dialog box, allowing the user to select automatic montage of a series of images, the position of the series of

Automatically montage image	es	
Starting Location of the	0	0
Horizontal Separation of Im	ages	0
Vertical Separation of Imag	es	0
🗹 Auto-scale when images	do not fit	
Layout options		
Defocus Horizontal/ Thickne	ess Vertical	l.
O Thickness Horizontal/ Defo	us Vertica	l.
O User Defined Layout (Click to	o Display a	nd Set)

images and the number of pixels to leave between images.

Intensity Scaling...

Brings up a dialog box, allowing the user to manually set the intensity values to be mapped to black and white. The values shown correspond to the last image displayed with automatic scaling.



Magnification

Allows the user to set the magnification to a set value. The magnification depends on a screen with a resolution of 72 dots/

	-	-
Magnification	30.0	Million
🗹 Auto-scale when	images d	o not fit

inch. If Auto-scaling is set, images will scale to fit the window.

CTF Scaling...

Brings up a dialog box, allowing the user to set the maximum scale of the reciprocal axis during plotting of the Contrast Transfer Function.



Diffraction Pattern...

Displays a dialog box, allowing the user to select the position of the diffraction pattern, the camera length and the minimum diffracted intensity that can be displayed. The user can also choose whether the objective lens aperture should be superimposed on the diffraction pattern. The indices of the diffracted beams can be superimposed on the diffraction pattern as well as the corresponding real space distances. Selecting Circular Diffraction spots instead of Gaussian Diffraction Spots results in solid circles. One can also set a cut-off such that diffracted beams with

Options	Settings	
🗹 Display Objective Jens Aperture	Camera Length (mm) :	520
Display Laux Circle	Divergence Angle (mrad) :	0.200
Show hkl	Min. Intensity to display 10-	4.5
3D indexing (h,k,0)	Display only g <= 2.00	1
Show d-spacings		
In real units [Å]		
Caussian Diffraction Spots		-

g-vectors larger than the cut-off will not be displayed.

Min Lens Intensity...

Displays a dialog box, allowing the user to manually set the minimum intensity required of a diffracted beam for inclusion in the formation of the image.

be conside	red		_
in the Obje	ctive Len	s: 10**-	8.0
			-
		~ ~	
	Cance		ж

Slice Method...

Allows the user to select the option to perform a three dimensional calculation of the projected potential by summing over the third dimension (1) in reciprocal space.



Show Microscopes...

Displays a dialog, showing the user which microscopes are known to MacTempas. The default parameters associated with a known microscope can be changed by the user and a new microscope may be made known to MacTempas. MacTempas currently only allows a maximum of 10 microscopes to be made known

	meroscop						
iame	Volt.[kV]	Cs[mm]	Div[mrad]	Del[Å]			
AGOOEX BL AGN	400 200 800	1.00 0.30 2.00	0.55 0.50 0.85	80 50 150	Microscope Pa	rameters	
000CX	200 200	1.20	0.65	100 80	Microscope Parameters		
M300	308	0.60	0.15	20	Microscope Name	Unti	tled
New) (De	lete) (Edit)	Accelerating Voltage (kV)		200
					Spherical Abberation Const	ant (mm)	1.2
			Canc	el) 🧲	Spread of Defocus (Å)		50
					Divergence Angle (half-wid	th) (mrad)	.5

Use Fit For Electron Scattering Factors / Use Fit For X-Ray Scattering Factors

MacTempas can use either the 8 parameter fit for the Electron Scattering Factors or the 9 parameter fit for the X-Ray Scattering factors. The menu item text will reflect the current setting.

Edit Scattering Factor Parameters...

Brings up a table of the fitting parameters. Double clicking in the value -field brings up a dialog box prompting for a new value. See next page.

Treat as Monolayer

When this option is set, the calculation treats the unit cell as a non-repeating structure such that the entire specimen is represented by a single unit cell with the thickness of the specimen as the thickness of the unit cell.

_	-		an unicour	TICKOT LA	eco de so	SCORE 19		<u> </u>		-
-	Z	Al	DI.	A2	02	A3	53	A4	D4	1
H	1	0.2022	30.8679	0.2437	8.5444	0.0825	1.2726	0.0000	0.0000	
He	2	0.0906	18.1834	0.1814	6.2109	0.1095	1.8026	0.0562	0.2844	
Li)	5	1.6108	107.6384	-			45551	0 0986	0.4951	
8e	4	1,2498	60.0042	IDES	83977	392	5.0534	0.1055	0,4157	
8	5	0.9446	46.4438		Gancel	OE	5.2220	0.1159	0.3767	1
C .	6	0.7507	36.9951	1.1.1.1	0.040	10.499.9	2.01.39	0.1247	0.5456	
N	7	0.5717	28.8465	1.0425	9.0542	0.4647	2.4218	0.1511	0.3167	
Q	8	0.4548	28,7803	0.9175	7.6220	0.4719	2.1440	0.1584	0.2959	
F	9	0.3696	20.2390	0.8109	6.6093	0.4751	1.9510	0.1459	0.2793	
Ne	10	0.3025	17.6396	0.7202	5.8604	0.4751	1.7623	0.1584	0.2656	
Na	11	2.2406	108.0039	1.3326	24.5047	0.9070	3.3914	0.2863	0.4346	
Mg	12	2.2682	73.6704	1.8025	20.1749	0.8394	3.0181	0.2892	0.4046	
A1	13	2,2756	72.3220	2.4280	19.7729	0.8578	3.0799	0.3166	0.4076	
St	14	2.1293	57.7748	2.5333	16.4756	0.8349	2.8796	0.3216	0.3860	
P	15	1.8882	44.8756	2.4685	13.5383	0.8046	2.6424	0.3204	0.3608	
S	16	1.6591	36.6500	2.3863	11.4881	0.7899	2.4686	0.3208	0.3403	
C1	17	1.4524	30.9352	2.2928	9.9795	0.7874	2.5536	0.3217	0.3228	
Ar	18	1.2736	26.6823	2.1894	0.0130	0.7927	22105	0.3225	0.3071	
К	19	3.9507	137.0748	2.5452	22.4017	1.9795	45319	0.4617	0.4340	
Ca	20	4.4696	99.5228	2.9708	22.6955	1.9696	4.1954	0.4518	0.4165	
Sc	21	3.9659	88.9597	2.9169	20.6061	1.9254	3.8557	0.4802	0.3988	
TI	72	3.5653	81.9821	2.8181	19.0486	1.8930	3.5904	0.4825	0.3855	
V.	23	3,2449	76.3789	2.6978	117.7262	1.8597	3.3632	0.4864	0.3745	
Cr	24	2.3066	78.4051	2.3339	15.7851	1.8226	3.1566	0.4901	0.3636	
Min	25	2.7467	67.7862	2.4556	15.6743	1.7923	2.9998	0.4984	0.3569	
Fe	26	25440	64.4244	2.3434	14.8806	1.7588	2.8539	0.5062	0.3502	
Ca	27	2.3668	61.4506	2.2361	141798	1.7243	2.7247	0.51 48	0.3442	
N	28	2.2104	58.7267	2.1342	13.5530	1.6891	2.6094	0.5258	0.3388	
Cu	29	1.5792	62.9403	1.8197	12.4527	1.6576	2.50.42	0.5323	0.3331	
Zn	30	1.9418	54.1621	1.9501	12.5177	1.6192	2.4164	0.5434	0.3295	
Ga	31	2.5205	65.6019	2.4855	15.4577	1.6879	2.5806	0.5992	0.3510	
Ge	5.2	2.4467	55.6930	2.7015	14.5950	1.6157	2.4461	0.6009	0.3415	
As	33	2.3989	45 71 79	2.7898	12.8166	1.5265	22799	0.5956	0.3277	
Se	34	2.2980	38,8296	2.6541	11.5359	1.4555	2.1463	0.5695	0.3165	
Br	35	Z.1659	33.0987	2.9037	10.4996	1.3951	2.0415	0.5506	0.3070	
Kr	36	2.0558	29.9992	2.9271	9.5977	1.8425	1.9520	0.5888	0.2986	
Rh	37	4 7760	140 7921	1 8599	0199.01	0 1770	13 2010	0.8597	0.4194	

Ch. 6 Menus - p.65



Commands Menu

Erase

Erases the selection made by the selection tool.

Draw Atomic Model...

Displays a dialog box, from which the user can select to display the original or transformed unit cell from any direction, including perspective view. The transformed cell corresponds to the unit cell that MacTempas uses in the multislice calculation. To view the cell as "seen" by the electrons, the transformed (new) unit cell should be viewed in the 001 orientation. It should be noted that the viewing direction is in units of the real space unit cell axes. One can also view a cross-section of the material in a given direction. A dialog box allows the user to specify the field of view in Å for the two directions.

Type of View			
Original Unit Cell			
Transformed Unit Cell (001	is Elec	tron Bear	n Direction)
C Extended View Width	20	Height	20
View Options	_		
Viewing Direction (UVW) :	0	0	1
Initial Scaling Factor to be used Displaying The Unit Cells Di of	l for Windo	wt	60
1. A.S.			

Draw the CTF

Draws the Contrast Transfer Function for the current microscope values. The original microscope values are taken from the structure data, but the user is free to change the values associated with the CTF independent of the values used in calculating the image. Clicking in the CTF will show a bar with the values



of the CTF and the resolution. The bar moves with the mouse.

Draw Pendelløssung Plots...

In case the user has selected to store a set of diffracted beams for plotting of amplitudes and phases as a function of specimen thickness, this brings up a dialog box allowing the user to set the plotting conditions. One can choose to have the amplitudes

MacTempas User Manual

or the intensities plotted as well as the phases of the diffracted beams. Each reflection can be plotted by itself, or several reflections can be superimposed on the same plot. Instead of plotting the values, the values can also be written to a file for further manipulation or inspection.



Define Projected Potentials...

This allows the user to specify which potentials to be used in the layered structure.

Stack Potentials...

This allows the user to specify the sequence of potentials that should be used in the multislice calculation. This applies only to layered structures. See Chapter 9 for a more detailed instruc-

	- IKAN AND	Defined	Phas	egratings	-	0.80		87.10°	100	- 00/11/11/24	
*	Name	Δz	#	Name	Δz	#	Name	Δz	#	Name	Δz
1	c60slc_0	3.00									
3	c60sic_2	3.00									
4	(6051C_3	3.00									
		Specir	nen							1	
	TOP	1,2,	3, 4, 1,	2,3,4						De	posit
										(In	sen
										Re	peat
										R	eset
-	24.0										
										C	OK.

tion on how to create a layered structure.

Slice Unit Cell...

Use this option to subdivide a structure into separate layers for use in a layered structure calculation. The direction perpendicular to the slices and the number of slices must be specified.

Anna anis Karabatan	10		.w
zone axis for slicing		10	-
Number of separate la	ivers		1
Calculate Each Pro	ected	Potential	
Carculate cach Proj	Jectieu	rotencial	

Parameters Menu

N	Jain Paramet	ers	
	Parameters	Calculate	T
	Main Param	eters	
	Atom Basis		
	Symmetry (Operators	
	Atom Coord	dinates	

This brings up a dialog box showing the current conditions for the simulation. The values are taken from the input given to the

Crystal Parameters	Spec	cimen Paran	neters	2.10	dex	
[A] 3.8140 Alpha [deg.]	30.00	0 1	0 0	0	1 0	
(Å) 3.8140 Beta (deg.)	30.00 Num	ther of Gires	nercell	9	1	
[A] 30.5200 Gamma [deg.]	30.00 Cma	Gmax IA-11				
pacegroup # (Int. Tables) 13	35 Thie	Thick (beg.inc.end) 30 0				
of Atoms in Basis 9	tow Ston	Store Ampl /Phases				
of Symm. Ops. 32	now Cent	Cent of Lave Circle h 0.00 + 0.00				
of atoms in UCell 38	now En 1	En Tilt [mrad] & angle * 0.00			0.00	
of different atoms 5	Elig. 1	The set the section 0.00 0.0				
	Туре	of Absorpti	on _	Nor	e.	
oltage [kV] 300 Cs [mm onvergence angle [mrad] 0.20 pread of defocus [Å] 30	1 0.eo	Two fold Three fold	Mag. 0 1 0	w/h	oriz.)	
Defocus (beg,inc,end) [Å] -2400	-100 -2700	Coma	1000	0.0)	
bj. lens apert. rad. [A-1]: 1.25		Mechanic	al Vibratio	n (A)		
h k Cent. of Obj. Lens Aprt. 0.00 0.00 Cent. of the Optic Axis 0.00 0.00	Mag. Angle" 0.00 0.0 0.00 0.0	Sigma of Angle w	(a,b) th x-axi	0.00 0 s [°] 0	.00	
Over-ride calculation status						
lark for re-calculation 🗌 Projected	Potential 📃	Exit Wavefur	ction		e	
lark as calculated 📃 Projected	I Potential 📃	Exit Wavefur	ction	Imag	e	

New... command in the FILE menu. The parameters can be changed as to bring about a new simulation.

Ch. 6 Menus - p.71

Atomic Basis...

Brings up the list of all the atoms forming the set of basis atoms for the current structure. The atomic coordinates etc. can be edited and atoms can be added to or deleted from the list.

۴.	Name	x-coord.	y-coord.	z-coord.	dw-fact.	Occ.
t I	Ca	0.000000	0.000000	0.000000	3.600000	1.000000
2	\$F	0.0000000	0.000000	0.109700	3.600000	1.000000
i I	Di	D.DODDOD	0.000000	0.302200	3.600000	0.670000
	84	0.000000	0.000000	0.268100	3.600000	0.130000
	CU	0.000000	0.000000	0.445600	3.600000	1.000000
1	0	D.SODDOD	0.000000	0.446000	5.400000	1.000000
ć	0	0.000000	0.000000	0.375000	5.600000	1.000000
	0	0.000000	0.000000	0.205000	3.600000	1.000000
1	٥	0.500000	0.000000	0.250000	3,600000	0.065000
	_					
	-		-			-

Symmetry Operators...

This brings up the list of symmetry operators either associated by the space group or entered manually by the user. The symmetry operators can be edited, and new ones may be added to
	8.8.2	16	-y+1/2,-x+1/2,-z+1/2
1	x+1/2,x+1/2,z+1/2	17	-X,-Y,-Z
	-3,-9,2	18	-x+1/2,-y+1/2,-z+1/2
÷	-x+1/2,-y+1/2,z+1/2	19	X,Y,-Z
5	-y, x, #	20	x+1/2,y+1/2,-x+1/2
5	-y+1/2,x+1/2,x+1/2	21	γ ₄ -× ₄ -2
,	γx.2	22	γ+1/2,-x+1/2,-x+1/2
8	y+1/2,-x+1/2,2+1/2	23	-'y', X, -2
•	-X,Y,-3	24	-y+1/2,x+1/2,-2+1/2
11	-x+1/2,y+1/2,-z+1/2	25	x,-y,2
10	X,-Y,-2	26	x+1/2,-y+1/2,z+1/2
12	x+1/2,-y+1/2,-z+1/2	27	-3,9,2
13	¥,K,-2	28	-x+1/2,y+1/2,z+1/2
14	y+1/2,x+1/2,-t+1/2	29	-9,-3,2
15	-9,-15,-2	30	-y+1/2,-x+1/2,z+1/2

the list or existing ones deleted.

Atomic Coordinates...

This shows all the atoms within the unit cell. This list of atoms are generated by applying the symmetry operators on to the set of basis atoms above. This list can not be changed, the changes must take place in the atomic basis or the symmetry operators.

