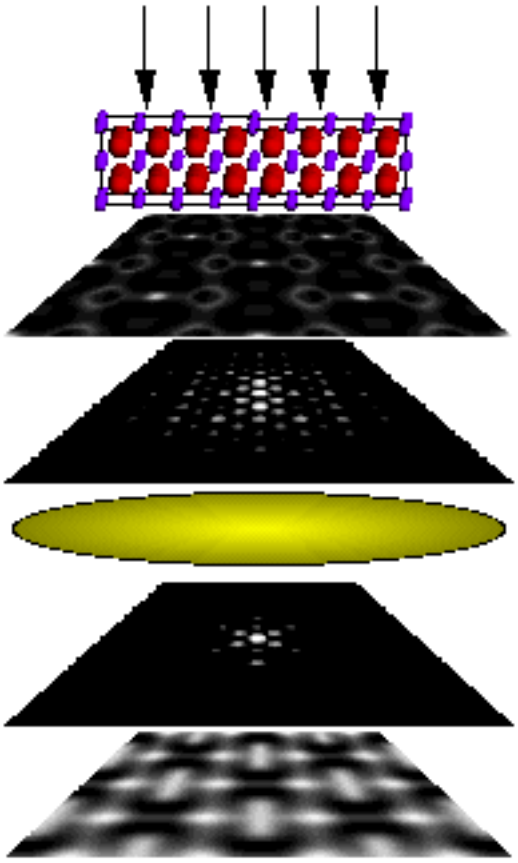
MacTempas

HRTEM Image Simulation Software Package



User Manual

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Chapter

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Installation

The application MacTempas and its associated files are all compacted in the file MacTempas.sea which is a self-extracting archive. After double-clicking on the application on the disk, select a location on your hard disk for placement of the folder "MacTempas Folder" which will be created automatically as part of the extraction procedure. There are two versions of MacTempas supplied on your disk, MacTempasPPC and MacTempas68K for the PowerPC and 680XX Macintoshes respectively. You should run the appropriate version for your machine.

Installing the Hardware Protection Key

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. Before installing the hardware key, drag the file "TRSecurity" found inside the folder “MacTempas Folder:Put In System Folder:Put in Extension Folder” to a closed System Folder. If you are running System 7 or later, the file will be automatically placed in the extension folder within the System Folder. Make sure you install the init TRSecurity before installing the hardware protection key. Shut down the computer. With the computer turned off, unplug the keyboard from the back of the computer and plug the hardware key into the freed up port in the back of the computer. Connect the keyboard cable to the other end of the hardware protection key, making sure that all connections are good. Restart the computer.

Activating the Hardware Key and Personalizing the Program

Before MacTempas can be run on a new system, you must run the program, MacTempasKey. Enter your name and affiliation as appropriate. This program is not required again unless the program is moved to another machine. At times when the operating system is changed it may also be necessary to run the

installation program once more. If this happens, a message will come up and say to contact Total Resolution or to run the installation program.

Changing Hardware or Versions of the MacOS

If you have just changed your computer or installed a new clean version of the MacOS, you must ensure that the extension TRSecurity is placed on the new machine/new extension folder. Without the extension in place, the program will not recognize the hardware key and MacTempas will run in demonstration mode.

MathLib is a shared library that is used on PowerMacs. When MacTempas runs on a PowerMac it uses the math routines that are contained in the file Mathlib. Unfortunately, the first MathLib provided by Apple was very poorly optimized and thus the Mathlib provided with MacTempas is intended for your use in case the version you have on your computer is older than the one provided with MacTempas. If you are running MacOS 7.5.3 the Mathlib is automatically built into the operating system and you don’t need the supplied MathLib. The file Mathlib if present is found in the Extension Folder inside the System Folder.

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[1]. 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[2]. 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).

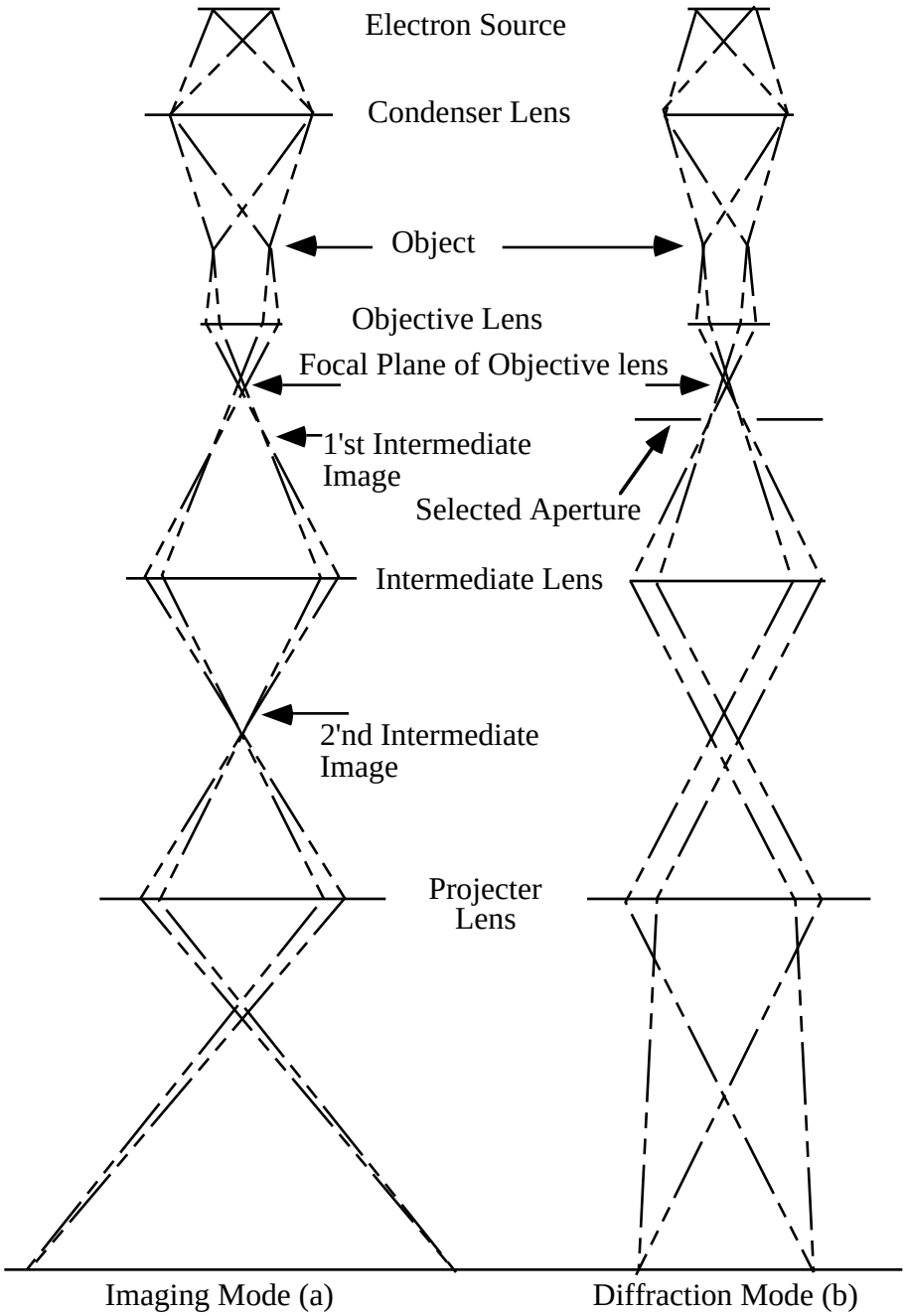

Figure 1. Geometrical optics representation of the TEM in imaging mode (a), and diffraction mode (b)

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

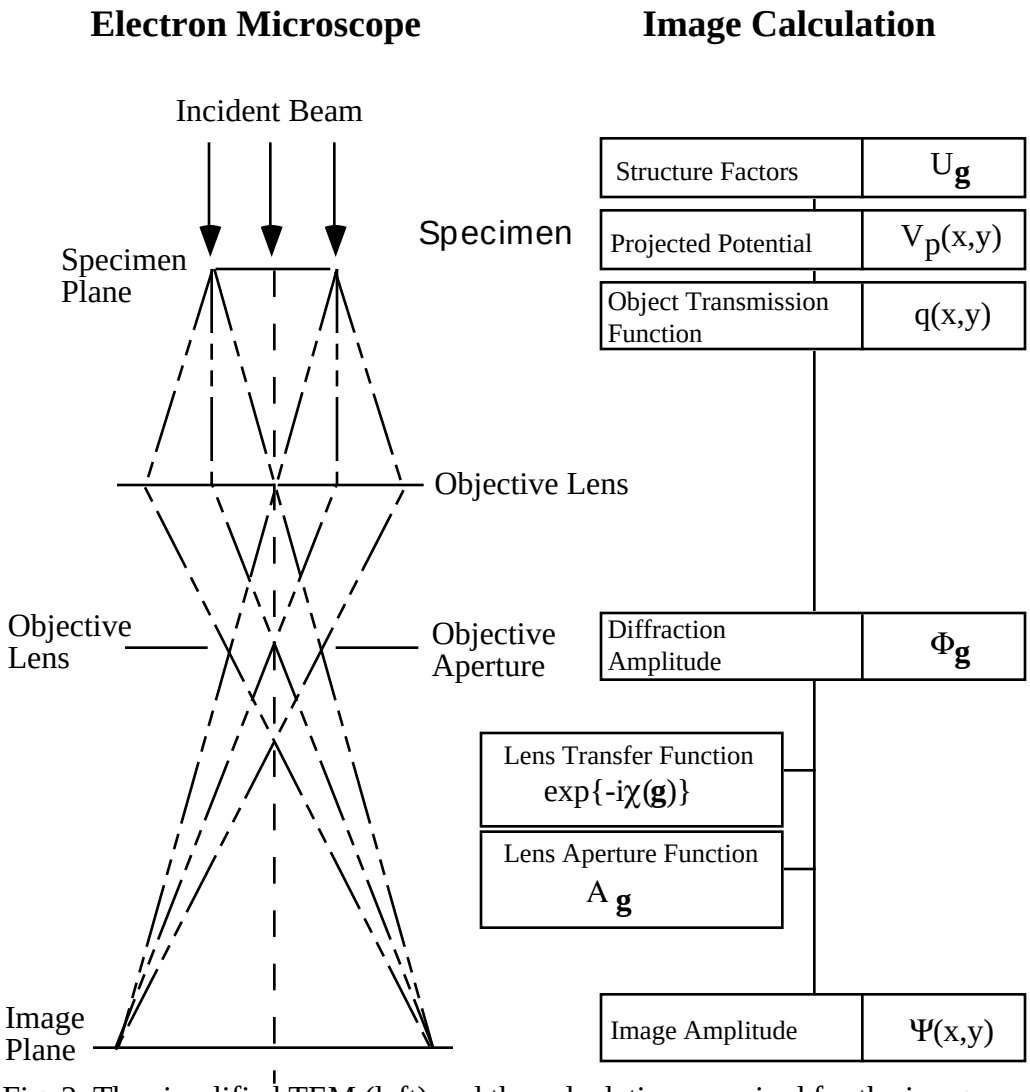

Fig. 2. The simplified TEM (left) and the calculations required for the image simulation (right). The three principal planes are marked.

Cowley and Moodie (1957) showed that the interaction of an electron beam with a specimen could be described by the so-called multislice approximation, in which electrons propagate through the specimen and scatter from the crystal potential, the electron scattering is described by the so-called phase-grating function, a complex function of the potential, and the electron propagation is computed with a propagation function dependent on the electron wavelength. Since then there have been numerous formulations of the multislice approximation derived from the Schrödinger equation.

Simulating TEM Images

The problem of simulating images thus becomes a problem of computing the electron wavefields (wavefunction) at three microscope planes. Currently the best way to produce simulated images 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 of these steps will be covered in the next sections. However, because of space constraints, it is impossible to cover everything in great depth. For detailed derivation, the reader is encouraged to read the many excellent texts on the subject.

Chapter 2

Theory of Image Simulation

Modeling the Specimen

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

$$\phi(\mathbf{r}) = \int d^3\mathbf{r}' \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \qquad 1$$

where $\rho(\mathbf{r})$, the charge density is:

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

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

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

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

$$\varphi(\mathbf{r}) = \int \Phi(\mathbf{u}) e^{-2\pi i \mathbf{u}\cdot\mathbf{r}} d\mathbf{u} = \sum_{\mathbf{H}} \Phi(\mathbf{H}) e^{-2\pi i \mathbf{H}\cdot\mathbf{r}} \qquad 4$$

since because of the periodicity of the unit cell, $\Phi(\mathbf{u})$ is non-zero only when $\mathbf{u} = \mathbf{H} = h\mathbf{a}^* + k\mathbf{b}^* + l\mathbf{c}^*$, $\mathbf{H}$ being a reciprocal lattice vector.
The potential $\Phi(\mathbf{H})$ is given as a sum over all atoms in the unit cell

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

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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 which are used by image simulation programs[3].
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 calcu-

lated as:

$$\Phi(\mathbf{H}) = \sum_{\substack{unit\ cell \\ atoms\ i}} f_i^{el}(\mathbf{H}) Occ(\mathbf{r}_i) \exp[-B_i \mathbf{H}^2] e^{2\pi i \mathbf{H}\cdot\mathbf{r}_i} \qquad 6$$

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

Simulating the Interaction Between the Electrons and the Specimen

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

$$[-\frac{h^2}{8\pi^2 m}\nabla^2 - e\phi(\mathbf{r})]\Psi(\mathbf{r}) = E\Psi(\mathbf{r}) \qquad 7$$

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 $\mathbf{k}_0$, $k_0 = 2\pi/\lambda$, so that the incident electron wave is written

$$\Psi_0(\mathbf{r}) = \exp\{i(\omega t - 2\pi \mathbf{k}_0 \cdot \mathbf{r})\} \qquad 8$$

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

$$V(\mathbf{r}) = \frac{8\pi^2 me}{h^2}\phi(\mathbf{r}) \qquad 9$$

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 solu-

tions to the electron wavefield at a position T within the specimen.

The Weak Phase Object Approximation

In the Phase Object Approximation (POA)[4], the phase of the electron wavefunction after traversing a specimen of thickness T is given as

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

with

$$\sigma = 2\pi me\lambda\left[1+\frac{eE}{mc}\right]\Big/h^2 \qquad 11$$

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:

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

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[5]. The FT of the wavefunction gives the amplitude and phase of scattered electrons and in the WPOA one has:

$$\Psi(\mathbf{u}) = \delta(\mathbf{u}) - i\sigma \mathrm{V}_p(\mathbf{u})T \qquad 13$$

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 scatter-

ing 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 **k** is written as a linear combination of Bloch waves b(**k**,**r**) with coefficients ε[6]. Each Bloch wave is itself expanded into a linear combinations of plane waves which reflect the periodicity of the crystal potential.

$$\psi(\mathbf{r}) = \sum_j \varepsilon^{(j)} b^{(j)}(\mathbf{k},\mathbf{r}) = \sum_j \varepsilon^{(j)} \sum_{\mathbf{g}} c_{\mathbf{g}}^{(j)} \exp[-2\pi i(\mathbf{k}_0^{(j)} + \mathbf{g})\cdot\mathbf{r}] \quad 14$$

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

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

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[7,8] is 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:

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

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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

$$\psi_1(x,y,z+dz) = \exp[-i\sigma\int_z^{z+dz} V(x,y,z')dz']\psi(x,y,z) \equiv q(x,y)\psi(x,y,z)$$

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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

$$\psi(x,y,z+dz) = \exp[-i\sigma dz\nabla^2_{x,y}] \cdot \psi_1(x,y,z) \qquad 18$$

The last equation represents a convolution in real space and is

solved more efficiently in Fourier space[9], where the equation transforms to

$$\Psi(\mathbf{H}, z+dz) = \exp[-i\pi\lambda dz\mathbf{H}^2]\cdot\Psi_1(\mathbf{H}, z) \equiv p(\mathbf{H}, dz)\cdot\Psi_1(\mathbf{H}, z) \quad 19$$

where $\Psi(\mathbf{H},\mathbf{z})$ are the Fourier coefficients of ψ(x,y,z). p(**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[10].

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

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

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

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

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 1/2 of 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.

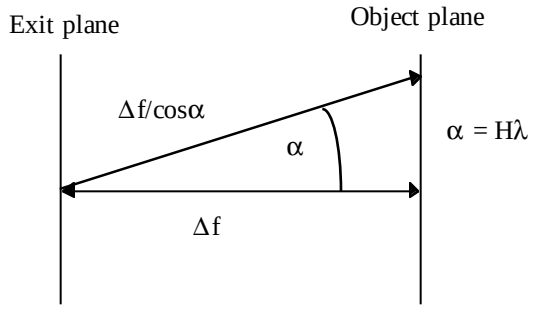
The Image Formation

After the electron wavefield emerge from the specimen, it is subjected to the varies magnetic field of the lenses that form the 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

$$I(x,y) = \left|\psi(x,y,z = exitplane\ \ of\ \ specimen)\right|^2 = \psi_e(x,y)\psi_e^*(x,y)$$

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 Δf.

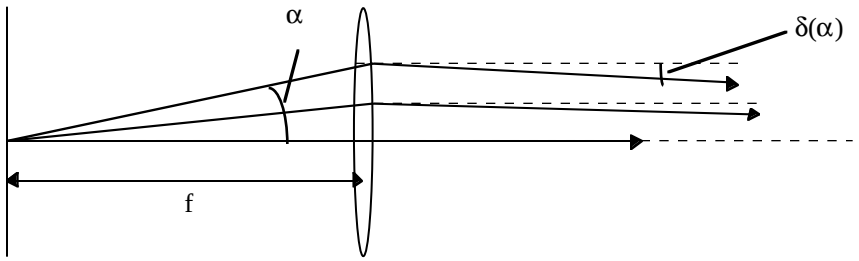


The electron traveling along the optic axis will have a path length of Δf while an electron that has been scattered an angle α=Hλ, will travel a distance Δf /cosα. This can be expressed as a phase difference

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

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 (α=0) which is written as[11]:

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

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 wavefield.