-	_			10	Atoms in th	e Unit Cell			
		Type	Name	X-coord.	Y-coord.	2-coord	DW-f	Oof	
	1	1.5	14 C	D.000000	0.00000	0.000000	5.600	1.000	
	2	1.1	De .	0.50000	0.533333	0.52228	3.600	1.000	
	× -	2	31.	0.000000	0.033333	0.18978	5.600	1.000	
	4	2	Sr.	0.50000	0.999993	0.83978	5.600	1.000	
	5	1	51	0.00000	0.00000	0.89358	3.600	1.000	
	1 K -	2	iir .	0.50000	0.90000	0.09000	3.600	1.000	
	1.		51	0.000000	0.00000	0.59229	3.600	0.870	
	1	3	Di .	D.50000	0.50000	0.88228	3,600	0.070	-
	1.4	Ξ.	8	p.000000	0.000000	0.68788	3.400	0.970	
	11	2	D1	0.50000	0.999993	0.19788	5,600	0.870	
	11	1	Di	0.000000	0.000000	0.25518	5.600	0.150	
	12	3	81	D 50000	0.50000	0.76818	1.400	0.130	
	18	1	59	D.000000	0.00000	0.75198	5.600	0.130	-
	14	3	Di	D.50000	0.533333	0.251.98	3.600	0.130	
	15		Es .	0.00000	0.00000	0.44553	3.600	1.000	Constanting of
	18	4	Es:	0.50000	0.999993	0.94588	5.600	1.000	(Page #)
	17	4	Ca .	0.000000	0.000000	0.55448	3.600	1.000	E .22)
	11		Es .	0.50000	0.90000	0.05448	3.400	1.000	
	1.9			0.50000	0.00000	0.44833	5.600	1.000	((a
	21		1	D.000000	0.533333	0.14511	3.600	1.000	6.00

Ch. 6 Menus - p.73

Calculate Menu

The active commands in this menu depends on the current sta-



tus of the calculation. If the simulation has already been carried out for the current set of parameters, then no commands will be active. If a change has been made or the file is a newly created structure file, the commands showing which subprograms needs to be run are shown active.

Full Calculation

Use this command if you would like the program to run the multislice calculation to its end starting from the point required by the last change made to the simulation parameters.

Projected Potential

Execute this command if you only want to run the PHSGRT program at this time. After the phasegrating is run, the multislice option is highlighted.

Exit Wavefunctions(s)

Execute this command if you only want to run the MSLICE program at this time.

Image(s)

Execute this command if you only want to run the IMAGE program at this time.

"

Weak Phase Object Images

is a separate module that allows the calculation of images that would be produced in the case of an ideal Scherzer lens and validity of the weak phase object approximation. The "wpo" calculation is discussed more in detail elsewhere.

itarting Resolution (Å)	3.00		
nding Resolution (Å)	1.00		
🖲 Auto Decrement Resolu	ition		
Fixed Decrements in st	eps of 0	.2	
Viewing Options			
Viewing Options	splay X & Y	1	1

Integrated Diffraction Pattern- calculates the diffraction pattern by adding up the intensities for each tilt angle within the cone of incident electron directions.

Thickness (A)	200	G-max (1/Å)	Z.00
Disk Size (evod)	0.20		
Image Size Width	512	Height	512

CBED Pattern

Will calculate the CBED pattern for the given input parameters

o ose bioen na	ve care	anac		Course managine encounter
one Axis	1	1	1	
urface Normal	1	1	1	
t Direction	0	D	0	
max for calculati	on [1/Å]		3.50	
ecimen Thicknes	s [Å]		1500	
icroscope Voltage	[kV]		100] 🔍 🕈 🔶
adius of disk in pi	xels		64	-
isk Size				
🔵 🖂 Snap to	spot o	dista	inces	
name Size				
Snap to	spot o	ent	ers	
🗾 🗹 Include	e partia	l be	ams	- + g-max for display [1/A]

using the Bloch-Wave approximation.

Tables

The current operations in this menu are:

Reciprocal Space Info...

Displays information about reciprocal space data for the current structure. The data can be sorted on the reciprocal vector or the



opacing a raigie carcalatoria

Ch. 6 Menus - p.76

	4[X]	9[<u>1/Ă</u>]	S.F.	Ex.[Å]
0 0 0	0.00	0.00	178.96	(0
0 0 2	15.26	0.07	8.34	8496
004	7.63	10.13	3.32	21336
006	5.09	0.20	9.67	7326
0 1 1	3.78	0.26	1.33	53531
013	3.57	0.28	16.66	4252
015	3.23	0.31	32.10	2207
1 1 0	2.70	10.37	(64.88	1092
112	2.66	0.38	15.94	4444
114	2.54	0.39	13.57	19631
1 1 6	2.38	0.42	3.05	23227
0 2 0	1.91	0.52	58.34	1214
022	1.89	0.53	0.58	122628
024	1.85	0.54	1.16	60828
026	1.79	0.56	4.36	(16259
121	1.70	0.59	2.26	31413
1 2 3	1.68	0.59	17.25	9766
1 2 5	1.64	0.61	14.20	4990
2 2 0	1.35	0.74	32.11	2206
222	1.34	0.74	10.52	136107
224	1.33	0.75	0.87	81177
031	1.27	0.79	1.73	:40916

structure factor/extinction distance.

Angles Between Reflections...

Shows the angles between a set of the reflections in the structure.

		An	gias Botios	ien Reflec	LIGHA			
0.	002	0.0-2	0-1 1	0-1-1	411	01-1	1.1.1	1
20.9	0	188,88	192.99	91.12	92.00	97.12	12.10	3
8 8 - 2	180.00		97.12	82.88	97.12	82.88	\$7.12	1
8-8.1	92.99	41.12	0	14.25	165.25	165.55	89.12	1
8-1-1	97.12	82.88	14.25	0	180.00	165.15	90.95	1
	87 38	81.12	165.7%	182.08	1	10.35	88.12	1
0 1 -1	\$7.12	82.88	100.00	165.75	14.25	0	80.80	1
-1. J. T.	82.85	97.12	31.69	76.58	85.12	90.08	1	
-1.0-1	97.12	12.05	90.00	105.12	90.00	109.12	14.25	1
101	82.65	97.12	09.12	20.00	31.60	90.54	165.75	
1.0-1	97.12	82.68	90.99	26.92	90.00	99.12	100.00	
818	69.45	118.85	152.35	165.57	15.45	27.61	87.51	
81-3	110.55	69.45	16657	152 38	2262	18.49	82.49	
-1.1.0	90.00	\$6.09	15456	134.51	45.44	-8.44	45.44	
-88 8	90.09F	91.11	49.44	08.04	111216	104.54	41.44	
1.1.0	90.00	156.58	13456	13456	45.44	1-8-44	13436	1
1-1.0	20.00	98.88	45.44	41.44	13456	154.56	194.96	1
112	79.99	1163.32	132.02	125.44	44.56	47.04	132.02	
11-2	100.02	15.55	135.44	112.02	47.98	44.58	135.44	1
1-1 2	79.90	168.82	4456	41.94	132.02	135.44	132.02	
1-1-2	100.02	15.58	47.98	44.58	135.44	1152.82	135.44	
-1 1 2	78.98	108.82	132.02	115.44	44.96	07.98	\$4.56	
5-1 1-2	100.02	15.58	135.44	53.521	47.90	+456	47.95	1
-8-8.2	79.98	100.02	4436	41.98	152.02	158.44	44.95	1
-1-1-2	100.02	15.58	47.90	44.56	135.44	132.82	47.80	1
0	ALCO PROPERTY.	Contraction in	All the second second	da a constantes	0.000.000	NAME OF TAXABLE		T

Ch. 6 Menus - p.77

Seaech for Angle...

One cacn search for reflections which satisfy a search criteria based on the ratio of the length of the vectors and/or the angle between the measured reflections. This can be used when trying to index diffraction patterns. One can give aan accepted toler-

ingle betw. reflections 45.0	100	Tolera	nce 5.00		
Exclude Kinematically Fort	xidder	Reflection	2015		
earch Method					
Search Only Based on De	esired	Angle			
🔾 Use Angle & Ratio					
Measured d-spacing Ra	atio	1.50	Tolerance	5.00	76
Use Angle & Measured V	'alues				
Measured d-spacing 1	2.00	Ă	Tolerance	2.00	*
Measured d-spacing 2	2.00	A	Tolerance	2.00	%

anse for the ratio, the measured spacings and the measured angle.

Spacing and Angle Calculator...

Shows the lengths and d-spacing for a type/family of reflections. Two reflections can be displayed and the zone in which a set of reflection exist and the angle between them is shown in

Ch. 6 Menus - p.78



the right part of the window. The reflections can be typed in and

when the "Calculate?Update Angles" button is clicked, the angls between the family of reflections and the zone in which they occur will show to the right. The length of the vectors and the corresponging plane spacing is updated live when typing in a new reflection. The lattice parameters can be changed and invoked through the "Update Lattice Parameter" such that the calculator can be used for structures different from the one that is loaded.

Quantitative Menu

This is the menu for quantitative comparison between experimental images and exit wavefunctions with calculated data. Structure refinement and imaging parameter refinement is also



invoked from this menu.

Ch. 6 Menus - p.79

Operating The Routines

All the routines except for the "Symmetry Transform Calculator" requires the presence of an image that is considered the experimental image. The purpose of this package is to provide the user with tools that permit the following attempts:

Quantitative comparison of experimental images with those simulated from a structure model

•A

•A

•Q

Automatic refinement of parameters such as the thickness of the specimen, the defocus of the objective lens, crystal tilt, aberrations, etc..

Automatic refinement of structural parameters such as atomic positions, debye- waller factors and occupancy factors for selected atoms in the structure.

The experimental image can be compared with the computed images using a number of goodness of fit criteria. A sub-area of the experimental image can also be compared to a sub-area of the simulated image. For more information on quantitative comparison methods and structure refinement see the chapter on "Introduction to Quantitative Comparison of simulated HRTEM images with experiment".

Note: All the procedures expect that the experimental image covers the exact area of that of the unit cell used in the calculation. Thus it is up to the user to make sure that the unit cell motif is extracted from the experimental data prior to usage. The term Unit Cell is loosely used since it only refers to the size of the model used in the simulation. The experimental image does not need to be sampled equal to the simulation, since the routines will resample the experimental data to fit that of the simulation.

For parameter refinement and structure refinement, there are a number of algorithms that attempt to look for the "one" solution of parameters that maximize the fit between the experiment and the simulation. This plug-in uses an algorithm based on simulated thermal annealing which is described further in the chapter at the end of this manual. No claims are made as to the effectiveness of this method and there is no guarantee that the final solution represents the global maximum/minimum in the goodness-of-fit parameter. The effectiveness of optimization routines depend on the starting parameters. There is no recipe for setting the initial starting condition and it is necessary to develop some experience using the optimizing routine in this program. Some trial and error is a definitive part of the parameter/structure refinement. Suffice it to say, "good hunting".

Load Experimental Image

Load Experimental Image is the starting point for loading in the image to be used in the comparison. The command will bring up a standard File Open dialog with a look that depend on the version of the MacOS you are using and what else of Finder utilities you have loaded.



The dialog will show you all files and it is up to you to select an appropriate file to read in. Currently, two distinct types of files are supported. These are

1) Binary Files

The data can be Real 4 or Integer 1,2,4 byte 2) **TIFF** Real 4, Integer 1,2,4 byte

If the routine recognizes the file as a TIFF file, it will just open the file and display the image.

For Binary files, you will need to specify the data type, the width and the height of the image. Byte swapping is also supported. If the program does not recognize the file as type TIFF it will bring up the following dialog

File			Format
Langth:	209	7152	- C Test
Data Tvi	1.4		Dims Are flavo
Integer 2	Bytes S	aned	Ges Size By Counting
time.			Binary
size	_		Offset 0
Width: 1	1024	Angle :	Commenter Print
Height: 1	1024	90	Swap Data syses
Simulati	ion Samp	ing	
Width:	256	Height: 32	Angle 90.00

Although the dialog box indicates that the program supports input of text files, this is currently unsupported.

The dimensions of the simulated image are shown at the bottom of the dialog box for binary files.

Once the routine has read in the file, it will display the image in



a separate window. Load Standard Deviation Image

Ch. 6 Menus - p.82

This command allows you to read in a standard deviation image to be used together with the average experimental image for comparison with simulated images. The standard deviation image will be used in conjunction with the average image for computing c-square deviations between the experimental data and the computed data. Otherwise the input works exactly as for the loading of the average experimental image. Again, the image will show up in its own window after it has been loaded.



Note: If the windows are covered by other windows, it may be necessary to move these to bring the "experimental" image and the "standard deviation" image to the foreground since there is no command to bring these automatically to the front.

Compare Experiment With Simulation

This brings up a dialog window which shows the loaded experimental image and the standard deviation image (if loaded). The name of the experimental image together with the dimensions are shown to the left of the image.



When comparing images, the experimental image will be resampled such that the sampling corresponds to the sampling of the calculated image.

The example used here will be that of a set of simulated images of Silicon in the [110] zone axis orientation The simulated images for a range of thickness and defocus are shown below.



Tools

Pointer Tool

Used essentially to ensure that no other tool is active Selection Tool

Use this to mark an area in the image which will be used for comparison. It is okay to go outside the image, since the selection will be cropped to the actual image.

Hand Tool

Use this tool to move the image within the display area. Magnification Tool

Use this tool to magnify the image. Holding down the Option key when clicking within the image will reduce the magnification. Holding down the shift key when clicking will increase/ decrease the magnification by a factor of 2.

Reset View

Click here to reset the view/pan/zoom of the image.

Selecting an area for comparison

The user choose to compare the entire image region or only a

selection of the image.

Two radio buttons are provided:

Compare Entire Image

By clicking on this radio button, the selection area will automatically be set to be that of the entire image. The comparison area is shown as a red rectangle.

Set/Compare Selection

The user must first use the selection tool to mark a region of the image. Upon clicking on the radio button, the selection will be marked in red and this area will be used for comparing images. For changing an already defined regions, mark a new one and click again in the radio button.



Note: The selection does not work for the Difference Image which automatically compares the entire image.

Selecting a method of comparison

The method of comparison or goodness of fit criteria is chosen by a set of radio buttons and the choices and the corresponding description and output is given below. The goodness of fit criteria is computed for each image that is compared and displayed in a table. This table can be saved in a text file for further use.

Statistical cross correlation coefficient (CCC):

A note about the cross correlation coefficient: The cross correlation coefficient measures similarity in the pattern between the experimental image and the computed image. Since the images to be compared are set to a mean level of 0 and normalized, any scaling of the type I(exp) = a * I(calc) + bwould give a cross correlation coefficient of 1. Exact fit is given as CCC = 1 and a reverse contrast gives CCC = -1.

Real Space



This is a straight calculation of the normalized cross correlation coefficient between the experiment and the calculated image(s). For it to give meaningful results, the origin of the experimental image neeeds to coincide with the calculated image.

<u> </u>									307	_		_	_				
	-540	-990	1900	1000	-200	-754	-004	-680	-\$10	+\$50	23000	-1058	-1100	-11502	10000	-1236	-1300
a .	-10046	-8.6765	-8.2149	1.64(8	8,9665	8.6629	8.7972	0.8585	0.9518	0.9787	0.0406	8.0600	3.4058	-1.5118	+6.2165	- # 9938	4.9164
24	-8.9538	-8.6396	1.1164	1.6862	18,9515	18.6640	1.8054	0.8787	0.9646	0.9658	8.8542	14.65.4	1.4647	-8.5958	-8.2662	1-6.0937	-8.0812
40	-8.0058	-1.4815	8.2655	6.7651	8.91-05	8.6512	8.0085	0.8950	0.9716	0.9474	6.9658	18.0458	8-654	-4.5151	4.7158	1-8.69.40	-8.6610
26	-1759	-1.1412	11.4101	67162	8.6702	E.Or So	1.1985	0.901.5	0.8585	0.6241	0.0015	6.6275	8.5804	-6.4001	-E.7412	1-8.0475	-1.0545
22	-3.417	0.2046	16 5500	8.2179	18,7951	18.7266	8.1412	10.8992	0.8464	0.8454	8.8463	18,2963	8.3565	142.3421	-66122	-6.9832	-8.0014
55	-10167	0.4815	14.846	1.2314	16.7858	8.5819	16701	008046	0.8918	0.8181	0.0028	117546	10.1035	-6 2593	-8.9479	-8.750t	-17642
204	0.3085	0.58998	0.7062	0.7278	8,1024	2.3714	8.4010	owoa.oc	0.7348	0.7114	8.7878	1.6900	10.1911	-0.1129	1-0-4911	148.40798	-8.7257
120	0.8489	0.7121	8.2156	87102	8.74/10	EXM2	8.8789	10.1181	0.4(3)	0.0353	18.40-6	110407	1.0419	-0.8822	-1.10-04	-8.10708	-188111
156	10.8125	0.7641	RESI	0.6910	18.1200	-0.1 tm	-0.1047	- 8.080e	0.8193	0.0484	0.0014	1.1814	8.7810	-0.0138	1-0.2440	1-8-4637	-3.5710.
192	0.140	0.8369	0.054	0.6108	1-01118	1-0.9666	-4.5108	-8.5879	- R.222.9	-8.1895	10.8451	8.1658	18.1465	8.0480	-61195	-6.5866	-1.4475
168	0.8026	0.8754	8.8951	1.63(6)	1-05-01	-0.5861	1.1.480	-1.4616	-1.4251	1.8.4871	-0.1141	-0.8996	8,0768	0.1015	8.8180	1-8.1258	-8,0440
4 100 100	1000000	2012/01/2	19.0.000 AT	Avocute.	0000000	000009/201	1000000	1.2270000	0000100	1000000000	10000410	HEM055	wondeall	DOM: NO	000000000	0.2040000	10000

Reciprocal Space

There are two options for calculating the cross correlation in reciprocal space.

The first is an exact calculation which is equivalent to the real space CCC. The entire Fourier transform of the experimental image is compared with the Fourier transform of the simulated image and the CCC is just the reciprocal space equivalent of the calculation in real space. Thus the reciprocal space CCC is equal to the real space CCC.

<u>Exact</u>



Only Amplitudes



This calculates the CCC based only on the Fourier amplitudes and (optionally) the program will try to estimate the shift between the experimental and the calculated image. This can be very useful for aligning images and to compare images when the origin of the experimental image is not known. The numbers given are the shift of the origin that should be attempted to be made on the experimental image before the experimental data is

2								- FD00F-	Amplitude	s —							
ę.	+300	-3210	-300	-1210	-500	-550	-500	-650	-700	-750	-800	-850	-900	-950	-1000	-1050	-1100
8	0.9751-0,0	0.9634	0.5188	0.9665	1303	0.9798	1.5486	0.5440	8.9980 8,0	0.9598	1.9729	0.3760	1.9625	0.9791	0.9452	0.9598	0.8888
24	0.9796	0.9651	0.5618	0.96-62 0, - 8	11116	0.9015	8.2167 8,0	0.6932	8.0602 8,0	0.9695	1.9740	0.9709	6.9771 6,0	0.9559	0.9271	0.8475	0.0211
40	0.9795	0.9584	0.9155	0.9471	1.9511	0.750Z 0.8	12765	0.7130	1.9662	0.9482 09	1.9655	0.9001	1.9054	0.9587	0.9096	0.9282	0.7267
50	0.8788	0.9520 0,8	0.8556	0.9045	1.1512	0.4061 0,8	1.4213 1,0	0.7295	8.9111 8,0	0.9009	1.9590	0.9451	8.9609 8,0	0.9288	0.0071	0.0	0.6-475 0,8
72	0.9748	0.9347	0.0502	0.7927	1.6122	0.8268	3 5665 9.0	0.7458	1.0500	0.0000	8.65TI 19	0.9055	0.9574 8,0	0.9994	0.8519	0.8483	0.5485
53	0.3552	0.9020	0.1145	0.5765	13143	0.5276	8.6404 8.0	0.7496	8.7510 8,0	0.6625 0,0	1,7017	0.8411	1.0977 1,0	0.8541	D.6102 D,1	0.1814	0.4629
10.0	0.9973	0.8444	0.8877	0.5001	1.4181	0.65%	1,7172	0.2485	1.6254	0.4953	1.4727 1,0	0,9330	8.7505 1,0	0.7424	0,7476	0.181.8	0.3776
120	0.8258	0.6792	0.2815	0.4100	11157	0.7856	8.7841 8.0	0.7588	8.4164	0.2165	8,0	0.2915	1.4504	0.5455 0,0	0.6165	0.5784	0.2866
135	0.8484	0.3795	0.3888	0.6212 0.1	1.1655	0.9090	8 8334 8.0	0.2284	1 2237	0.4699	1.505E	0.0	1.3222	0.2545	0.4007	0.4891	0.2826
152	0.5428	0.3639	0.6332	0.7516	1.6215 8,0	0.0441	8.6753 8,0	0.6979	8.4062 8,0	0.0512	8.606£ 8,0	0.6992	8.6257 8,0	0.5444	0.4460 D,8	0.2882	0.2514
Ins	0.2291	0.6658	0 1155	0.8528	11711	0.8811	1.9054	0.6617	16106	0.7687	1.7957	0.7955	8.7719	0.7485	0.7094	0.5514	0.5189

compared to the simulation using the exact formulation.

Chi-Square

This computes the chi-square deviation between the calculated image(s) and the experimental images, optionally using the standard deviation image for obtaining the uncertainty associated with the average pixel value.



If no standard deviation image is given, the uncertainty in the pixel value is set to 10% of the intensity in the pixel. The Chisquare goodness of fit criteria is sensitive to the mean level of the images and the scales the calculated images so that they have the same mean as the experiment before computing chisquare. Lower values indicate better fit, with a value of 1 meaning that the experiment and simulation agree within the uncertainty of the experimental values.

								Chi	-Square								
	-300	-793	-400	~450	-500	-550	-#10	-890	+760	-750	-800	-650	+900	-281	-1000	+1099	-1100
Ó	229.1	1018.8	3018.2	148.7	124.4	\$22.0	110.8	10.18	01.01	182.1	111.8	128.4	18.35	48.73	14.16	84.18	FIL:
24	270.1	50) 7	321.0	\$115	520.5	12127	11.58	3.861	41.02	112.0	140.0	117.4	11.10	30.24	22.78	45.48	158.1
40	205.6	299.8	312.1	52.5.0	292.9	181.0	38.90	1,541	54.75	108.5	111.8	185.1	18.61	25.76	22.48	99.58	142.1
56	260.2	208.1	209.8	208.0	280.5	100.4	21.41	1.4042	25.18	28.85	126.0	12.01	48.40	22.91	07.80	03.08	122.1
72	255.5	269.8	276.1	2083	190.6	58.05	11.52	8.9942	26.68	00.5 8	101.4	71.15	09.06	34.11	15.27	26.42	115.7
66	2.01.1	29(1.8)	104.1	217.4	126.%	38.25	7.818	0.0158	27.38	mass.	97.88	87.41	18.83	1.882	10.14	22.74	108.8
104	236.8	225.4	198.5	155.9	63.22	125.16	5 201	1.105	32.78	49.71	90.11	58.16	1.127	4194	8.735	22.49	95.28
120	219.8	179.4	134.1	99.32	51.79	25.01	1.857	2.168	49.69	109.7	112.7	71.16	2266	5.542	12,88	30.19	91.18
136	193.6	1.23 2	00.04	11.87	41.41	23.079	8.809	6.468	70.5%	104.0	111.5	188.4	81.04	35.93	85.22	498.30	90.41
152	1.49.1	75.51	61.48	57,89	-2.51	22.99	12.21	1261	1123	153.4	157.0	135.0	189.1	85.41	75.61	80.08	92.87
ten	98.23	11.01	48.17	41.31	48.81	26.11	18.82	28.02	101.0	101.1	118-4	1646	155.6	141.0	it at at	128.5	46.12

Root Mean Square Difference

This calculates the root mean square difference between the experiment and the simulation. Lower values indicate better fit with 0 being exact fit between experiment and simulation. Since the values depend on the mean level of the images that are compared, the simulation is scaled to have the same mean as the experiment.

Ch. 6 Menus - p.91



The table below shows the result from the RMSD comparison.

									RMSD								
	-300	-350	-400	-450	-500	-550	-600	-650	-700	-750	-800	-850	-900	-950	-1000	-1050	-1100
8	0.6512	0.6889	0.7110	0.7239	0.7195	0.6468	0.4129	0.1257	0.2806	0.4421	0.4808	0.4461	0.3627	0.2737	0.2292	0.2885	0.5240
24	0.6452	0.6812	0.7035	0.7162	0.7018	0.5786	0.3091	0.0771	0.2536	0.4256	0.4670	0.4249	0.3309	0.2427	0.2064	0.2674	0.4945
40	0.6391	0.6723	0.6939	0.7054	0.6768	0.4976	0.2382	0.0488	0.2312	0.4085	0.4527	0.4040	0.3002	0.2146	0.1859	0.2467	0.4672
56	0.6326	0.6610	0.6792	0.6864	0.6327	0.4025	0.1815	0.0264	0.2137	0.3895	0.4346	0.3779	0.2645	0.1852	0.1656	0.2253	0.4430
72	0.6244	0.6435	0.6516	0.6436	0.5414	0.2988	0.1319	0.0114	0.2026	0.3691	0.4082	0.3373	0.2114	0.1473	0.1429	0.2016	0.4182
88	0.6157	0.6223	0.6128	0.5782	0.4411	0.2344	0.1039	0.0220	0.2047	0.3608	0.3874	0.2972	0.1535	0.1106	0.1255	0.1870	0.3997
104	0.6036	0.5900	0.5525	0.4897	0.3578	0.1967	0.0901	0.0409	0.2245	0.3714	0.3845	0.2800	0.1090	0.0803	0.1159	0.1860	0.3848
120	0.5808	0.5253	0.4540	0.3916	0.2981	0.1797	0.0935	0.0687	0.2764	0.4107	0.4164	0.3324	0.1867	0.1236	0.1389	0.2154	0.3743
136	0.5455	0.4353	0.3680	0.3327	0.2702	0.1801	0.1110	0.0997	0.3420	0.4545	0.4566	0.4027	0.3076	0.2351	0.2156	0.2737	0.3729
152	0.4788	0.3362	0.3073	0.2963	0.2551	0.1880	0.1370	0.1392	0.4153	0.4951	0.4927	0.4608	0.4096	0.3645	0.3410	0.3551	0.3779
168	0.3787	0.2801	0.2756	0.2756	0.2481	0.2005	0.1687	0.1882	0.4827	0.5276	0.5209	0.5031	0.4812	0.4698	0.4670	0.4405	0.3897
•																	

Difference Image



There are two calculations performed in this case. One gives a difference image(s) which are pseudo colored such that where the experiment and simulation agree within one standard deviation, the pixel is black, less than 1.5 standard deviations, the pixel is colored green, and outside the pixels are shown in



shades of red or blue

depending on whether the values in the simulation are lower or higher than those in the experiment.

In addition to the difference image(s), the chi-squared value is also computed for each image and shown in its own table.

	711	-11U	20				192		RMSD	0.0	99	- 510	- 10				725	E
	-300	-350	-400	-450	-500	-550	-600	-650	-700	-750	-800	-850	-900	-950	-1000	-1050	-1100	^
8	0.6512	0.6889	0.7110	0.7239	0.7195	0.6468	0.4129	0.1257	0.2806	0.4421	0.4808	0.4461	0.3627	0.2737	0.2292	0.2885	0.5240	
24	0.6452	0.6812	0.7035	0.7162	0.7018	0.5786	0.3091	0.0771	0.2536	0.4256	0.4670	0.4249	0.3309	0.2427	0.2064	0.2674	0.4945	
40	0.6391	0.6723	0.6939	0.7054	0.6768	0.4976	0.2382	0.0488	0.2312	0.4085	0.4527	0.4040	0.3002	0.2146	0.1859	0.2467	0.4672	
56	0.6326	0.6610	0.6792	0.6864	0.6327	0.4025	0.1815	0.0264	0.2137	0.3895	0.4346	0.3779	0.2645	0.1852	0.1656	0.2253	0.4430	
72	0.6244	0.6435	0.6516	0.6436	0.5414	0.2988	0.1319	0.0114	0.2026	0.3691	0.4082	0.3373	0.2114	0.1473	0.1429	0.2016	0.4182	
88	0.6157	0.6223	0.6128	0.5782	0.4411	0.2344	0.1039	0.0220	0.2047	0.3608	0.3874	0.2972	0.1535	0.1106	0.1255	0.1870	0.3997	
104	0.6036	0.5900	0.5525	0.4897	0.3578	0.1967	0.0901	0.0409	0.2245	0.3714	0.3845	0.2800	0.1090	0.0803	0.1159	0.1860	0.3848	
120	0.5808	0.5253	0.4540	0.3916	0.2981	0.1797	0.0935	0.0687	0.2764	0.4107	0.4164	0.3324	0.1867	0.1236	0.1389	0.2154	0.3743	
136	0.5455	0.4353	0.3680	0.3327	0.2702	0.1801	0.1110	0.0997	0.3420	0.4545	0.4566	0.4027	0.3076	0.2351	0.2156	0.2737	0.3729	
152	0.4788	0.3362	0.3073	0.2963	0.2551	0.1880	0.1370	0.1392	0.4153	0.4951	0.4927	0.4608	0.4096	0.3645	0.3410	0.3551	0.3779	
168	0.3787	0.2801	0.2756	0.2756	0.2481	0.2005	0.1687	0.1882	0.4827	0.5276	0.5209	0.5031	0.4812	0.4698	0.4670	0.4405	0.3897	*
4																		1/1

Fractional Mean Absolute Difference

This calculates the fractional mean absolute difference between the experimental data and the simulated data.



0									IMAD								
÷	-531	-350	-#01	150	-501	-550	-021	-650	-701	-750	-001	-850	-901	-950	-1010	-1050	-1 100
e .	1.561)	0.5804	15:5913	0.6103	Letisz-	0.5615	1.1668	0.0830	8.2298	0.3706	2:4046	0.3721	1.2911	0.21.49	1.1748	0.2273	1.4466
24	1.5621	0.5852	8.5948	0.6190	2.60%8	0.5846	8.2769	0.0572	8.2133	0.3870	8.4082	0.3500	8.2615	0.1877	1150	0.1909	1.4195
40	1.2516	0.5905	8.5968	0.6840	1.5914	0.4512	8.2118	0.0966	8.1918	0.3804	8.5952	0.5462	1.2405	0.1852	1.1312	0.1914	1.3938
96	1,5521	0.5725	0.5815	0.1905	8.5412	0.1442	8.1515	0.0197	2.1845	0.5456	1.5513	0.5907	12155	0.1576	8.1188	0.1759	8,5687
72	1540	0.5552	0.5515	0.1903	1.4523	0.2549	1.1018	0.0075	1,1725	0.3252	1.3614	0.5015	1.1748	0.1956	8,0958	0.1580	1.548
98	1.5311	0.1996	8.0219	0.4779	1.3647	0.1548	1.0568	0.0136	0.1748	0.3897	1.8521	0.2990	1.1211	0.0826	1.0915	0.1510	1.5295
104	0.0258	0.03300	1-2578	0.1991	8.2812	0.1868	8.0748	0.0001	0.1002	0.11 87	3.5236	0.7478	8.0047	0.0619	8,0947	0.1547	6.0004
120	13041	0.4888	0.8617	5.11.22	0.0316	0.1998	2/0719	0.0816	8.2198	0.1280	0.0302	0.2872	0.1291	0.1076	1.1201	0.1804	0.11.08
136	1.4011	0.3404	1.099.0	0.2872	8.8117	0.1896	8.0804	0.0764	1.0848	0.3871	1.2502	0.2330	1.2214	0.3376	1.1991	0.3274	1.21-8
152	1.3911	0.2645	1.2464	3992.0	0.2001	0.1476	1.1082	0.1806	8.5245	0.3952	1.5611	0.3526	1.3248	0.3171	1.5018	0.2972	8.5178
169.	1.1016	03228	1,2218	0.3289	2,1993	0.1806	1.1392	D.1476	8.8942	0.4858	8.4107	p-mot	1.0104	0.4877	1.4118	0.3781	1.2121
•																	

Refine Parameters

Refine Parameters... will attempt to find the image simulation parameters that produce a simulated image that gives the "best" fit between the experiment and the theory. The search routine is based on the concept of "Simulated Thermal Annealing" and requires a starting configuration of parameters, a starting temperature and an ending temperature, a maximum change in parameters and a goodness of fit criteria that measures the "Energy" of the system. For further explanation of Simulated Thermal Annealing, see the last chapter in this manual. Invoking the command brings up the following dialog which is used to give the input parameters to the algorithm.