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 Δf 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

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

gives:

$$I = \int |\Psi(f - f_0)|^2 D(f - f_0) df \;\Rightarrow\; \Psi(\mathbf{H}) \rightarrow \Psi(\mathbf{H}) \exp[-1/2(\pi\lambda\Delta\mathbf{H}^2)^2]$$

26

This states that each Fourier term (diffracted beam) is damped according to the equation above[11].

Beam Divergence / Spatial Incoherence

The electron beam is not an entirely parallel beam of electrons, but form rather a cone of an angle α. 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[12] so that:

$$I(\mathbf{r}) = \int |\psi(\mathbf{r}, \alpha)|^2 D(\alpha) d\alpha \;\Rightarrow\; \Psi(\mathbf{H}) \rightarrow \Psi(\mathbf{H}) \exp[\pi\alpha\lambda(C_s\mathbf{H}^2\lambda^2 + \Delta f)]^2 \qquad 27$$

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[13]. 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:

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

and

$$\Psi(\mathbf{H}) = \delta(\mathbf{H}) - i\sigma V_p(\mathbf{H})T \qquad 29$$

Applying the phase shift due to the spherical aberration and the

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

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

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

The FT of the intensity is now given as

$$I(\mathbf{H}) = FT(\psi \cdot \psi^*) = \sum_{\mathbf{H}'} \Psi(\mathbf{H}')\Psi^*(\mathbf{H}-\mathbf{H}') \approx$$
$$\sum_{\mathbf{H}'} \left(\delta(\mathbf{H}') - i\sigma A(\mathbf{H}')V(\mathbf{H}')e^{i\chi(\mathbf{H}')}\right)\left(\delta(\mathbf{H}-\mathbf{H}') - i\sigma A(\mathbf{H}-\mathbf{H}')V(\mathbf{H}-\mathbf{H}')e^{i\chi(\mathbf{H}-\mathbf{H}')}\right) \approx$$
$$\delta(\mathbf{H}) + 2\sigma A(\mathbf{H})V(\mathbf{H})\sin\chi(\mathbf{H}) \qquad 31$$

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 $A(\mathbf{H})\cdot\sin\chi(\mathbf{H})$ The equation above states that each reflection **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 $\sin\chi$ and the damping curves. When $\sin\chi(\mathbf{H}) = -1$ for a large range of frequencies **H**, which is the condition referred to as Scherzer defocus[11], the image can be thought of as:

$$I(x,y) \approx 1 - 2\sigma U(x,y) \qquad 32$$

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.

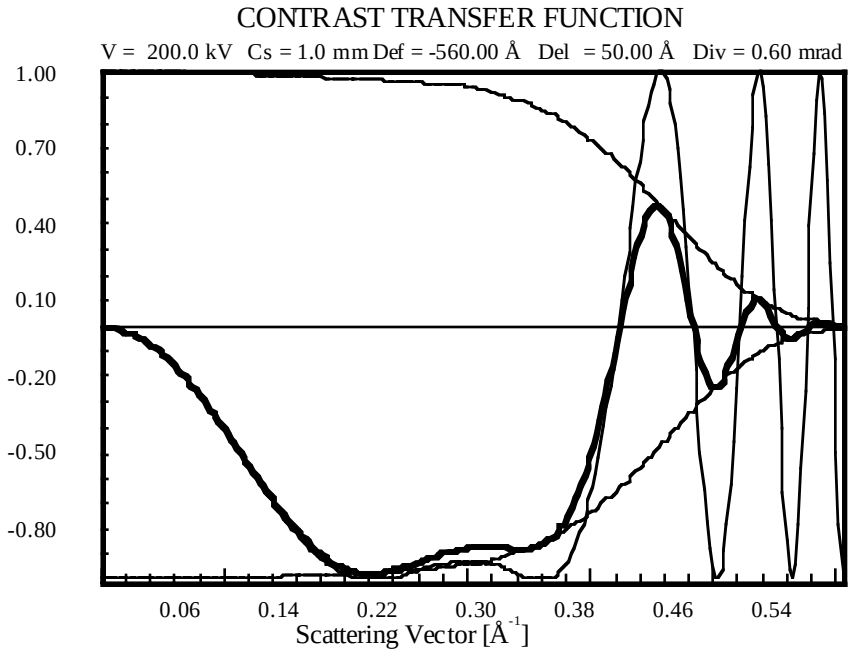


Figure 3. Plot of the Contrast Transfer Function for a 200kV microscope with the parameters indicated.

References

References

1. 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
2. Allpress J.G. et al (1972) n-beam Lattice Images. I. Experimental and Computed Images from $W_4Nb_{26}O_{77}$, Acta Cryst. A 28, 528-536
3. Doyle P.A. and Turner P.S. (1968) Relativistic Hartree-Fock X-ray and Electron Scattering Factors, Acta Cryst.. A 24, 390-397
4. Cowley J.M. and Iijima S. (1972) Electron microscope

image contrast for thin crystals, Z. Naturforschung 27a, 445-451

5. Gibson J.M. (1994) Breakdown of the weak-phase object approximation in amorphous objects and measurement of high-resolution electron optical parameters, Ultramicroscopy 56, 26-32
6. Howie A. (1963) Inelastic scattering of electrons by crystals, Proc. Roy. Soc. A271, 268-275
7. Goodman P, Moodie A.F. (1974) Numerical evaluation of N-beam wave functions in electron scattering by the multislice method, Acta Cryst. A30, 322-324
8. Self P.G.et al. (1983) Practical computation of amplitudes and phases in electron diffraction, Ultramicroscopy 11, 35
9. Ishizuka K. and Uyeda N. (1977) A new theoretical and practical approach to the multislice method, Acta Cryst. A 33, 740
10. 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
11. Scherzer O (1949) The Theoretical Resolution Limit of the Electron Microscope, Journal of Applied Physics 20, 20-29
12. Frank J (1973) The envelope of electron microscope transfer functions for partially coherent illumination, Optik 38, 519-536
13. O'Keefe M.A. (1979) Resolution-damping functions in non-linear images, Proc. of EMSA 37, 556-557

Chapter 3

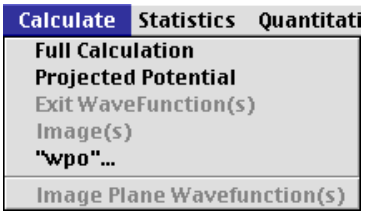
Introduction to MacTempas

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.

The Three Simulation Steps

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.

ExitWavefunctions(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 resolution-limiting aberrations are included via parameters like defocus, spherical aberration, incident beam convergence, spread of defocus, and the position and size of the objective aperture.

wpo - 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.

ImagePlaneWavefunctions(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 surface 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 "***ProjectedPotential***" calculation considers only the specimen structure, "***ExitWavefunctions(s)***" calculation treats the interaction of the specimen with the electron wave, and the "***Image(s)***" calculation simulates how the wave leaving the specimen interacts with the lens system of the electron microscope. Once a simulation has been made, any additional simulation will usually not require a full re-calculation; any change in microscope parameters will not affect the results of the "***ProjectedPotential***" and "***ExitWavefunctions(s)***" 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 output of "***ProjectedPotential***", but "***ExitWavefunctions(s)***" and "***Image(s)***" will need to be re-run. Of course, any change in the specimen structure will require the re-running of all three subprograms.

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 microscope 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 PHSGRT subprogram 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 MSLICE subprogram 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 subprogram 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 "***ProjectedPotential*** " subprogram processed the <**structurename** >.**at** data to produce the specimen potential.

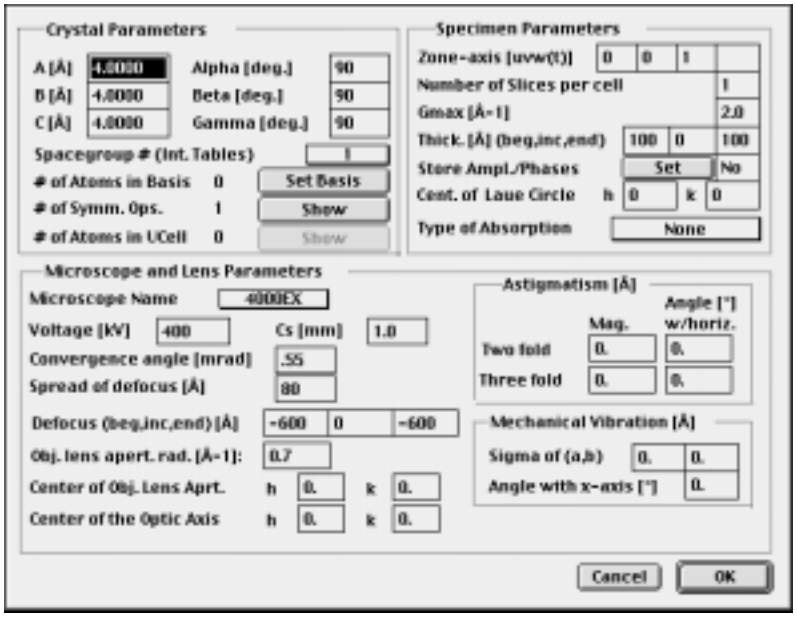
(8) <**structurename** >.**m_prnt** contains information about the way in which the "***ExitWavefunctions(s)*** " subprogram processed the <**structurename** >.**pout** data with the <**structurename** >.**at** to produce the exit-surface wave; that is, it contains information from the multislice computation.