Temperature		Acove	Parameters	Inital	Value or	aw channe
Starting	8.01000	Thickne	161 **	1	2	not chompe
Ending	0.09010	Gart	and a second	1		100
fractional Change	0.9900	G Defocus (A)			990	100
- Per degree of free	iam	Spheric	al A66. [mm]	0	<i>k</i> 5	1
Attempts per Temp.	25	Spread	of Defocus (Å	1 9	8	10
# Successful Attemp	\$ 5	Conver	gence (mrad)	a	150	.1
Fit Method		1		Value	Angle	
Cross Correlation ((00)	Oystai	Tift[mrad]	0	0	.8
🖓 Cor Hearler Amplica 🖓 Chi Savare	963	27 Astig	matism (Å)	0.00	0.00	10
		I T Astig	(Å) mettem	0.00	6.00	1000
Set Image Comparis	on Region	* For this number of	mess this is a f "unit cell" s	in inte	er givin icknesse	gthe H.
Output 0 Create a log file wit Create a movie of th	ptions h "energy" ver le progress wi	sus "time" indew 1	ine between	frame	s (secon	dsje 🛙

Setting Image Comparison region

The area to be used for the comparison is set using the button

Specify Area to Campare Concerner Richters Investi Ameri View

"Set Image Comparison Region"

Selecting the image region is done in the same fashion as under comparing images in the previous section.

Running the parameter refinement

After selecting the area (not needed if the entire image is compared, which is the default) and selecting the parameters to be refined and clicking OK, a progress window for the parameter

Temperature		-	Active Parameters				
Starting	0.0100	0		inta	Value	max change	
Ending	0.0001	p	Thickness [Å]	2	0.0	5	
Fractional Change	0.9000		Defocus (Å)	-	300	100	
For degree of freedom		ref E	Spherical Abb. [mm]	0	60	.1	
Altempts per Tem	n. 25		Spread of Defocus U	4 3	ġ.	10	
# Successful Arter	mps 5		Convergence (mrad)	0	200	.1	
Fit Method				Value	Angle		
Cross Correlation (CCP)		Crystal Tilt [mrad]	D	0	.1		
CCF Fourier Amp	aktudes		🗍 2F Astigmatism (Å)	0.00	0.00	10	
C Chi Square			🗍 🗐 1F Astigmatism (Å)	0.00	0.00	1000	
(Set Image Compa	rison Region	0	*Per thidness this is as in "Unit cdf" silce thidnesses	kiger g	ving the r	vorsber of	
Output Options	ith Sectors's		"				
Create a movie of	the progress	wind	w Time between frame	en (nec	onds]:	1	

refinement appears. If the refinement is not progressing in a satisfactory way, the computation can be canceled by hitting the



Apple-Command Key together with the Period. Optionally, a

log file can be written so that the "energy" as a function of time/ temperature can be plotted and also a Movie can be produced and played back. The number of frames/second for output can be set.

At the end of the run, the dialog box can be just dismissed and the final configuration of parameters will be discarded, or the parameters can be saved in the form of a new structure/simulation file.

Refine Structure

The structure refinement works in the same way as the parameter refinement. However, in this case it is the structure that is being varied, notably the coordinates of selected atoms and possibly debye-waller factors and occupancy. After invoking the

Temperature		Active	Atom Parameters				
Starting	0.01000		Maximum shift [Å]	0.100			
Ending	0.00010	Set Active Elements					
Fractional Change	0.9000	0.0					
Per Degree		00	mparison Area / Active A	voms			
Attempts per Temp.	25	Set Chemical Constraints					
# Successful Attemps	5	C	Set Bord Valences				
Fitting Method							
🗹 Use image agreemer	va av		mage Agreement	tion			
Include Bond Distan	ce Constraint:	5	- cross correla				
Include Bond Valenc	e Sum Optimi	zation	Chi Square				
Output Options							
Create a log file with	'energy' versa	us 'time'		[secs]			
Create a movie of the	e progress win	dow Tit	ne between frames	1			

command, the following dialog box appears.

There are several options associated with the structure refinement, such as which elements are active, which coordinates to be varied, etc.. The standard parameters for the simulated thermal annealing need to be specified, together with the goodness of fit parameter. Output options such as a log file and movie are identical to that under parameter refinement. In addition, it is possible to include in the calculation of the "Configuration Energy" terms that depend on selected bond distances and selected bond valence sum.

Set Active Elements

brings up the following dialog. By default all elements are active. Selecting an active element and clicking on the "Make inactive" button will move the selected element to the Inactive list. Just because an element is listed under active elements does

nactive Sr		Active
Ĩ.	Make Inactive	B
	E Make Active	0

not mean that one of its types will be used in the refinement pro-

cedure. It is necessary to specify the area used for comparison and which atoms are to be optimized before the simulated thermal annealing is carried out.

Set Are to Compare & Active Atoms

is used to set the image comparison region (as previously explained) and to select atoms for refinement. The following window appears.

MacTempas User Manual

Experimental Image		Starting Model	Comparison Area	_
			Selection	3
	1		C Dittire Image)
			Atoms to refine	
DISCOURS IN			Add Selection	0
	Ŧ		Add/Group Selection)
			Atoms in Comp. Ryn	0
be tools to manip slate model and image			(Optimizy	5
Show only non-symmetry related at	oms.	Others move according to symmetries.	Deselect All Atoms	3
East Ves	w)	View Model From 0 0 1		_

Area used for comparison

Selection Sets the comparison region to the selection rectangle. Entire Image Sets the comparison region to the entire image.

Atoms to optimize

Add Selection Includes in the list of atoms the ones that fall within the selection rectangle set in the model window.

Add/Group Selection Adds the atoms to the refinement list, but constrains all the atoms in this list to move as a unit. Atoms Within the Comparison rectangle

Sets the refinement list to include the atoms that lie within the rectangle set for comparing images.

Optimize...

Specifies which properties of the selected atoms (those in the refinement list) that are varied. The following window appears.

Name	x	On	Y.	On	z	On.	DW	On	OCC On	í
Co.	0.0000	4	0.0000	4	0 0 0 0 0 0	0022	3.6000		1.0000	
Sr	0.8903	1	0.0000	4	0.0000		3,6000		1.0000	
Cu	0.4456	4	0.0000	4	O.D.DOGO		3.6000		1.0000	
0	0.4460	1	0.0000	1	0.5000		3.6000		1.0000	- 1
Bi	0.2681	16	0.0000	4	0.0000		3.6000		0.1300	- 1
51	0.3022	4	0.0000	4	0.0000		3.6000		0.8700	- 1
Sr.	0.1097	1	0.0000	1	0.0000		3.6000		1.0000	- 1
0	0.5540	4	0.5000	4	O.D.D.D.D.D.D.D.D.D.D.D.D.D.D.D.D.D.D.D		3.6000		1.0000	- 1
Ð	0.7319	1	0.0000	1	0.0000		3.6000		0.1300	- 62
Bi.	0.6978	4	0.0000	4	0.0000		3.6000		0.8700	- 47
0	0,4460	1	0.500D	1	0.0000		3.6000		1.0000	11

Normally varying the z-coordinate will not have any effect since the image simulation procedure uses a 2d projection of the atoms in the unit cell. However, in the case where symmetry operators are used, changing he z-position may result in changes in x and y for symmetry related atoms.

Each time an atom (or group) is varied, a new set of parameters for this atom/group is created and tested. If "Test each variable separately" is checked, each try will only vary one parameter. Thus if x and y are checked above, both x and y for an atom are changed at the same time unless "Test each...." is checked, in which case one time x is varied and another time y is varied.

Deselect All Atoms

clears the refinement list and allows the user to start all over, defining the set of atoms to optimize.

Show only symmetry related atoms/Use Symmetry elements If the structure is a perfect crystal defined by a set of basis atoms and a set of symmetry operators, it is possible to refine positions of the atoms in the basis and to move symmetry related atoms accordingly such as to preserve the crystalline spacegroup.

Checking this options will result in only atoms in the basis to be visible in the model. Thus the atoms to be refined is selected from the basis and any change in the basis is reflected in the entire structure.

Experimental image	Starting Model	Comparison Area
		(Selection)
		Atoms to refine
		Add Selection
		Add/Group Selection
		Atoms in Comp. Rgn
as heats to rearrigative model and image		Optinize.
Show only non-symmetry related ato	ns. Others move according to symmetries.	Deselect All Atoms
(View Model From 0 0 1	

Set Chemical Constraints



brings up the following window.

which can be used to specify a configuration energy that includes terms that depend on bond distances. It is possible to specify the optimum bond distance between two atoms, whether it is a 2d or 3d constraint, and to specify the weight of the term in the energy calculation. Even though bond distances have been defined, the inclusion of a bond-distance energy term can be turned off/on from the main "Refine Structure" window.

Include Bond Valence Sum Optimization

This allows the bond valence sum to be used to measure the energy of the configuration. It brings up the following window in which the active bonds are specified, the maximum distance between atoms for calculating the bond valence sum, the valence of the first atom in the atom-pair and the bond constant. Each "bond" has its own weight and the entire bond-valence

Bond St	rength	Calcu	lation Criteria:			
Active	From	То	Max. Distance (Å)	Valence	Bond Constant	Weight
2	Ûł.	Ċð.	3.0	1	1.815	10
0			3.0	1	1.815	1.0
8			3.0	1	1815	10
8			3.0	1	1815	10
0			3.0	1	1.815	10
Overal	Weight	. 16	0.0100			
19212122			1020-221 JL			

energy terms has an overall weight.

Running the Refinement

Once the parameters are set and OK has been clicked, a progress window appears. The current atomic configuration is shown together with the corresponding simulated energy. The energy as a function of "time/temperature" is shown in its own window and can be monitored to ensure that the system moves in a desirable fashion. It is important to understand that no specific recipe can be given to ensure that the system finds a meaningful minimum in the configuration energy. The success of the optimization depends on how far the starting configuration is from the "solution" and the choice of annealing parameters. It is not a straight forward, just run and you get the correct answer, black box approach. An understanding of the system, a good feel for choosing a reasonable starting structure and some experience in choosing "annealing parameters" is definitely a requirement in order to have confidence in the resulting ending configuration. Experimenting with different input parameters is advised.

As with refining simulation parameters, it is possible to save a log file or to produce a movie of the annealing process. Saving a movie can be very useful when the refinement takes hours/days and it is necessary to see how the system varied over time. It is important to realize that a movie file grows quickly in size and that the number of frames/second should be chosen appropri-



ately if the movie is to cover a period of many hours.

At the end of the run, the final structure can be dismissed or saved as a structure file for later use. The run can also be aborted by the normal "Apple + Period" key combination.

Symmetry Transform Calculator

Choosing the "Symmetry Transform Calculator" brings up a modal dialog window that can help the user find another set of symmetry operators and basis atom positions in the case of a change in axis and origin of the unit cell. Input is the original symmetry operators which come from the spacegroup that is in use together with the original basis atoms. The new unit cell axes **a',b'** and **c'** are given by the transformation matrix T such that

$$\begin{bmatrix} \mathbf{a}' \\ \mathbf{b}' \\ \mathbf{c}' \end{bmatrix} = \begin{bmatrix} T_{11} & T_{12} & T_{13} \\ T_{21} & T_{22} & T_{23} \\ T_{31} & T_{32} & T_{33} \end{bmatrix} \begin{bmatrix} \mathbf{a} \\ \mathbf{b} \\ \mathbf{c} \end{bmatrix}$$

together with a translation of the origin specified in the old unit cell system (fractional coordinates).

Invoking the command brings up the following window.

0.0	Syn	nmetry Operator Transformations					
Original Operators ×3;2 +1/2,y+1/2,2+1/2 -8,y,2 -9,4/2 -9,4/2 -9,4/2,2+1/2,2+1/2 -9,4/2 -9,4/2 -9,4/2,2+1/2,2+1/2 -9,4/2 -9,4/2 -9,4/2,2+1/2,2+1/2 -9,4/2 -9,4/2,2+1/2,2+1/2 -9,4/2 -9		New Operators 2,9,2 341/2,9+1/2,2+1/2 -3,-9,2 -4+1/2,9+1/2,2+1/2 -9,42 -9,41/2,9+1/2,2+1/2 -9,42 -1/2,9+1/2,2+1/2 -9,47 -4+1/2,9+1/2,2+1/2		Transfe takes ti the new Transfer X' = TX 1.000 0.000 0.000 Transfer	ermation he old a mation m 0.000 1.000 0.000	a that ses into natris 0.000 0.000 1.000	
Original Basis	191	New Basis	122	d.bdb	D.0D0	O.DOD	
Ca 0.000,0.000,0.000 Sr 0.000,0.000,0.110 Bi 0.000,0.000,0.302 Bi 0.000,0.000,0.268 Cu 0.000,0.000,0.268		Ca 0.000,0.000,0.000 Sr 0.000,0.000,0.110 Bi 0.000,0.000,0.302 Bi 0.000,0.000,0.268 Cu 0.000,0.000,0.268		Note that translati- cannot g represen spacegro	t an arbit on of the enerally i ited by th sup	rary origin be e same	
Original Unit Cell Atoms Cs 0.000,0.000,0.000 Cs 0.500,0.500,0.500 Sr 0.000,0.000,0.110 Sr 0.000,0.000,0.890 Sr 0.500,0.500,0.890 Sr 0.500,0.500,0.890 Bi 0.000,0.000,0.302 Bi 0.000,0.000,0.802 Bi 0.000,0.000,0.698		New Unit Cell Ca 0.000,0.000,0.000 Sr 0.000,0.000,0.100 Sr 0.000,0.000,0.110 Sr 0.000,0.000,0.810 Sr 0.000,0.000,0.890 Sr 0.000,0.000,0.302 Bi 0.000,0.000,0.802 Bi 0.000,0.000,0.898		Switch S	pacegro gonal ort New	(Set)	
810.500.0.500.0.198		8+0.500.0.500.0.198	÷			onvert	

Lists

Original Operators

This list shows the symmetry operators of the current spacegroup. Initially these are those of the crystal in use, but the spacegroup can be changed by the command "Switch Spacegroup".

Original Basis	The basis atoms are the atoms of the current crystal.
Original Unit Cell Aton	<i>ns</i> This list gives the atoms that are produce by the operation of the symmetry operators of the spacegroup in use on the original basis atoms.
New Operators	These operators are the result of applying the transformation operations that are given by the change in coordinate system together with a translation of the origin to the generators of the original spacegroup.
New Basis	This is the transformed basis.
Original Unit Cell Aton	<i>ns</i> This list gives the atoms that are produced by the operation of the new symmetry operators of the spacegroup used on the new set of basis atoms.
	Convert Clicking on this button initiates the computation of the trans- formed set of symmetry operators, the transformed basis and the new atomic positions.

MacTempas User Manual

0.0	Syn	metry Operator Transforma	ations				
Original Operators		New Operators		Transformation that			
x,y,z	0	x,y,2	0	takes the old axes into the new axes Transformation matrix X ⁺ = TX			
x+1/2,y+1/2,z+1/2		x+1/2,y+1/2,z+1/2					
-x,-y,z		x,-y,-2					
-x+1/2,-y+1/2,z+1/2		x+1/2, y+1/2, z+1/2					
~y,X,Z		x,z,-y		11 117	100	-	
-y+1/2,x+1/2,z+1/2		x+1/2,z+1/2,-y+1/2		0	0	1	
y,-x,2		x,-2,y		-1	0	0	
y+1/2,-x+1/2,2+1/2		x+1/2,-2+1/2,y+1/2	1	0	1.	d	
-x,y,-Z	4	-x,-y,Z	4	•	1	9	
-x+1/2,y+1/2,-z+1/2	*	-x+1/2,-y+1/2,z+1/2	Ŧ	Translation of origin			
Original Basis		New Basis		0.000	0.000	0.000	
Ca 0.000,0.000,0.000	0	Ca 0.000,0.000,0.000	0	Note that	at an arbit	bitrary	
Sr 0.000,0.000,0.110		Sr 0.110,0.000,0.000		translation of the origin cannot generally be represented by the same spacegroup			
Bi 0.000,0.000,0.302	- E.	Bi 0.302,0.000,0.000					
Bi 0.000,0.000,0.268	4	Bi 0.268,0.000,0.000	- A				
Cu 0.000,0.000,0.446	4	Cu 0.446,0.000,0.080	Ť.				
Original Unit Cell Atoms		New Unit Cell					
Ca 0.000,0.000,0.000	0	Ca 0.000,0.000,0.000	0	Switch	witch Spacegroup:		
Ca 0.500,0.500,0.500		Ca 0.500,0.500,0.500		Take	-		
Sr 0.000,0.000,0.110	-	Sr 0.110,0.000,0.000	-	Tetra	igonai		
Sr 0.500,0.500,0.610		5r 0.610,0.500,0.500					
Sr 0.000,0.000,0.890		Sr 0.890,0.000,0.000		11.5			
Sr 0.500,0.500,0.390		Sr 0.390,0.500,0.500		Exp	ort New	Set	
Bi 0.000,0.000,0.302		Bi 0.302,0.000,0.000		-			
Bi 0.500,0.500,0.802		Bi 0.802,0.500,0.500					
Bi 0.000,0.000,0.698	4	Bi 0.698,0.000,0.000					
Bi 0.500,0.500,0.198	*	Bi 0.198,0.500,0.500			(Convert	

Note:

It is important to realize that arbitrary input does not result in a symmetry which still can be presented by the same spacegroup with a change in symmetry operators.

Export New Set

Process Menu

Allows the user to export the new basis and the symmetry operators as a new structure file.

The Process menu is the largest menu and is the source of all
Process Text Windo	w Help
Image Calculator	0%C
FFT	26 F
Hanning Masked FFT	Ó% F
Inverse FFT	261
Power Spectrum	
Edit Mask	1000 M
Apply Mask(s)	JCH M
Wiener Filter	V.W.Z
Spatial Filters	•
Invert	
Transform	*
Statistics	•
Colibrate	728IC
Extract From Complex	•
Auto Correlation	
Cross Correlation	
Azimuthal Average	- F
Template Matching	
Find Peaks	
Edit Lattice	
Fit Lattice	
Analyse Displacement	Sec
Average/Extract Motif	
Change Image Origin.	.4
Cryst. Image Processin	ng
Find Focus From Imag	e
Find Focus Preference	s
✓ Always Create New Im	age

image processing functions. There are menu sub-menus as well.

Image Calculator

This is a general image and number calculator using reverse

MacTempas User Manual

00		Imag	e Calcula	tor				
Stack	100	144		121213	45454	8		BBB
4:	Image	Selecto	or : (A1	: untitled	d		•	New
	FFT	Min	Mean	Mod	Real	x+iy	=x	=x+iy
3:	FFT ⁻¹	Max	Stdv	Phase	Imag	x e ^{iy}	ceiling	floor
				Inten.	Conj.		Create	Window
2:	x ²	x ³	x ⁿ	x!	Rnd	Roll	+/-	Rad
	VX	∛x	X ^{1/n}	1/x	%	Swap	π	Deg
1:	7	8	9	+	AC	Sin	Cos	Tan
	4	5	6	-	С	Sin ⁻¹	Cos ⁻¹	Tan ¹
	1	2	3	*	Drop	Log ₁₀	Log ₂	Loge
X: A1.0	0		Exp	1	Enter	10×	2×	e×

polish notation (HP style calculator).

Crystallographic Image Processing...

Can be invoked on a real space image which is square or has a



Fourier transform need to be calculated. Once the Fourier transform has been calculated, the reciprocal space lattice needs to be set using the a* and b* tools and clicking on two consecutive

Ch. 6 Menus - p.111



reflections that defines the reciprocal space. The number of

reflections used in the lattice refinement and information extraction can be limited by the circular aperture tool. Once the lattice reflections have been marked, the next action is to invoke the command "Refine Lattice".



After the lattice has been refined, click on "Find Motif". This will extract all the phases and amplitudes from the Fourier transform at the lattice points and one can test for possible symmetries of the motif.

0.00		202020303000000000000000000000000000000
ла		
20 35.16 96m 62.85	Show Unit Cell Outline	Click Defines New Origin Set Current Origin as Origin for P1
	Contraction of the second s	and the second sec

Each symmetry can be tested and imposed on the reflections to form a new unit cell motif. The origin of the unit cell motif is dhown on the right and can be changed.



Once a solution has been chosen, a new image can be created with the imposed symmetry and can be used as an image to compare with simulation.

Text Menu

This menu determines the appearance of text drawn in the MacTempas image window. The following text attributes can be set:



Windows Menu

Use this menu to bring a window to the top of the screen in case it has been completely covered by another window.



Chapter 7

Input File Format

The structure file created by New... in the File Menu is a file of type 'TEXT' and can be produced by a text editor. At times it is desirable to edit the file directly, rather than using MacTempas to create this file. In fact the user may sometimes want to write a program to generate the data in the structure file. For that purpose in particular, the format of the structure file <structure-name>.at is given below:

Line #	Parameter(s)	Meaning
1	litle	structure
2	SpaceGroupNumber	Just as is says, one of the 230 spacegroups, (1-230).
3	abcabg	The lattice parameters and angles
4	Gmax	The maximum reciprocal lattice vector in the multislice calcula- tion. The potential is evaluated out to twice this value, units Å-1.
5	iu iv iw	The direction of the electron beam in units of the real space crystal lattice vectors.
6	NSymops Nslices (I3d)	Number of symmetry operators, number of slices per unit cell, and a flag indicating 2d (0) or 3d (1) potential calculation only if Nslices is different from 1.

Line #	Parameter(s)	Meaning
7	NBasis Ntypes	The number of atoms in the basis, the number of different types of atoms. A different type is associ- ated with a different chemical symbol or a different Debye- Waller factor.
8	it symb x y z dw occf	The type of atoms (a number from 1 - NTypes), Chemical sym- bol, x-,y-,z coordinates in relative units of the lattice vectors, Debye-Waller factor and occu- pancy factor.
9	The same as line 8 for	atom number 2.
10	The same as line 8 for	atom number 3.
8+NB	asis MicName Cs Del Th	The name of the microscope, the spherical aberration (mm), the spread of defocus (Å) and semi-angle of divergence (mrad).
9+NBa	asis Voltage	Accelerating voltage (kVolt).
10+NI	Basis	
	Lh Lk	The center of the Laue circle in units of the h and k of the trans- formed reciprocal unit cell. (Real numbers).

Meaning
The specimen thickness or T1,T2,DT First thickness, last thickness, increm. The commas are required.
Amplitudes to be stored as for possible plotting, (YES/NO).
The indices of the reflection to be stored, or if IPlot == NO then :
Objective lens defocus or
First defocus, last defocus, incre- ment. The commas are required.
-
Radius of the objective lens Aperture in units of Å-1.
The center of the objective lens
aperture in units of h,k of the transformed reciprocal unit cell.
L
The center of the optic axes in the same units as Ah,Ak.

Ch. 7 Input File Format - p.117

Meaning
Symmetry operator number 1. An example is $x+1/3,y+5/6,z+1/3$. The commas are required.
The calculation status of this structure. For a new structure this should be 1
Halfwidth of mechanical Vibra- tion in A.

Note: If different wordprocessing software is used, Microsoft Word, Write Now etc., make sure that the text file is saved at the end as type TEXT.

Chapter 8

Sample Calculation

As an example of a calculation using MacTempas we consider a BCSCO super-conductor structure. Using the structure determined by Tarascon et al (1988), we show the steps necessary to input the model structure, examine it, compute the diffraction pattern and simulated images, and display and print them.

The Structure

As published by Tarascon et al in Phys. Rev. B 37 (1988) p.9382-9389, the tetragonal structure has the following parameters -

Space group: I4/mmm

Cell parameters: a=b=3.814Å, c=30.52Å, a=b=g=90

with nine atom positions in the basis:

Atom	Wyckoff notation	Х	у	Z	Occupancy
Ca	2a	0	0	0	1
Sr	4e	0	0	0.1097	1
Bi	4e	0	0	0.3022	0.87
Bi	4e	0	0	0.2681	0.13
Cu	4e	0	0	0.4456	1
O(1)	8g	0.5	0	0.446	1
O(2)	4e	0	0	0.375	1
O(3)	4e	0	0	0.205	1
O(4)	4d	0.5	0	0.25	0.065

Isotropic thermal parameters for all atoms are fixed at 3.6 Å².

Ch. 8 Sample Calculation - p.119

Entering the Structure

To enter a new structure into MacTempas, we first go to the **FILE menu** (Section 3.3), and select **New Structure File...** After entering a filename in the New File dialog, MacTempas will put up a dialog into which the relevant information must be entered. Note that the program shows a default cubic structure. We need to change the data to reflect that of our structure.

Filename

BCSCO

Specify a filename under which to file the input data. It should be descriptive enough to be easily remembered when you need to open it later. Make sure you use no extension.

A) 3.8340 Algha (deg.) 90.00 Dm A) 3.8340 Beta (deg.) 90.00 Nar A) 3.05200 Gamma (deg.) 90.00 Nar A) 30.5200 Gamma (deg.) 90.00 Nar A) 30.5200 Gamma (deg.) 90.00 Thu X Algena (deg.) 90.00 Thu Star X Algena (deg.) 90.00 Thu Star X Algena (deg.) 90.00 Thu Star X Algena in UDell 18 Star Tap X different acoms S Tap Tap Croscope and Loos Parametiers Croscope (lange (lange) Tap Tap rese (lange (lange) 20.00 Cod Tap Tap rese (lange (lange) 20.00 Cod Tap Tap reserversences angle (mraid) 0.15 Phase Plate Tap reserversences angle (mraid) 20.00 -300 -300<	ants (swert) D 1 ber of Six x (A-1) k, (beg, inc Ampl, /Ph of Laws (wit (mrad) of Absorp	0 D es per cel ases 0 ciscle 0 6 angle 1 storn	2 0 0 5 0 0 5 0 0 0 00 0 00	Implies 0 1 0 2 0.00 200 200 200 1 Vess 0.00 0.00 0.00 0.00 0.00
A) 3.8140 Algina (deg.) 90.00 A) 3.8140 Beta (deg.) 90.00 A) 3.5200 Gamma (deg.) 90.00 acagroup # 0nt. Tableol 138 The X Aomis In Basis 9 Snow Snow X Aomis In Basis 9 Snow Ea X Aomis In Basis 9 Snow Ea X Semin Ops. 32 Blow Ea X Semin Ops. 32 Blow Ea X defitesen acoms 5 Snow Ea X defitesen acoms 5 Snow Ea K defitesen acoms 5 Snow Ea Reservers angle (mrad) OHSOO Ea Top Invergence angle (mrad) 0.15 Phase Bate Snow read of deficus (A) 20.00 0.00 300 -300 s time apert read. (A-1) 1.25 Stronij central beam k Rag Angle	D 1 ber of Six x [A-1] k. (beg.ine e Ampl./Mo of Laue I of Absorp	0 D es per cel jases 0 Ciscle 6 angle " ston	0.00 0.00 0.00	1 D 2 2.00 200 Yes 6.00 0.00
A) 3 8140 Beta (blog.) 90.00 Nur A) 30.5200 Camma (deg.) 90.00 Final atterroup 4 det. Tableol 139 This X Atoms in Basis 9 Show Stow Stow X Atoms in Basis 9 Show Stow Stow Stow X Atoms in Basis 9 Show Stow East Stow Stow X Atoms in UCell 38 Show East Stow East Stow East X different atoms S Chick Stop Typ Typ croscopps and Lons Parameters Chick Final 0.60 Typ croscopps angle (traal) 0.15 Phase Plate Num Tock Tool Tool </td <td>ber of Six x [A-1] k. (beg.ink e Ampl./Ph of Laue (http://www.i of Alasarp</td> <td>es per cel jand) 20 ases (Ciscle 6 angle " stion</td> <td>0 0 Sat 0.00 0.00</td> <td>2 2.00 200 Yes 8 0.00 0.0</td>	ber of Six x [A-1] k. (beg.ink e Ampl./Ph of Laue (http://www.i of Alasarp	es per cel jand) 20 ases (Ciscle 6 angle " stion	0 0 Sat 0.00 0.00	2 2.00 200 Yes 8 0.00 0.0
A) 30.5200 Camme (deg.) 90.00 acegroup 4 Ont. Tableol 139 X Atoms in Basis 9 Show X Symm Ops. 32 Heav of atoms in UCell 38 Show d different atoms 5 Tap croscope and Less Parameters croscope Anne 04500 Itage (UV) 200.01 CL (mm) 0.60 med of defocus (Å) 20.00 0.00 m focus Reguinc.end0 (Å) -300 0300 is lems apert. and (Å-1) 1.25 Strong central beam <u>k</u> <u>k</u> Rag Angk ²	x (A-3) k. (beg, inc e Ampl, /Ph of Laue (inf Absorp	vend) 20 lases (Cixcle 6 angle " stion	0 0 Sat 0.00	2.00 200 Yes ¢ 0.00 0.0
A does in Basis 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9	k. (beg.inc e Ampl.,/M of Laue I itt (mrad) of Alasorp	,and) 20 ases (Circle 6 angle [*] ston	0.00 0 Sat 0.00 0	200 Yea k 0.00 0.0
Adoms in Basis 9 Snow Star Symm Ops. 32 Blow East Set of a constraint of the set of	Ampl./Ph of Laue I itt (mrad) o of Absorp	ases d Cincle & angle " xion	Sat 0.00 0.00	¥ea k 0.00 0.0
Yomm Ops. 32 How Cee Second State Second	of Laur I III (mrad) of Absorp	Circle & angle " stion	a.aa a.aa	• 0.00 0.0
of elorno in UCell 38 Stow Eq. of different actions 5 Typ Encecope and Loos Parameters Croscope Name Or000 Encecope and Loos Parameters Croscope Name Or000 Encecope and Loos Parameters Croscope Name Or000 Encecope and Loos Parameters Or000 Encecope Name Croscope Name Or000 Cs (mm) 0.60 Encecope angle (mrast) 0.15 Phase Plate wad of defocus (Å) 20.00 0.00 -300 is lams apert, and (Å-1) 1.25 Strong central beam 4	of Absorp	& angle " ston	0.00 h	0.0
d different atoms 5 Typ croscope and Loss Parameters croscope Name 04300 tage (kv) 200.00 Cs (mm) 0.60 reorgence angle (mrzs) 0.15 Phace Plate wad of defocus (Å) 20.00 0.00 m focus theg.inc.end(kl -300 0.00 -300 s fem apert. red. (Å-1) 1.25 Strong central beam + k Rag Angle*	of Absorp	sion	0.00	0.0
Tep knoscope and Loss Parameters croscope Name 04300 ktage (k/v) 300.00 Cs (mm) 0.60 nvergence angle (mras) 0.15 Phace Plate wad of defocus (Å) 20.00 0.00 n focus theg.inc.end) (Å) -300 -300 -300 s. Imm apert. ned. (Å-1) 1.25 Stroniji central beam k Nag. Ang.k*	of Alasorp	sti on		
croscope and Lens Parameters croscope Name Criscop tage (k/v) 300.00 Cs. (mm) 0.60 moregence angle (mras) 0.15 Phase Plate read of defocus (Å) 20.00 0.00 0.00 s. kms apert. read. (Å-1) 1.25 Strong central beam			_	418
Crosscope Name CH350 Itage (JUV) 300.00 CL (mm) 0.60 revergence angle (mrad) 0.15 Phase Plate read of defocus (Å) 20.00 0.00 m rocus Reguinc.end) (Å) -300 0 -300 is limit apert. read. (Å-1) 1.25 Stroniji central beam 6 Nag. Angle*				
trage (k/s) 300.00 Cs (mm) 0.60 revergence angle (mrad) 0.15 Phase Plate read of deficus (Å) 20.00 0.00 focus flegunc.end) (Å) -300 0 -300 is tims opent.end. (Å-1) 1.25 Stroniji central beam 6 Nag Angle*	AIRE	ation (ALC	Casa	
Itage (AV) 200.01 CL (mm) 0.60 invergence angle (mrad) 0.15 Phase Plate read of defocus (A) 20.00 0.00 m focus theguno.end/(A) -300 0 -300 is lems apert, red. (A-1) 1.25 Strong central beam t k Neg Angle*		-		ugle [1]
rvergence angle (mrad) 0.15 Phase Plate read of defocus (Å) 20.00 0.00 n focus theguno.endi (Å) -300 0 -300 s lems apert, rad. (Å-1) 1.25 Strong central beam <u>k k Rag. Angle</u>	Taxa Data	4 0		0.0
read of defocus (A) 20.00 (0.00 m focus theguns, end) (A) -300 (0.00 m s term opert, and (A-1) (1.25 Strong central beam () (A Rag Angle*	100100	1.1		0.0
focus (beguinc.end) (A) -310 0 -300 s lens epert. red. (A-1) 1.25 Strony central beam <u>+ + Nug Argin</u>	Trinen 12	v as		0.0
s. lens apert. nad. (A-1) 1.25 Stroniji central beam + k Neg. Angle*	Conte	1000	9	0.D
Strong central beam + + Mag. Angle"		ical Histari	ice (A)	
	Since	12.14	10.00	0.00
AL of Obj. Lens Aprt, 0.00 0.00 0.00 0.0	Starte	14 VAL 14	0.00	0.00
nt, of the Optic Axis 0.00 0.00 0.00	Angle	With 3 - 22	an 1.1	(4.4
er-ride calculation status				
ek for re-calculation 🔅 Projected Potential 🔅		unction	Die	age .
rk as calculated 🛛 Projected Potential 📋	Exit Waven	112422		age
	Exit Waven Exit Waven	unetian		Contraction of the local division of the loc

Space group # 139 From the structure information, we know that the cell is tetrago-

nal with a space group I4/mmm. From Table 6.2.1 of the International Tables for Crystallography, we find that the space group number for I4/mmm is 139. Choose the correct spacegroup from the popup menu.

a

Enter the correct value for the lattice parameter a. In this example MacTempas knows that **b** is equal to **a** for the tetragonal space group #139, and so enters **b** automatically once **a** has been set. Similarly, MacTempas puts in the correct unit cell angles, since they are defined by the space group (in this particular example). Note that cell parameters are input in Å, not in nm.