Chapter 4

Running MacTempas

Generating an Input Structure

The first step in running a simulation is generating the structure input file. This is done through New... in the FILE menu. This generates the input dialog window below which requests the following information: All fields have default values and in order to create a valid input file, only valid data for the atoms must be entered.

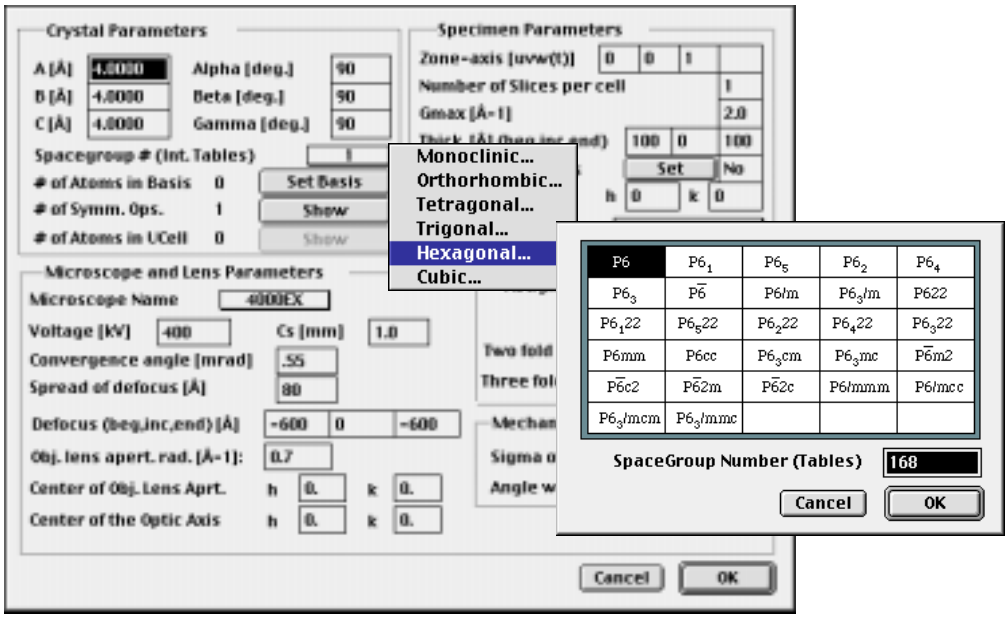


a, b, c,
alpha, beta, gamma
These are the unit cell dimensions in Ångstrøm units, and the unit cell angles in degrees. MacTempas will automatically set the angles depending on the spacegroup, if possible. The program will also automatically set lattice parameters depending on the spacegroup. Thus if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

and **c** are set equal to **a**

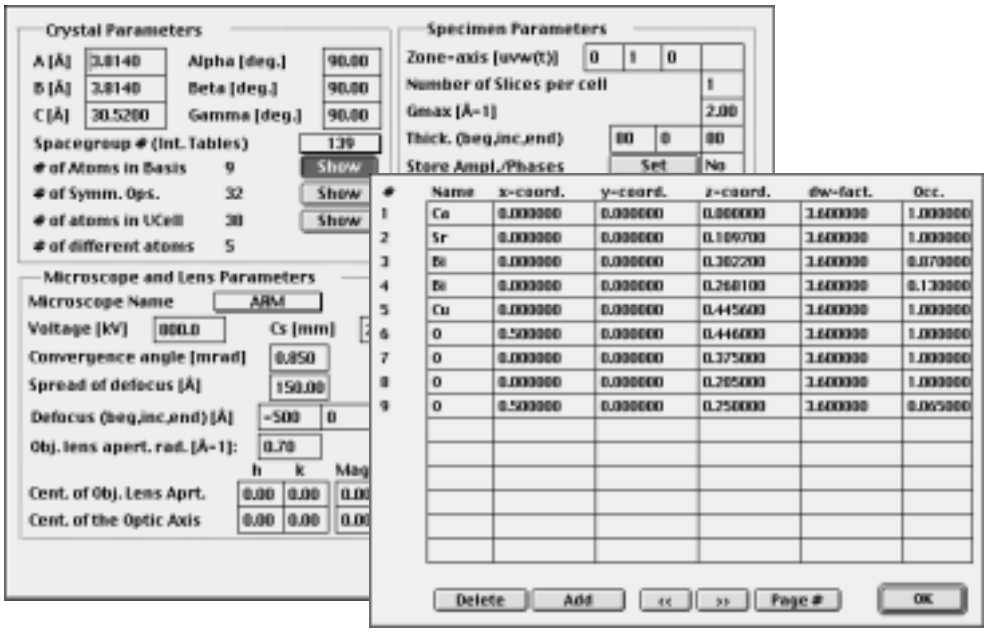
Space group#

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.



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.

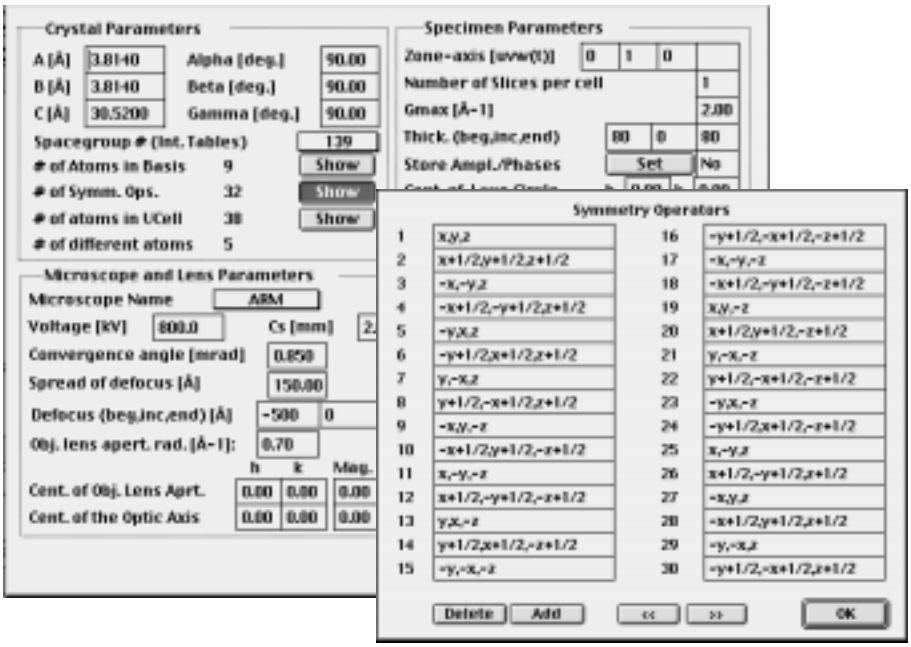
Set Basis
Use this button to bring up the dialog window that enables the input of the atoms in the basis.



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.

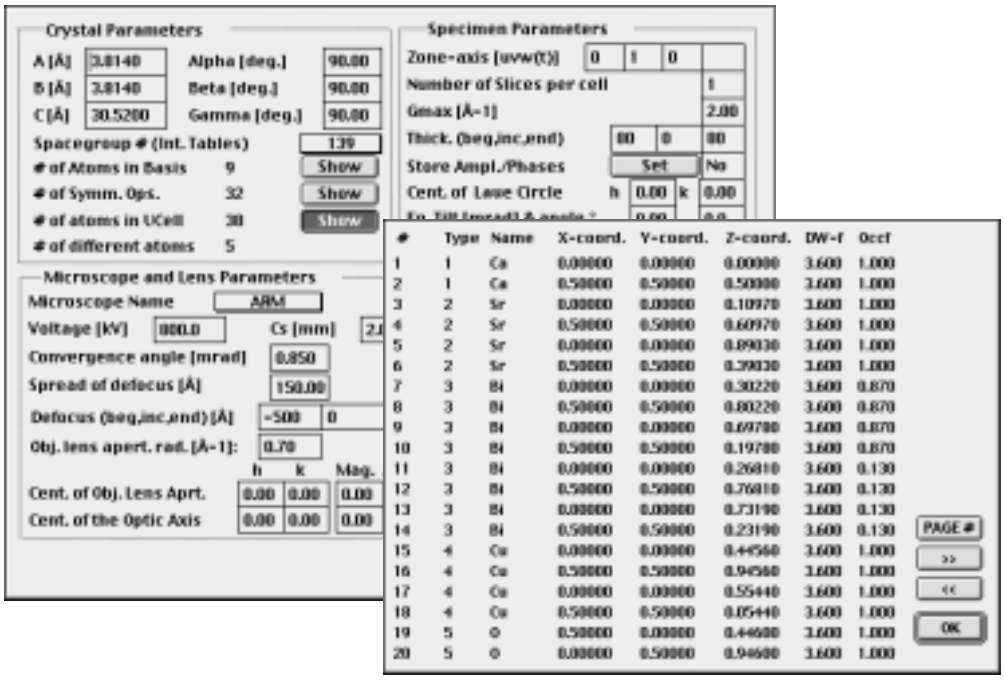
Show (Symmetry Operators)
The symmetry operators are automatically created by specifying the spacegroup. By clicking on this button, a window dis-