С

30.52

3.814

The value of the C cell parameter is input in Å.

Gmax

(default=2)

Gmax is the size of the "multislice aperture" and defines how far out in reciprocal space the diffraction calculation will extend. The value of \mathbf{G}_{max} is automatically set to 2.0 reciprocal Ångstrøm units, so that MacTempas will compute all of the dynamically-diffracted scattered beams out to this value, by considering all their interactions with phase-grating coefficients out to twice \mathbf{G}_{max} (a default of 4.0 reciprocal Ångstrøm units). Note that these default values (2 for the multislice and 4 for the phase-grating) are normally large enough to ensure that all significant contributions to the dynamic scattering are included; however \mathbf{G}_{max} is displayed in the MacTempas menu so that it can be set to a larger value if greater precision is required with a structure that includes heavy atoms.

Zone Axis

0,1,0

The correct response is the set of three integers that defines the direction of the electron beam with respect to the specimen (or the specimen orientation with respect to the incident electron beam direction). In this example we choose to enter 0,1,0 in order to image the specimen down the b-axis.

Number of slices per unit cell

(default=1)

9

This value will be computed by MacTempas from the repeat distance of the structure in the beam direction and the current value of G_{max} . This number can be changed if desired (as, of course, can all the parameters entered in response to the prompts listed in this chapter).

Show (Basis)

Click on the command to bring up the dialog box for entering the information regarding the number of atoms in the basis. We enter the nine different atom positions listed for the basis atoms. For each of the atoms in the basis, MacTempas requires the chemical symbol, x,y,z coordinates, DW factor and occupancy factor. From the information given above, we use the following information for the nine atoms that are given in the structural basis.

Chemical Symbol	Ca
X, y , Z	0,0,0
Debye-Waller Factor	3.6
Occupancy	1

The data for the first atom include the chemical symbol for calcium (used by MacTempas to select the correct scattering factor table), the atom coordinates, the temperature factor (or Debye-Waller factor), and the occupancy factor.

The second atom position is entered in the same way with responses of -

Chemical Symbol	Sr
x,y,z	0,0,0.1097
Debye-Waller Factor	3.6
Occupancy	1

The third atom position is similar, except that the occupancy is set at 0.87 -

Chemical Symbol:	Bi
х,у,Z:	0,0,0.3022

B Factor:	3.6
Occupancy:	0.87

After all nine atom positions have been entered, MacTempas will need the parameters of the electron microscope for which to compute the simulation.

	Ca. Sr	D. DODODO	D.DODDDD	D. COCOOD	3 400000	1 ababaa
	Sr			an and and and at	2000000	1.000000
		D.DODODO	D DO DO DO D	D 10970D	3.600000	1.000000
	Bi .	0.0000000	0.000000	0.302200	3.600000	0.870000
	8i	0.0000000	0.000000	0.269100	3.600000	0.130000
	Cu	0.0000000	0.000000	0.445600	3.600000	1.000000
	0	0.500000	0.000000	D.446000	3.600000	1.000000
	0	D.DGDGDO	D.DODDOD	0.375000	3.600000	1.000000
	0	D.DODODO	D.DODDOD	0.205000	3.600000	1.000000
	0	0.500000	0.000000	0.250000	3.600000	0.065000
				1		
1					-	

Microscope

4000EX

If the input microscope name is listed in MacTempas's microscope file, various operating parameters will be set automatically. If the entered name is unknown to MacTempas, values will need to be given for each of the operating parameters. In this example, we use 4000EX, and find that MacTempas sets the spherical aberration coefficient to 1.0mm, the Gaussian halfwidth of depth of focus to 80Å, and the semi-angle of beam convergence to 0.5milliradian.

Specimen Thickness 40 20 80 The foil thickness response may be in one of two forms, either a single value in Ångstrøm units, or a construction combining a starting and ending thickness with an incremental value. The

construct that we have entered requests MacTempas to store diffraction results for thicknesses starting at 40Å and continuing through 80Å in steps of 20Å. That is, at specimen thicknesses of 40Å, 60Å and 80Å.

Store Ampl./Phases

As well as storing all the beam amplitudes at specified specimen thicknesses, MacTempas can store a selected few beam amplitudes at each single-slice increment in thickness, then plot amplitude (or intensity) and phase as a function of thickness for any of the stored beams. To store beams for plotting, click on the command to enter the indices for the reflections that will be stored. In this starting example we will not be entering any information here.

Voltage

The voltage would need to be entered if an unknown microscope type were selected. Since we have selected a 4000ex, MacTempas will choose a value of 400keV.

Center of the Laue Circle

The pair of values specified as the Laue circle center are used by MacTempas to define the direction and amount by which the specimen is tilted from the exact zone-axis orientation specified above, and, in fact, specify the center of the Laue circle in units of the h and k coordinates in the diffraction plane. Note that the values supplied need not be integers, but should not define a tilt of more than a few degrees. The default values of 0,0 specify exact zone-axis orientation.

Objective Lens Defocus

-200 -200 -800

So far, we have supplied all the information MacTempas requires to carry out the dynamical diffraction part of the simulation; now we input the imaging conditions. The first imagingcondition prompt is for the objective lens defocus. We choose to enter four values of defocus by specifying defocus values from -200Å to -800Å in steps of -200Å. Note that a negative value denotes an objective lens weakened from the Gaussian condi-

No

(400) ·

0.0

tion; that is, underfocus is negative.

	Aperture Radius The value for the radius of the objective aperture shou spond to the radius in reciprocal Ångstrøm units, as m from a diffraction pattern exposed with the aperture su posed. We will enter 0.67 to represent a typical value.	0.67 ld corre- easured iperim-
	Center of the Objective Aperture In order to simulate dark-field images, MacTempas pro an objective aperture displaced from the center of the tion pattern. As for the Laue circle center, the aperture defined in units of h and k. We leave the default value	0,0 ovides for diffrac- center is as of 0,0.
	Center of the Optic Axis To provide for microscope misalignment, or for condit tilted-beam imaging, the coordinates of the diffraction which the optic axis lies can be specified in the same n the center of the aperture. Again, we use default value	0,0 tions of pattern at nanner as es of 0,0.
Verifying the Input	After the response to the last data-entry prompt, MacT draws the windows it uses. To re-display the input infe- click on the "Main Parameters" in the Parameters me this stage any desired changes can be made by using th to move the cursor to the desired parameter, and makin change.	'empas ormation nu. At ne mouse ng the
Running the Cal- culation	When all the data in the top field are satisfactory, we g "Atomic Basis" in the parameters menu to check that parameters have been entered correctly. At this stage is worthwhile getting MacTempas to display a model of ture by.going to the Commands menu and clicking on the Unit Cell".	o to all atom it is also the struc- "Draw
	When we are satisfied that all data are correct, we run	the simu-

Ch. 8 Sample Calculation - p.125

	lation by.clicking on "Full Calculation" in the calculate menu. Note that MacTempas displays the current status of the calcula- tion in the Status Window. First, MacTempas computes the phase-grating for the structure (the status window shows the number of coefficients generated so far), then the dynamical diffraction for each slice of the specimen (current slice number is shown in the Status Window), then four images are computed at each of the three specimen thicknesses that we specified (the image number is shown in the window).
Displaying the Results	Once MacTempas has finished the computation, the results (dif- fraction patterns, images and diffractograms) can be displayed. (Also beam amplitude and phase plots if any of these has been stored).
	To display the images, we.go to the Display Window and select "IMAGE", then "DISPLAY". MacTempas will ask which of the 12 images is to be displayed, then display the requested image in the center of the screen. The image can be moved around with the pointer tool
	To get all 12 images onto the display screen simultaneously, select the options menu and the "Montage" option. Back in the source window, set "ZOOM" to 0.5 (to reduce the image magnification in order to fit all 12 images on the screen), then "DIS-PLAY".
	Now go back to the montage option and deselect "Montage".
	To display the projected potential for comparison with images, select "PROJ.POT" in the source window, then "DISPLAY".
	To display the diffraction patterns at the stored specimen thick- nesses, select "DIFFR.PATT" in the source window, then "DIS- PLAY". To change the size of the patterns, choose "Diffr. Patt". from the Options Menu and choose a different camera length.

The size of the diffraction spots also depend on the divergence angle set in the main parameters. It may be necessary to adjust both the camera length and the divergence angle to get a suitable display of the diffraction pattern.

To display the power spectrum of one of the images, we choose "IMAGE" from the **source** window. Respond by answering which image and then choose "FFT" from the **operand** window. Finally click on "DISPLAY" to view the power spectrum. The options for the power spectrum are the same as those for display of diffraction patterns. The circle drawn in diffraction patterns and power spectra corresponds to the objective aperture and can be turned off from the diffraction option. **MacTempas User Manual**

Ch. 8 Sample Calculation - p.128

Chapter 9

The Weak Phase Object Approximation

The Weak Phase Object (WPO) approximation is a useful tool to find out what kind of information about a specific structure may be revealed at different levels of resolution. The WPO approximation has already been described earlier, and some of that information is repeated here. There are two important assumptions that are made in the WPO approximation.

Wavefunction Approximations

The wavefunction of the electron can be written as

(x, y) = 1 - i t (x, y)

where (x, y) is the electron wavefunction at a point (x,y) and (x, y) is the projected electrostatic potential at the same point. Sigma is the interaction parameter between the electron and the potential of the atoms and t is the specimen thickness. This first approximation is good for very thin specimens containing light atoms.

Ideal ScherzerAn ideal Scherzer lens is a lens that transfers all diffracted
beams with a g-vector that is less or equal to 1/resolution, and
blocks all diffracted beams with a larger g-vector. In addition it
adds a phaseshift of 90 degrees (relative to the central beam) to
all beams passing through the lens. This in addition to the 90
degree phaseshift introduced by the scattering event itself (the
'i' in the equation for (x, y) above) causes all scattered
beams that pass through the lens to be 180 degrees out of phase

with the central beam.

Under the two assumptions above, the image intensity in the WPO approximation can be written as

$$(x, y) = 1 - i t (x, y)$$

such that the image intensity is low in areas of high electrostatic potential, the location of atoms. Atoms of higher atomic number show up as larger and darker regions in the image. This type of image will often be similar in appearance to images calculated by a full multislice calculation for equivalent resolution for a thin specimen for Scherzer defocus.

The WPO approximation is invoked from the menu bar in the same fashion as the multislice calculation. The input to the WPO calculation is a starting resolution in Å and an ending resolution. The steps in resolution can be fixed (user determined) or automatic. When automatic steps are chosen, the program will calculate the first image corresponding to the reflections that lie within 1/BeginningResolution and each new image will be calculated for the next set of reflections corresponding to a higher resolution until the end resolution is

lesolution Options		
tarting Resolution (Å)	3.00	
nding Resolution (Å)	1.00	
Auto Decrement Resolu	ution	
Fixed Decrements in st	eps of 0.2	
iewing Options		
lumber of Unit Cells to di	splay X & Y	1 1
oom Factor to be Used in	Displaying Ima	ipes 2.0

MacTempas User Manual

Ch. 9 The Weak Phase Object Approximation - p.132

Chapter 10

Creating a Layered Structure

A layered Structure is a special type of "structure" where the composition varies in the direction of the electron beam. An example of this would be a crystalline material having surface layers of amorphous material. Another example would be a crystalline structure where the repeat distance in the electron beam direction is too large for the repeat to be used as the slicethickness and the unit cell must be sub-divided into several



Ch. 10 Creating a Layered Structure - p.133

slices with different atomic content. As an example we will work with three layers which we will call LayA, LayB and LayC. Each of these "layers" are what we would call a "single" structure. That means they are defined as a unit cell with lattice parameters and atomic content. The one thing they have in common is that the lattice parameters A and B with respect to the electron beam are the same and that we will use identical sampling in each case, see figure above. The idea of the layered structure is that the 3 layers can be arranged in any chosen sequence to make up the total structure. The steps in creating and calculating the image for a "**layered**" structure are as follows.

1) Define the 3 layers LayA, LayB and LayC as single structures with the same unit cell dimensions perpendicular to the electron beam (A and B).

- Calculate the phasegrating for each structure LayA, LayB and LayC using the same value for Gmax.
- 3) Now create a "**New**" Structure in MacTempas using the

Specimer	n Parame	ters	107	-	21
Zone-axis (Total define Gmax (Å-1) Thick. (beg,	uvw(t)] ad thickno inc.end)	0 Aşsş f A	0 1 20	1	0 2.00 20
Store Ampl.	/Phases	0	Se	t	NU
Cent.of Lau	e Gircle	h	0.00	I K	0.00
Eq. Tilt [mra	id] & angl	e"	-0.0	1	0.0
Type of Abs	orption	18 E.	N	one	
-600 Me	o fold ee fold echanical jma of (a,	0 0 Vibre b)	tion		
8.0 An	gle with	x-axis	(Y)	0.0	1.1
and the second se	forgetters integrable submitted	statistical statistics	and the second second	and the states	
	Specimes Zone- axis (Total define Gmax (Å- 1) Thick. (beg, Store Ampl. Cent. of Lau Eq. Tilt (mrd Type of Abs Type of Abs Tw Thr -600 MR Angle* Sig 1.0 An	Specimen variance Zone- axis (uvw(t)) Total defined thickne Gmax (Å- 1) Thick. (beg.inc.end) Store Ampl./Thoses Cent. of Lave Circle Eq. Tilt (mrod) & angl Type of Absorption - Astigmati 0 Two fold Three fold -600 Angle* 1.0	Specimen varianeters Zone-axis (uvw(t)) 0 Total defined thickness (Å Gmax (Å-1) Thick. (beg,inc,end) 5 Store Ampl./Thoses 5 Cent. of Lave Circle h Eq. Tilt (mrod) & angle* Type of Absorption 5 - Astigmatism (Å 0 Mag. Two fold 0 Three fold 0 - Mechanical Vibra Sigma of (a,b) Angle* 4 0.0	Specimen rarameters Zone-axis [uvw(t)] 0 Total defined thickness [Å] Gmax [Å-1] Thick. (beg,inc,end) 20 Store Ampl./Thases Se Cent. of Laue Circle h Eq. Tilt [mrad] & angle* -0.0 Type of Absorption N Mag. V Two fold 0 Three fold 0 Angle* Sigma of (a,b) 0.00 Angle with x-axis (?) Angle with x-axis (?) 0.00	Specimien Parameters Zone-axis [uvw(t)] 0 1 Total defined thickness [Å] Gmax [Å-1] Thick. (beg.inc.and) 20 0 Store Ampl./Phases Set Cent. of Lave Circle h 0.00 Eq. Tilt [mrad] & angle* -0.0 Type of Absorption None Astigmatism [Å] Angle Mag. w/ho Two fold 0 Three fold 0 Angle* Sigma of (a,b) 0.00 0.0 Angle with x-axis (?) 0.0

option "Layered Structure". You will be asked to fill out infor-

mation regarding the lattice parameters A and B etc. There are no input for atoms, because a layered structure has no atom information per se. Even though you are asked to fill out a specimen thickness, this value has no meaning at this time, because the content of the structure has not been defined. The values of A and B come from the structures LayA, LayB and

12	copper inp	dz = 3.61Å dz = 4.50Å	=	
	New	Delete	Show	
		Connet		

Ch. 10 Creating a Layered Structure - p.135

LayC. When you create the layered structure, a default value of 2 $Å^{-1}$ is supplied and you must change it in the main parameters if a different value was used in calculating the phase-gratings for LayA, LayB and LayC.

4) Once the information in 3) has been filled out, the file is created and you must define the "structural" or "phasegrating" content of the layered structure. This is done by going to the Command Menu and executing the command "**Stack Phaseg-ratings**". If this is a new file, there will be no phasegratings listed and the command "**New**" must be used to define the layers. By invoking "**New**", you get a list of the available phaseg-

	MacTempasPPC - Open	
😽 File Edit View	Folders Documents Finder	t
C Ext	mple Structures 💠	a Olodum
Rate	Sier Lute	a Eject
cabbe, bent	40 E Sun, Jan 29, 199	Desktop
El inp.pout	16 K - Wed, Hor 6, 199	
		Cancel
		Open
New Jak		*
3 3005 4		8 60°.

rating files (—.**pout**). Double Click on LayA.pout and fill in the value for the slice-thickness that was used in the calculation of LayA.pout. Continue and do the same for LayB and LayC.

Name of this layer	Thickness (Å)
copper	3.6147
Cancel	ОК

Now the program has information as to which phase-gratings it can use and the final part is to define the sequence of these phasegratings up to the desired thickness. Use "**Stack**" and the sequence can be defined in different ways. One way is to type in the sequence as

1,1,1,1,1,2,2,2,2,2,2,3,3,3,3,1,1,1 where 1 stands for LayA, 2 for LayB and 3 for LayC. One can also use the commands to define the sequence. At all times the specimen is drawn as a colored bar at the left. Once this is done, you have defined the structure.

# 1 2	Name copper isp	- Defined Δ <i>x</i> 3.61 4.50	Phase ¢	egratings Name	Δz	•	Name	Δz	•	Name	Δz
	TOP	— Speci 1,1,1	men (,1,1,2	2,2,2,2,1,1,	1.1					De In Re	posit sort peat eset
	55.0	0									ок

5)Now check the Main Parameters to see that everything is correct and finally run the calculation. The calculation will begin with multislice.

Ch. 10 Creating a Layered Structure - p.137

MacTempas User Manual

Ch. 10 Creating a Layered Structure - p.138

Chapter 11

HOLZ Interactions & Sub-slicing

With suitable algorithms, it is possible to include in the diffraction calculation the effects of out-of-zone scatterings, or nonzero (or higher-order) Laue zone (HOLZ) interactions. Basically, there are four ways to produce the set of phasegratings (or projected potentials) that describe the "multisliced" crystal. For structures with short repeat distances in the beam direction, the simplest method is to use one slice per unit cell. For structures with large repeats in the beam direction, several methods may be used, three of which rely on sub-dividing the slice into "subslices". Any of the four methods can be used in MacTempas.

Identical slices with only one sub-slice per unit cell repeat distance

A multislice computation in which every slice is identical contains no information about the variation in structure along the incident beam direction, and includes scattering interactions with only the zero-order Laue zone (ZOLZ) layers. For structures with short repeat distances in the beam direction such a computation is adequate, since the Ewald sphere will not approach the (relatively distant) high-order zones.

Identical sub-slices with n sub-slices per unit cell repeat distance

For structures with large repeats in the beam direction, a method of sub-dividing the slice is required in order to compute the electron scattering with sufficient accuracy. The simplest, but most approximate method, is to compute the projected

Ch. 11 HOLZ Interactions & Sub-slicing - p.139

potential for the full repeat period then use 1/n of the projected potential to form a phase-grating function that can be applied n times to complete the slice. This method avoids interaction with any "pseudo-upper-layer-line" (Goodman and Moodie, 1974), but ignores real HOLZ layers.

Sub-slices based on atom positions

An improvement on sub-dividing the projected potential is to sub-divide the unit cell atom positions. In this procedure the list of atom positions within the unit cell is divided into n groups depending upon the atom position in the incident beam direction. From these sub-sliced groups, different projected potentials are produced to form n different phase-gratings, which are applied successively to produce the scattering from the full slice.

Sub-slices based on the three-dimensional potential

A further improvement on sub-dividing the atom positions, is to sub-divide the three-dimensional potential of the full slice, since an atom with a position within one sub-slice can have a potential field that extends into the next sub-slice. Rather than compute a full three-dimensional potential and then integrate over appropriate sub-slices (a 128x128x128 potential would require over two million samples to be stored), it is possible to derive an analytical expression for the potential within the subslice $z_0 \pm z$ projected onto the plane at z_0 (Self et al., 1983). It is possible to apply this method routinely to structures with large repeats in the beam direction, thus generating several different phase-gratings for successive application, and even to structures (perhaps with defects) that are aperiodic in the beam direction and require a large number of individual non-repeating phase-gratings (Kilaas et al., 1987).

MacTempas sub-slicing

While ensuring that the calculation remains sufficiently accurate, MacTempas will normally choose the simplest (and quickest) method of specifying how slices are defined for any particular combination of specimen, zone axis, accelerating voltage, and maximum g. To this end, the user can choose to neglect HOLZ interactions if these are judged to be unimportant. If HOLZ interactions are important, then the user should select the "3D-Potential Calculation" radiobutton in the Options menu, rather than "2D-Potential Calculation".



When a two-dimensional calculation is selected, MacTempas will use one slice per cell if the cell repeat distance in the beam direction is small. If the repeat distance is too large for one slice per unit cell, MacTempas will avoid pseudo-upper-layerlines by producing n identical sub-slices.

When a three-dimensional calculation is selected, (3D-Potential Calculation activated), MacTempas uses a sub-divided threedimensional potential when the repeat distance is large, and defaults to one slice per cell if the distance is small enough. Note that the number of sub-slices per unit cell can be forced to be greater than one by setting it explicitly in the Parameter menu; this will ensure that any HOLZ interactions are included even for small repeat distances. Of course, if the repeat distance is very small, leading to a distant HOLZ in reciprocal space, both the calculation and the experiment it is modeling will interact only very weakly with the HOLZ reflections. Use of the Layered Structure option to produce the scattering from a structure that is layered or aperiodic in the incident beam direction is effectively an application of the method of sub-slicing based on atom positions. Thus the user could create a number of sub-slices by assigning selected atoms to different structure files, then forming a phasegrating for each sub-slice, and using the Stack Phasegratings command to specify how the sub-slices are to be used to describe the specimen structure. This is the suggested method to try first if upper Laue layers are to be included or 3-dimensional effects are important as it is much faster than using a complete 3D calculation.

Other methods

Van Dyck has proposed other methods to include the effects of HOLZ layers, including the second-order multislice with potential eccentricity (Van Dyck, 1980) and the improved phase-grating method (Van Dyck, 1983). Tests of these procedures show that the extra computation involved in using potential eccentricity may be worthwhile, but that the improved phase-grating method diverges too easily to be useful.

Goodman P, Moodie AF (1974) Numerical evaluation of Nbeam wave functions in electron scattering by the multislice method. Acta Cryst. A30, 322-324.

Kilaas R, O'Keefe MA, Krishnan KM (1987) On the inclusion of upper Laue layers in computational methods in high resolution transmission electron microscopy. Ultramicroscopy 21, 47-62.

Self PG, O'Keefe MA, Buseck PR, Spargo AEC (1983) Practical computation of amplitudes and phases in electron diffraction. Ultramicroscopy 11, 35-52.

Van Dyck D (1980) Fast computational procedures for the sim-

ulation of structure images in complex or disordered crystals: A new approach. J. Microscopy 119, 141-152.

Van Dyck D (1983) High-speed computation techniques for the simulation of high resolution electron micrographs. J. Microscopy 132, 31-42.

MacTempas User Manual

Ch. 11 HOLZ Interactions & Sub-slicing - p.144
Chapter 12

Structure Refinement Through Matching of Experimental and Simulated HRTEM Images

12.1.Introduction

The goal of performing simulation of HRTEM images is to compare these with the experimental data in order to determine the structure. In practice this means that various models are proposed and that images are calculated until a match is found. At that point, the structure is presumed to be known (atomic positions and atomic numbers) with some given uncertainty. Alternatively, one starts with a given model and varies the model in a systematic fashion searching for a global maximum in the fit between experiment and simulation. This entails that one needs an efficient method to compare the experimental and the calculated image. It also requires knowledge of the uncertainty in the measurement (image intensities in the experimental image) and a way to relate this uncertainty to the uncertainty in chemical composition and atomic positions. This area of quantitative electron microscopy is fairly new and most images are still compared visually. However, it is an active area of research and many techniques from statistics are just now beginning to be used in HRTEM.

12.2. Acquiring quantitative data

In order to extract quantitative information from electron micrographs, the data must be represented by a set of numbers. Usually, images from TEMs are brought into a digital representation by one of the following methods.

i)Recording the image on a photographic plate and using a scanner to convert the film density into numbers which are stored in a computer.

ii)Recording the image on an image plate.

iii)Recording the image on a slow scan CCD camera with readout of deposited charge into a computer.

The first approach yields data that is not directly comparable to computer calculations because of the non-linear response of the film. It is however possible to calibrate the response of the film based upon a sequence of controlled exposures using varying exposure times and mapping the resulting scan values versus electron dosage[1]. The image plate and the CCD camera both yield numbers that are linear with respect to the electron dosage and only require a scaling of the data in order to compare to computed values[2]. There has been much discussion about the relative merits of the various recording media above and each has its own advantage. The CCD camera is currently limited to 2K by 2K pixels, although it may be possible to go to 6K by 6K by using multiple chips in the near future. Since its Modulation Transfer Function (MTF) can be characterized, it is straight forward to use deconvolution to compensate for the drop in high frequency response due to spread of electrons and due to spill over of charge to neighboring cells[3]. The image plate has many of the advantages of the CCD camera and covers a larger image area. However, the imaging plate is not gaining as much popularity as the CCD camera. Many laboratories are now starting to do much of the recording on CCD cameras, while still retaining the use of film.

12.3.Pre-processing of data

Once the image data has been converted to numbers, any necessary processing or transformation of the data can take place. The required pre-processing of the data depends on the nature of the information that is sought and thus there is no one optimal method, but rather a number of possible options.

12.3.1Sampling and resampling of data

If the image is distorted over the image field of view, either by the action of the imaging system or the recording system, the data can be re-transformed by a warping transformation of the image. This can be done if the transformation can be determined by imaging a perfectly crystalline material and noting deviations from where the atoms are known to be and where they are imaged[4]. On some systems, i.e. the Gatan Imaging Filter, the distortions are measured by recording the image of a square grid of circular holes.

An image of a crystalline material can be resampled onto lattice relative coordinates, such that the unit cell dimensions are represented by an integer number of pixels and commensurate with the dimensions of the final image. This will eliminate streaking in the Fourier transform of the image which is due to the truncation of the image by the edges on boundaries that do not represent a periodic continuation of the image. Streaking can be reduced by multiplying the image with a circular mask. The mask is represented by a circle of pixels with value 1 up to a specified radius and then falling off gradually to 0 within 5 to 10 pixels close to the borders of the image. A side-effect of masking is the increase in noise in the Fourier transform which is discussed below.

If only the image of a single unit cell of the crystalline material need to be determined and compared with the image obtained through an image simulation calculation, the image of the unit cell can be resampled onto the coordinate system and sampling interval used in the computation. This is equivalent to determining the matrix M defined through the equations

 $\mathbf{a}_{s} = M \mathbf{a}_{e}$

1)

Ch. 12 Structure Refinement Through Matching of Ex-

$\mathbf{b}_{s} = M \mathbf{b}_{e}$

Two steps are necessary; the rotation /scaling required to make the lattice base vectors identical; and secondly the determination of a common origin.

Finding the common origin between experimental and calculated image is determined by cross-correlation between the simulated and the experimental image[5].

12.3.2Fourier transforms and masking

The Fourier transform of HRTEM images of crystalline materials provides useful information about lattice spacings and can also be used to compare experimental Fourier amplitudes with theoretical calculations. Because the image being transformed is rarely a periodic function in W (width) and H (height), the Fourier transform of the image of a pure crystalline material is the convolution of the Fourier transform of a perfectly periodic signal (the crystal) with the transform of a window the size of the image dimensions, making a Bragg peak take on the shape of the transform of the window.

name	window	function	transform sin(πk)	peak profile	falloff 1	rel. noise level
none	$\frac{1\pi}{2}\cos\frac{\pi x}{2}$		$\frac{\cos(\pi k)}{1-\frac{1}{2}}$	\square	$\frac{\overline{k^1}}{1}$	1
cosine			1 - 4k $\sin(\pi k)$		к 1	1.23
Von	$1 + \cos(\pi x)$		$\pi k(1-k^2)$		$\overline{k^3}$	1.5
Hann						

Table 1:

The use of a mask changes the transform of the window and can be used to make the peak profiles decay faster, but at the expense of increasing the noise level. This is illustrated above,

showing the effects of employing masks on a 1-dimensional signal[6]. This also has an effect on locating peak positions in order to determine lattice spacings and on the estimate of the amplitude of the Fourier component. The standard error in both estimates increases as a function of applying a mask, with the cosine window being a good compromise.

12.3.3Noise reduction

In addition to reducing noise, it is also important to have an estimate for amount of noise present and to quote a signal to noise ratio. From two equivalent regions, the noise can be estimated from obtaining the cross-correlation coefficient for two regions. Given a cross-correlation coefficient ccf, the signal to noise ratio can be estimated as

$$S_N = \frac{CCF}{1 - CCF}$$
 2)

In order to reduce noise and to obtain a statistical average of the image of a single unit cell (motif), the positions of individual motifs can be determined by cross-correlation. Once these are found, statistically equivalent regions can be averaged to find the average motif and to determine the signal to noise ratio associated with individual pixels as a function of position within the unit cell. This determines a standard deviation for each pixel i and can be used to set confidence levels associated with matching of the experimentally averaged image with a calculated image[7].

$$\sigma^{2}(i) = \frac{1}{M-1} \int_{j=1}^{M} (I_{j}^{2}(i) - \langle I(i) \rangle^{2}) \quad 3)$$

where M is the number of equivalent regions being averaged. Using a low pass filter to perform a smoothing of the image may be effective depending on the noise level present, particularly when averaging over statistically equivalent regions can not be performed. Smoothing helps the eye see features more clearly; but has the disadvantage that it causes correlation between image pixels, which may distort the significance threshold of simulation mismatch criteria. Averaging can also be performed through symmetrization which is to average the motif with copies of itself to which symmetry operations known to be present are performed. This will reduce noise levels by a further factor of $1/\sqrt{M}$ when M symmetry related copies are averaged, but may also just disguise defects in imaging conditions.

12.4. Matching experimental and simulated images

There are a number of various ways to measure similarity or mismatch between two images. Below are a few of these[8]. The mean square difference:

$$D^{2} = \left\langle (I_{1} - I_{2})^{2} \right\rangle = \frac{1}{N} \quad (I_{1} - I_{2})^{2} \quad 4)$$

The Root Mean Square Difference:

$$D_{rms} = \sqrt{D^2} \quad 5)$$

The mean modulus difference:

$$D_{mmd} = \langle |I_1 - I_2| \rangle = \frac{1}{N} |I_1 - I_2| = 6$$

The Cross-correlation Coefficient:

$$CCF = \frac{(I_1(i) - I_1) (I_2(i) - I_2)}{\sqrt{(I_1(i) - \bar{I}_1)^2 (I_2(i) - \bar{I}_2)^2}}$$
7)

The brackets <> all indicate the mean of the enclosed quantity. In each of these equations, the sum is over all the pixels i in the image and N is the total number of pixels. The cross-correlation coefficient above is a normalized coefficient where the images are normalized to zero mean.