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”,

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 $\mathbf{G}_{max}$ may allow the Ewald sphere to approach the so-called pseudo upper-layer line that the multislice 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

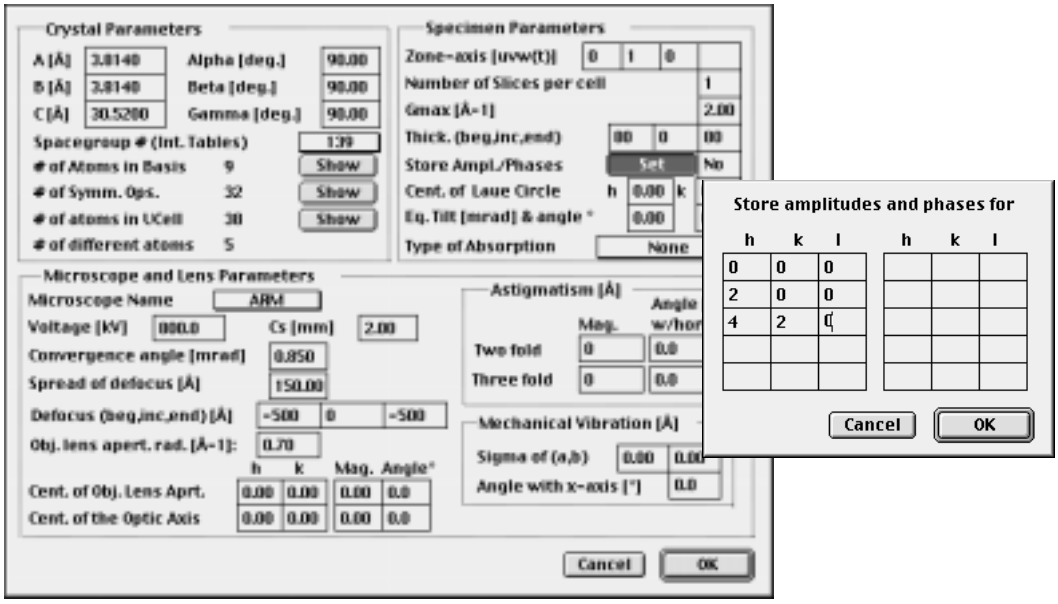
chosen in the Option menu.

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 $\mathbf{G}_{max}$ 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. 100 250 50 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

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.

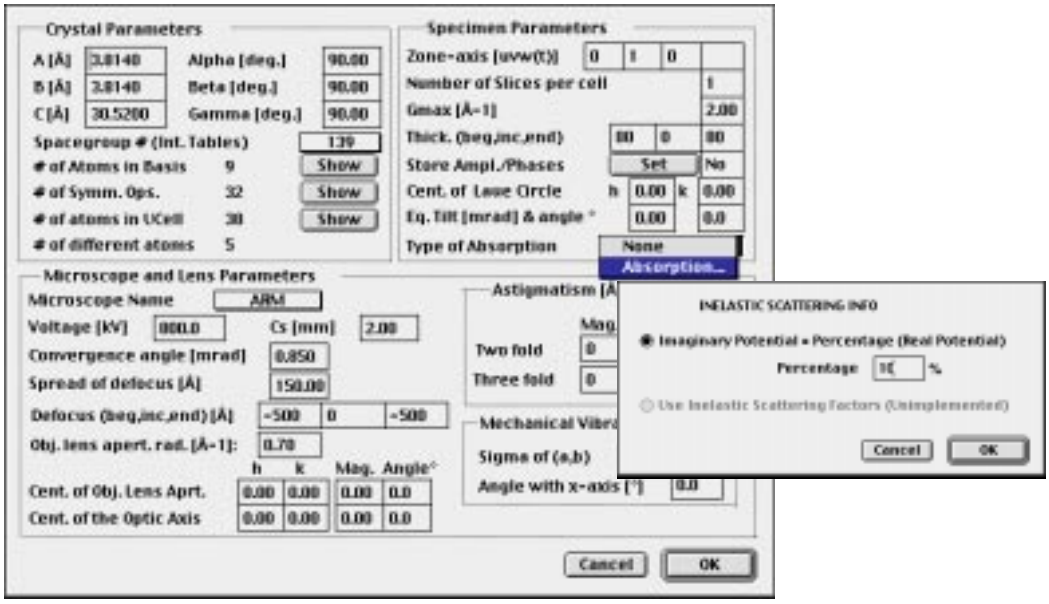
Center of the Laue Circle

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

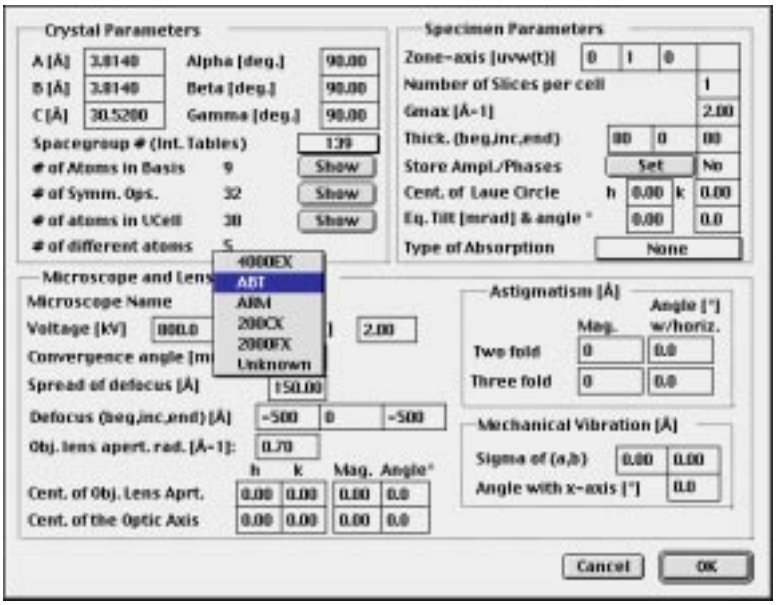
imaginary projected potential.



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

may be made known to MacTempas).



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 SPECIMEN 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.

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).

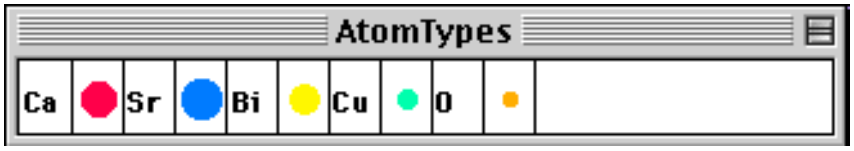
Chapter 5

Windows

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

Status Window

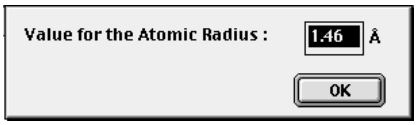
This window shows the current status of the program indicating the number of phasegrating coefficients calculated, the current slice number being calculated, the current image being calculated etc.



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.

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).

Color Window

This window is used to set the color of a particular atom species, the color of the foreground (the color of lines and text) and the background color. Colors for use in pseudocoloring is also picked from the color window. To choose a color, the Color Picker Tool must have been chosen. To select the **foreground** color click on the color with the **Option** Key held down. Holding down the **Option** key and the **Shift** key selects the **background** color.

Tools Window

The following tools are currently defined:

Eraser
By selecting the eraser tool, the cursor turns into an eraser which can be used to erase any part of the image screen. Double-clicking the eraser, erases the entire screen. The eraser

erases the screen with the current background color.

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.

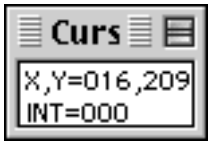
Selection Tool
This tool is used to select a portion of the screen for several possible operations such as copying, cutting, histogram computation etc. To select an area, click at a point in the display and drag the cursor while the mouse button is pressed.

Histogram Tool
When this tool is selected a histogram will be produced for a rectangular region defined by dragging a rectangle while the mouse button is held down. Double-clicking on the histogram tool produces a histogram distribution of the entire image screen.

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

Cursor 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.

MLUT Window

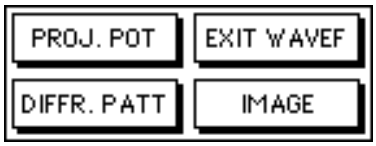
Mono Lookup Table window show the linear relationship between input values and output grey levels. Under normal conditions, each input value maps to the same output value. To change the mapping, the line can be modified by use of the cursor. To change the contrast of the image, the endpoints can be moved by the mouse if the mouse is clicked near the endpoint and the mouse button is held down while the mouse moves. The brightness is changed by clicking near the center of the line and dragging it to the desired location. To reset the lookup tables, just double-click in the MLUT window.