The CCF (which measures similarity rather than difference) can also be interpreted as the cross-product between two n-dimensional vectors (n being the number of pixels in the image). In that case, one can associate an angle with the CCF,

 $CCF = \cos \vartheta$, in the general interpretation of an inner product between two vectors as $I_1 \cdot I_2 = |I_1| |I_2| \cos \vartheta$ with the angle

being $\vartheta = \cos^{-1}(CCF)$. This angle is zero for identical images. If the images are normalized to zero mean and unit length as in the definition of the normalized cross-correlation coefficient above, the angle is 180 deg. for a reversal in contrast between the two images I₁ and I₂.

12.4.1Significance and Noise

Each of the above criteria must be tested for the significance of the measured value.

 D^2 can be compared to the mean square intensity (or intensity deviation due to noise) in either image.

 D_{rms} can be compared with standard deviation of the intensity in either image

A good way to test for the mismatch between two images, is to use a statistical measure for the probability of two images being equal given knowledge of the noise in the images. If one assumes Gaussian uncorrelated noise for each pixel in the experimental image, the optimum statistical measure is given by

$$\chi^2 = \frac{1}{N} - \frac{(I_1(i) - I_2(i))^2}{\sigma^2(i)} = 8$$

where N is the number of pixels in the image[9]. The value $\sigma^2(i)$ is the standard deviation associated with the pixel *i* and can be found as described above from a number of equivalent regions. If an experimental image I_e is compared to a calculated image I_c and there are M adjustable parameters in the calculation, the equivalent expression becomes[7]

$$\chi^2 = \frac{1}{(N-M)} - \frac{(I_e(i) - I_c(i))^2}{\sigma_e^2(i)}$$
 9)

A mismatch by one standard deviation adds one to the sum in the expressions above and a value of 2 of 1 implies that the two images are identical within the uncertainty given by the noise. The expected value for statistically equivalent images consisting of N points is 1 and random deviations from this value by more than $2/\sqrt{N}$ are considered unlikely.

Ch. 12 Structure Refinement Through Matching of Ex-

Writing

$$\chi^2 = \frac{1}{N} - \frac{\left(I_1(i) - I_2(i)\right)^2}{\sigma^2(i)} = \frac{1}{N} - f^2(i) - 10$$

leads to the definition of a Residual Image f(i) [10] which is used to visualize and to quantify the (mis)match between two images. It has the advantage that instead of presenting a single number for how well two images match, it is a two-dimensional mapping of the local fit. Thus a difference image will more clearly reveal areas of greater mismatch. The optimum match is still defined by minimizing 2 . It is important to note that the fitting parameters can also be applied to the Fourier transforms of the images which sometimes will lead to a reduction in the number of the data-points to be compared[11]. In the case of images of crystalline material containing no defects, the Fourier components will be non-zero only for frequencies corresponding to Bragg-reflections of the lattice, although this is strictly only true if the motif has been averaged over many repeating regions and resampled onto lattice coordinates such that streaking due to discontinuities at the boundaries is eliminated. The complex values for the Fourier coefficients take the place of the image intensities. It is interesting to note that the use of different matching criteria can lead to slightly different values for optimized parameters[12].

12.4.2 Adjusting for different means and contrast levels

Since absolute values for image intensities are not known and an experimental image may be linearly related to a calculated image, a useful way of normalizing the image intensities is to subtract the mean and divide by the standard deviation. This ensures that $D^2 = 0$ for linearly related images and a value of around 2 for unrelated data.

Similarly, the Cross-correlation coefficient will lie in the range from -1 to 1, taking the extreme values when the two images are linearly related and being near 0 for unrelated data.

Another approach is to scale the images to the same mean. This is done as follows

$$I_{calc} = \frac{I_{calc}}{\langle I_{calc} \rangle} \langle I_{exp} \rangle \quad 11)$$

where the calculated image is scaled to the mean of the experimental image.

In order to understand how the mean value, contrast and image pattern affect the image matching criteria, it is useful to consider how the Root Mean Squared Difference D_{rms} can be separated into three terms[13].

$$D_{rm} = \sqrt{\langle (I_1 - I_2)^2 \rangle} = \langle \langle I_1 \rangle - \langle I_2 \rangle \rangle^2 + [\sigma_1 - \sigma_2]^2 + 2(1 - \rho)\sigma_1\sigma_2$$

where

$$\sigma_{1,2} = \sqrt{\langle I_{1,2}^2 \rangle - \langle I_{1,2} \rangle^2} \quad 13)$$

and

$$\rho = \frac{\langle I_1 I_2 \rangle - \langle I_1 \rangle \langle I_2 \rangle}{\sigma_1 \sigma_2} \quad 14)$$

The first term measures the difference in the mean of the two images and vanishes if both images are normalized to the same mean value. The second term measures the difference in contrast between the two images, while the third term (where is the same as the normalized cross correlation coefficient) measures the difference (similarity) in the pattern of the two images. Thus it is important to note that the normalized cross correlation coefficient CCF only measures similarity in patterns and ignores variation in contrast and differences in mean levels. It is generally found that most of the mismatch between experimental and computer simulated images is due to the difference in contrast[14]. The difference in contrast can be an order of magnitude and the cause is generally attributed to the following factors.

misalignment specimen vibration inelastic scattering

specimen damage

There is however an ongoing debate as to the nature of the discrepancy in contrast as calculations indicate that the factors above are not sufficient to resolve the disparity. A possible explanation is that there is a general background in experimental images that is not accounted for.

12.4.3 Effect of noise on matching criteria

In order for two images to be considered equal, we need to consider the effect of the uncertainty or error in the matching criteria due to noise and the parameters determining the image. A study of the effect of noise on the cross-correlation factor reveals that in the presence of noise, the cross-correlation coefficient *CCF* for the two images I₁ and I₂+ , where represent random noise superimposed on image I₂, can be written as[13]

$$CCF(I_1, I_2, \eta) = CCF(I_1, I_2) / \sqrt{1 + \frac{\sigma^2(\eta)}{\sigma^2(I_2)}} = CCF(I_1, I_2) / \sqrt{1 + \vartheta_n^2}$$
15)

with

$$\vartheta_{\eta}^2 = \frac{\sigma^2(\eta)}{\sigma^2(I)}$$
 16)

The effect on the hyper-angle $\vartheta = \cos^{-1}(CCF)$ is in the small angle approximation

$$\vartheta(I_1 I_2, \eta) = \sqrt{\vartheta^2(I_1 . I_2) + \vartheta_{\eta}^2} = 17)$$

If two images are identical except for a small error in one of the image-formation parameters (defocus, thickness, etc.) the error in the angle ϑ is proportional to the parameter error. The error in the angle due to independent parameter errors is

$$\theta = \sqrt{\vartheta_i^2} \qquad 18)$$

Typical mismatches in CCF (pattern matching) due to parameter errors are

Parametet	Error	theta(mrad)
Noise		0.06
Composition	±0.03	0.02
Thickness	±2nm	0.2
Defocus	±15nm	0.4
Beam Tilt	<1.5mrad	0.8
Astigmatism	<15nm	0.2
Crystal Tilt	<2mrad	0.6
Beam Diverg.	<0.3mrad	0.1
Focal Spread	<5nm	0.15
Vibration	<0.04nm	0.2

Table 2:

12.4.4Chi-Square or Chi-based criteria

Although all the methods above measure either the match or mismatch between two images, the important questions is not to what degree do they match, but how well do they match given systematic and non-systematic errors. Thus the fitting parameter must take into account the statistical nature of the data and the accuracy to which we know the data-points. Thus the fitting parameter should depend on a maximum-likelihood (probability) model and be a measure of the probability that A is equal to B, given knowledge of the probability distribution of the datapoints. In the presence of Gaussian distribution of uncorrelated noise, each data point has a Gaussian probability distribution with the noise in one pixel uncorrelated to the noise in adjacent pixels, which leads to a ² criteria. The criteria takes into account the number of adjustable parameters and the error in each data point.

As mentioned above, any data point lying one sigma away from the expected value will add 1 to the sum in 2 .

Similarly, any data point which has only 1% probability of being measured given A = B, adds a value of 6.63 to the sum in

². Thus values of ² greater than about 6 states that there is less than 1% probability that A is equal to B. The fitting parameter depends on the model of the distribution of data-points due to statistical noise with a Gaussian distribution of uncorrelated noise leading to the criteria. However, it is important to determine the statistical nature of the noise in the image. This can be done by examining the noise distribution determined from a large number of image regions considered to be equivalent except for noise. A non-Gaussian distribution will lead to a modified criteria, but still based upon [7].

12.5. Structure determination

In order to determine the "unknown" structure, it is necessary to perform a comparison between calculated images, exit wavefunctions or diffraction patterns and experimentally obtained data. As described above the comparison can be done using different matching/mismatching criteria. Ideally, the determination of the structure is done by modifying the structure until the mismatch between the experimental and calculated data is within the error in the experimental data. In principle the imaging parameters themselves can be allowed to vary together with the atomic coordinates. However, in practice the imaging parameters are optimized separately if possible. This reduces the complexity of the problem and reduces the number of steps involved in the search for a solution which optimizes the matching criteria. In cases involving unknown defects in the presence of a "known" structure, the imaging parameters and specimen thickness are first determined from the known structure. Determination of an unknown set of input parameters requires the following 1)An image (in real or reciprocal space) obtained from the experimental data.

2)A computational method yielding an image to be compared to the 1).

3)A method for comparing 1) and 2)

4)A criteria based upon 3) for when 1) and 2) are statistically equivalent.

5)An initial set of adjustable input parameters which are to be optimized so that the final configuration results in satisfying 4)6)A method for varying the adjustable parameters so that the final configuration is found within finite time.

There is an essential assumption being made above, which states that the computational method used in 2) will produce the image in 1) given the correct choice of input parameters. This is a separate issue which will not be addressed here. The validity of this assumption can be debated and it is acknowledged that computational methods are in need of further refinement. However, in what follows, the assumption is presumed to be valid.

12.5.1Matching Images or Exit Wavefunctions

In order to compare calculation with experiment, one can compare either images or diffraction patterns. For perfect structures it may be beneficial to compare diffraction patterns since the number of data points to compare are given by the possible Bragg reflections of the structure[11]. However, for defect structures, the information that describes the defect is located in the diffuse scattering between Bragg spots, and it is more efficient to compare images. The entire discussion relates to both real space and reciprocal space, although only real space images will be referred to.

12.5.2Simulated Thermal Annealing

Simulated thermal annealing is a relatively new technique for finding the global minimum of a multivariate function[15]. The algorithm is based upon assigning an energy to the system which is a function of the parameters being varied, with the optimum configuration of the system being the minimum energy state, the ground state. A temperature is also assigned to the system and the temperature is slowly being reduced as the configuration is changed. From the initial configuration $E_0(x_1, x_2, ..., x_n)$, the parameters are varied in a random fashion and for each variation the new energy $E_j(x_1, x_2, ..., x_n)$ is calculated. The new configuration is always accepted if $E = E_j(x_1, x_2, ..., x_n) - E_{j-1}(x_1, x_2, ..., x_n) < 0$. Otherwise the new configuration has a probability P of being accepted, where

$$P = e^{-\frac{E}{T}}$$
¹⁹⁾

E and T being dimensionless quantities.

For each temperature, the system undergoes a given number of variations, accepting or rejecting the new configuration based upon the above criteria. When a specified number of successful transitions have taken place, the temperature is lowered by a certain amount, and the parameters are changed again. As a function of iterations, the energy of the system decreases towards what is hoped to be the minimum energy configuration and the process is terminated when either no more successful variations are made for a given number of attempts or the temperature reaches a lower limit.

When comparing calculated and experimental images, the energy of the system can be chosen to be 2 or any of the other quantities that measures image-mismatch. When basing the comparison on the cross-correlation coefficient, the energy can be taken as 1-CCF.

Simulated thermal annealing is a straight forward technique that has proven to be very powerful for finding global minimum without getting trapped in local minima. It is sensitive to the starting conditions and the choice of starting temperature and some experimentation may be required. Near the minimum, it tends to be less optimal than search techniques based upon gradient methods and switching to a different search algorithm may be an alternative once the simulated annealing algorithm has terminated.

12.5.3Simulated Evolution

Simulated evolution is another technique for obtaining the global minima which is modeled after Darwin's principle of "survival of the fittest"[16] It starts with an initial configuration of all the variables to be fitted and produces a number of sets 1 from the initial set using a random generator (mutation generator). This set 1 represents the first generation of children. The algorithm proceeds in the following way:

i)Evaluate a quality function Q (goodness of fit) for all l children.

ii)Select a subset $(\mu < l)$ of survivors which will be he parents of the next generation.

iii)Create a new generation by applying the random generator, after selecting and mixing a part of the parent's parameter vectors.

iv)Loop back to i) until one of the following criteria are met: a) a maximum number of generations have been reached or b) a critical goodness-of-fit has been reached.

12.5.4Other Techniques

There are other ways to do the refinement which is based upon changing the input parameters so that the system moves in a path where the gradient with respect to the fit is the largest[17]. Each method has its advantages. Simulated thermal annealing and simulated evolution are good techniques for getting close to the optimum fit. Once close to the minimum, gradient methods may be used for further refinement until the match is within the uncertainty of the measurement.

12.6.References

1. Völkl E. et al (1994) Density correction of photographic material for further image processing in electron microscopy, Ultramicroscopy 55, 75-89

2. Ruijter W.J. de and Weiss J.K. (1992) Methods to measure properties of slow-scan CCD cameras for electron detection, Rev. Sci. Insts. 63, 4314

3. Mooney P.E. et al. (1993) MTF restoration with slow-scan CCD cameras, Proc. Annual Meeting of the Microsc. Soc. of America 51, 262-263

4. Ruijter W.J. de and Weiss J.K. (1993) Detection limits in quantitative off-axis electron holography, Ultramicroscopy 50, 269-283

5. Frank J. (1972) Two-dimensional correlation functions in electron microscope image analysis, Electron Microscopy, The Institute of Physics, 622

6. Saxton W.O. (1996) Pre-processing of data, registration, distortions, resampling, noise removal, and noise estimation, NCEM workshop on quantitative HRTEM, April 18-20, NCEM/LBNL Berkeley CA, USA

7. Zhang H. et al (1995), Structure of planar defects in (Sr0.9Ca0.3)1.1CuO2 infinite-layer superconductors by quantitative high-resolution electron microscopy, Ultramicroscopy 57, 103-111

8. Smith A.R. and Eyring L. (1982) Calculation, display and comparison of electron microscope images modeled and observed, Ultramicroscopy 8, 65-78

9. Press, W.H. et al (1986) Numerical Recipes, The art of scientific computing. Cambridge University Press 1986 ISBN 0 521 30811 9, p. 502

10. King W.E. and Campbell G.H. (1994) Quantitative HREM using non-linear least-squares methods, Ultramicroscopy 56, 46-53

11. Thust A and Urban K. (1992) Quantitative high-speed matching of high-resolution electron microscopy images, Ultramicroscopy 45, 23

12. Möbus G. and Rühle M. (1994) Structure determination of metal-ceramic interfaces by numerical contrast evaluation of HRTEM micrographs, Ultramicroscopy 56, 54-70

13. Hÿtch M.J. and Stobbs W.M. (1994) Quantitative criteria for the matching of simulations with experimental images, Microsc. Microanal. Microstruct. 5, 133-151

14. Hÿtch M.J. and Stobbs W.M. (1994) Quantitative comparison of high resolution TEM images with image simulations, Ultramicroscopy 53, 191-203

15. Thust A., Lentzen M. and Urban K. (1994) Non-linear reconstruction of the exit plane wave function from periodic high-resolution electron microscopy images, Ultramicroscopy 53, 101-120

16. Möbus G. (1996) Retrieval of crystal defect structures from HREM images by simulated evolution I. Basic technique, Ultra-

microscopy 65, 205-216 17. Press, W.H. et al (1986) Numerical Recipes, The art of scientific computing. Cambridge University Press 1986 ISBN 0 521 30811 9, p. 521