Note: The above only currently works when the monitor is set to 256 colors.

Pseudo Window

Use this window to pseudo color images to enhance certain features in the image. To pseudo-color an image with grey levels, color is substituted for certain grey levels. To do this, select the Color Picker Tool, select a color from the Color Window and deposit the color anywhere in the grey-scale by dragging in the pseudo-color window. Double-clicking in this window resets the grey levels.

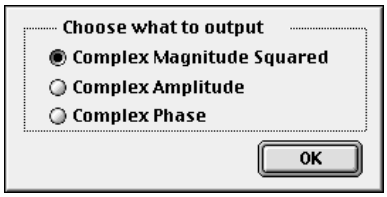
Source Window



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

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

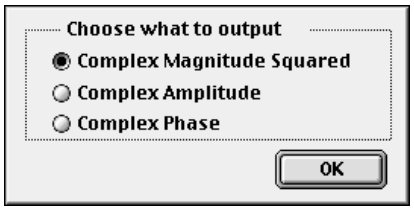
Exit Wavefunction - This is the output of the MSLICE subprogram and shows the distribution of electrons as they emerge from the bottom of the specimen, or at a predefined depth in the specimen. By holding down the OPTION key when selecting the button, one can select to display either the magnitude



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.

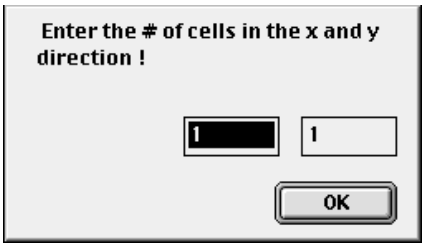
Operand Window

Selecting functions in this window defines the actions to be performed on the source defined by clicking in the Source Window. Prior to selecting an operation, a source must have been selected. The operation currently available are:

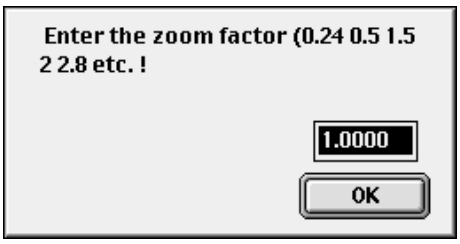
FFT
Use this to perform a Fourier Transform on the source selected in the Source Window. Operating on the Projected Potential will yield the structure factors, operating on the Exit Wavefield will yield the diffraction pattern and operating on the image will give the Power spectrum of the image.

#Unit cells
Use this to specify the number of unit cells that should be displayed. The input requires the number of cells in the a-direction and b-direction. The number of unit cells stay in effect until

explicitly changed.



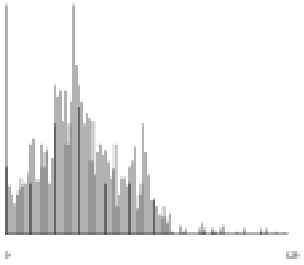
Zoom
Use this selection to Zoom the object to either magnify the object or to reduce the object. A zoom factor greater than 1. magnifies and a zoom factor less than 1. reduces the object. As



with the number of unit cells to be displayed, the zoom factor stays in effect until explicitly changed.

Histeq
This operation performs a histogram equalization on the source. Only a final object of the type “image” is suitable for histogram equalization.

Histog



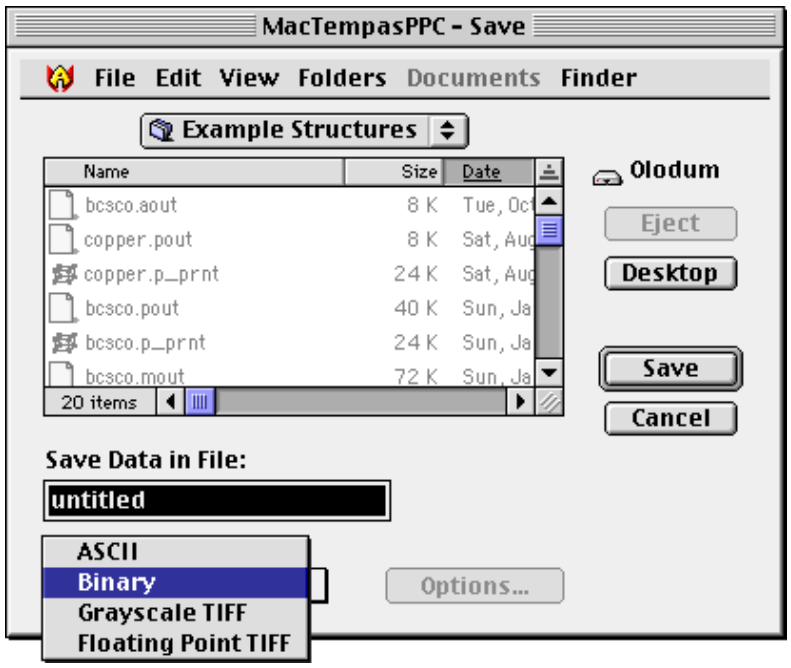
Selecting this operation will produce the histogram of the source.

Display

Before the result of operating on a selected source is displayed in the image window, DISPLAY must have been selected. 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 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

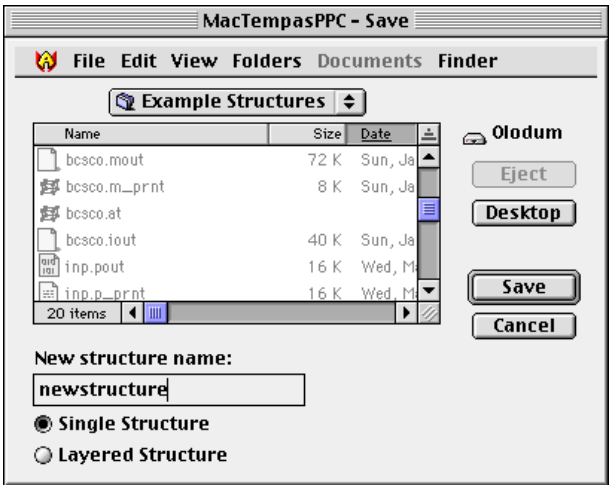
Menus

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 Menu

This menu contains the following commands:

New...
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. A new file can either describe a single structure or a layered structure (see Chapter 8 on how to create a layered structure) depending on the radiobutton selected. Many of the parameters will have default values.



Open...
Open an existing structure file. The standard Macintosh file open dialog is presented and only files of the type ‘TEXT” with the extension “.at” are displayed as selectable. The name of the imaging window will change to reflect the name of the current structure.

Close
Close the file currently in use

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

Save 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.

Save Selection...
Saves the selected portion of the screen into a file. The filetype (PICT, grayscale Tiff or Palette Tiff) can be selected from the file Save Dialog.

Save Window...
Saves the content of the image window into a file. The filetype (PICT, grayscale Tiff or Palette Tiff) can be selected from the file Save Dialog.

Open PICT File...
Open a PICT file and display it in the MacTempas image window. Only pictures that will fit within the MacTempas image window can be opened this way.

Page Setup...
Set the options for the page to be printed.

Print...
Print the MacTempas image window. If a Selection is made, the selection will be printed out. If a histogram window or Trace Window is the foremost window, that window will be printed.

Quit
Quit MacTempas.

Edit Menu

Undo
Undo the last operation. This operation does not work in MacTempas.

Cut
Cut out the selection made by the selection tool. The cut selection can be moved by holding the mousebutton down when the cursor is within the selected area.

Copy
Copy the selection made by the selection tool. The copied selection can be moved around as described in Cut above.

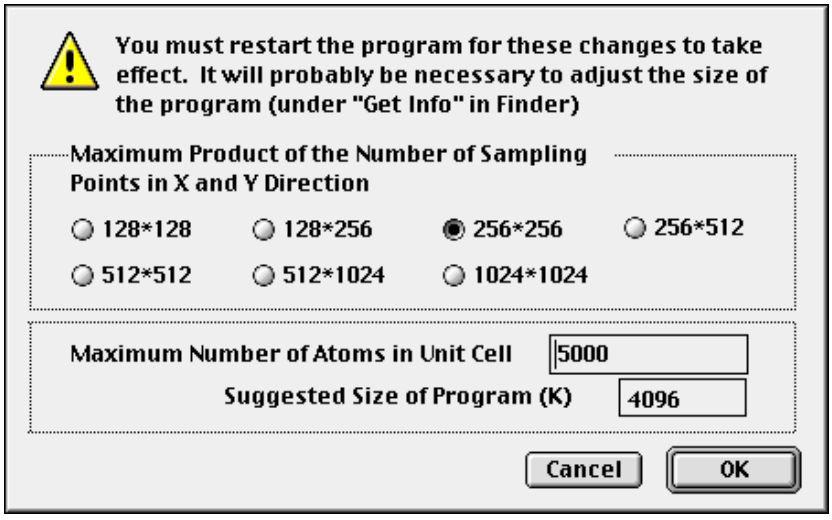
Paste
Paste a selection onto the image window. The source for the paste can be an image cut out from another application or through the cut/copy commands of MacTempas

Clear
Clears the selection made by the selection tool

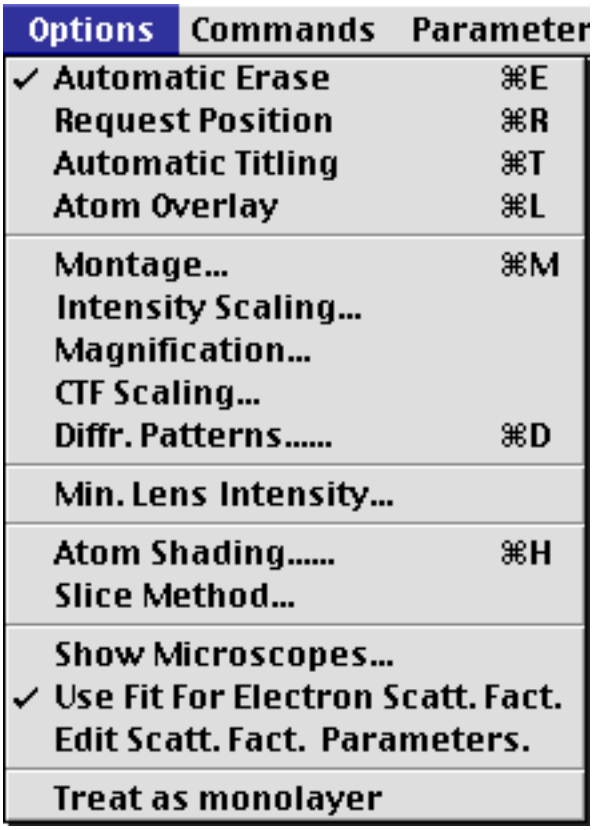
Select All
Select the entire MacTempas image screen for the next operation.

Show Clipboard
Shows the clipboard and the content of the clipboard.

Preferences
The maximum size of a calculation and the maximum number of atoms in MacTempas is by default set to 256*256 and 2500. These numbers can be changed if the size of the program is modified accordingly and sufficient memory is present in the computer. Since running MacTempas with virtual memory turned on is very slow, all memory requirement should be satisfied by physical Random Access Memory.



Options Menu



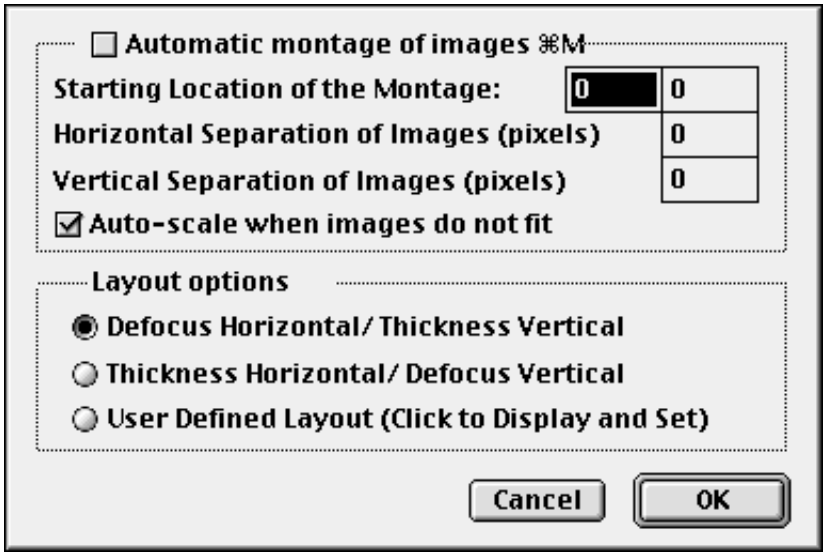
Automatic Erase
Toggles whether the screen is automatically erased before the image window is being drawn into.

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

Automatic Titling
Toggles whether each image will be shown with a descriptive title below the image

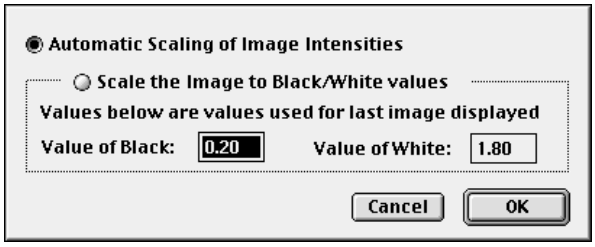
Atom Overlay
If set, the atom positions will be drawn in as circles on top of images. The circles are scaled to the atomic radius and the color is the color set for that atom species. If the Option key is held down while the image is "drawn", only the circles are drawn (no image).

Montage...
Brings up a dialog box, allowing the user to select automatic montage of a series of images, the position of the series of images and the number of pixels to leave between images.

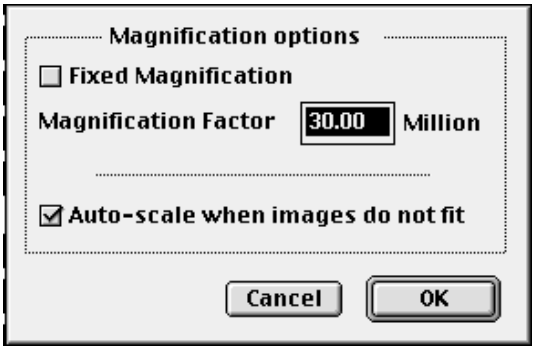


Intensity Scaling...
Brings up a dialog box, allowing the user to manually set the intensity values to be mapped to black and white. The values shown correspond to the last image displayed with automatic

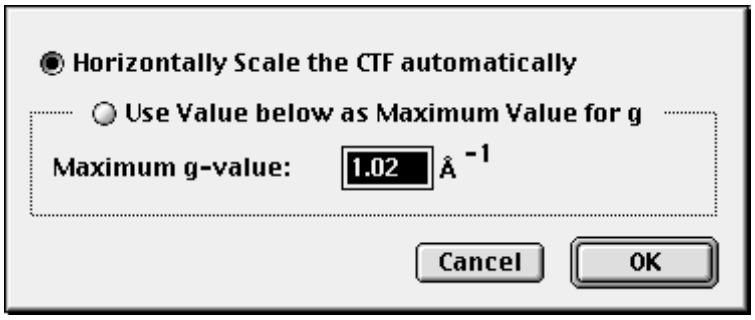
scaling.



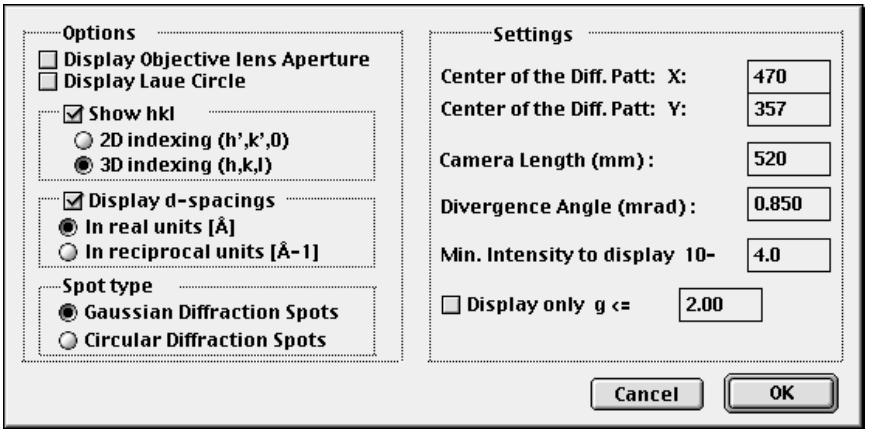
Magnification
Allows the user to set the magnification to a set value. The magnification depends on a screen with a resolution of 72 dots/ inch. If Auto-scaling is set, images will scale to fit the window.



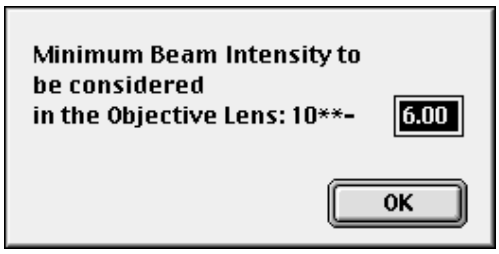
CTF Scaling...
Brings up a dialog box, allowing the user to set the maximum scale of the reciprocal axis during plotting of the Contrast Transfer Function.



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

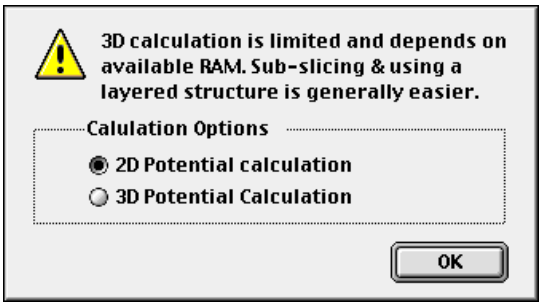


Min Lens Intensity...
Displays a dialog box, allowing the user to manually set the minimum intensity required of a diffracted beam for inclusion in the formation of the image.



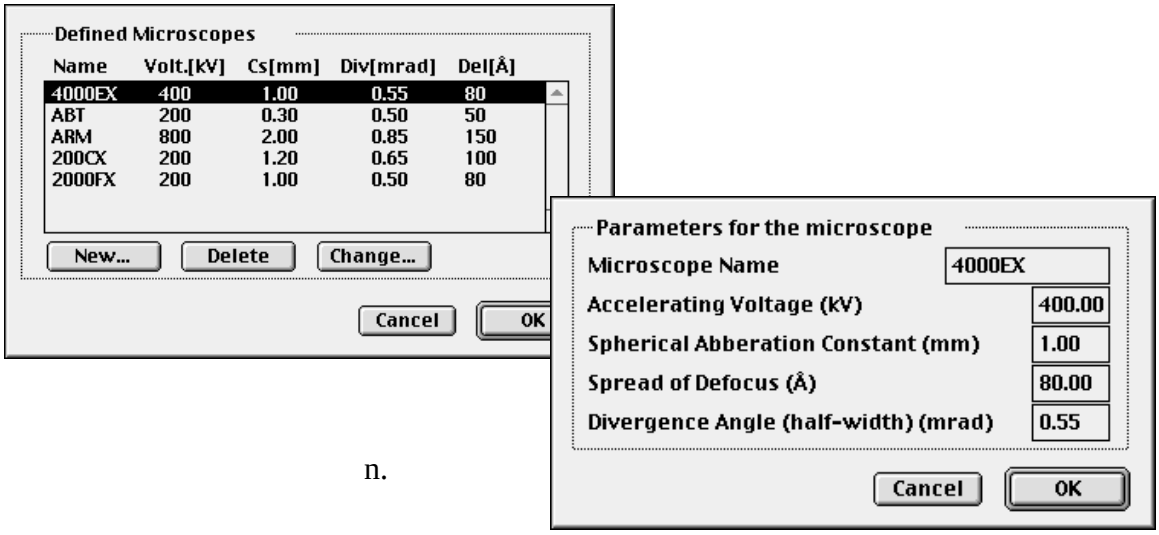
Atom Shading...
Allows the user to select whether atoms drawn should be displayed in color (circles) or as shaded balls (grey

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 (l) 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

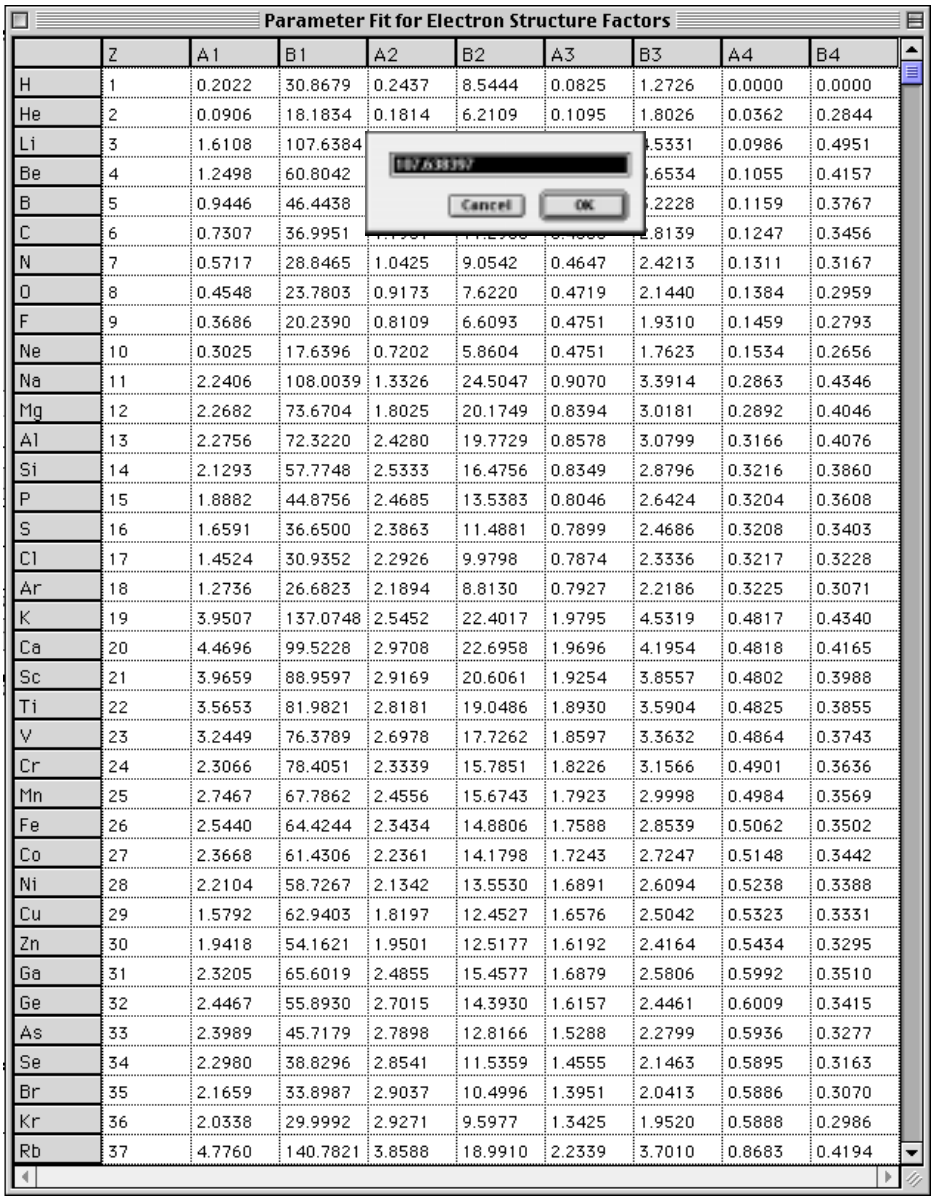


n.

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.

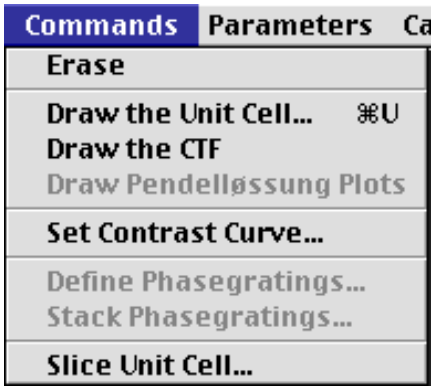
Treat as Monolayer
When this option is set, the calculation treats the unit cell as a non-repeating structure such that the entire specimen is represented by a single unit cell with the thickness of the specimen as the thickness of the unit cell.



Parameter Fit for Electron Structure Factors

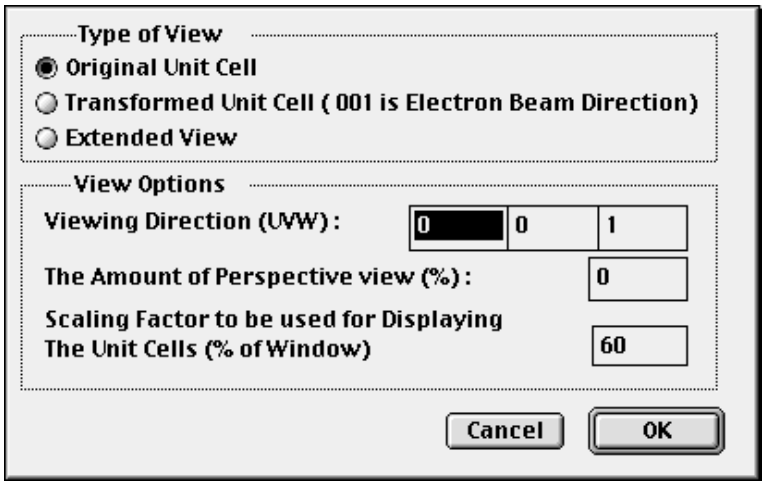
	Z	A1	B1	A2	B2	A3	B3	A4	B4
H	1	0.2022	30.8679	0.2437	8.5444	0.0825	1.2726	0.0000	0.0000
He	2	0.0906	18.1834	0.1814	6.2109	0.1095	1.8026	0.0362	0.2844
Li	3	1.6108	107.6384				.5331	0.0986	0.4951
Be	4	1.2498	60.8042				.6534	0.1055	0.4157
B	5	0.9446	46.4438				.2228	0.1159	0.3767
C	6	0.7307	36.9951				.8139	0.1247	0.3456
N	7	0.5717	28.8465	1.0425	9.0542	0.4647	2.4213	0.1311	0.3167
O	8	0.4548	23.7803	0.9173	7.6220	0.4719	2.1440	0.1384	0.2959
F	9	0.3686	20.2390	0.8109	6.6093	0.4751	1.9310	0.1459	0.2793
Ne	10	0.3025	17.6396	0.7202	5.8604	0.4751	1.7623	0.1534	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
Al	13	2.2756	72.3220	2.4280	19.7729	0.8578	3.0799	0.3166	0.4076
Si	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
Cl	17	1.4524	30.9352	2.2926	9.9798	0.7874	2.3336	0.3217	0.3228
Ar	18	1.2736	26.6823	2.1894	8.8130	0.7927	2.2186	0.3225	0.3071
K	19	3.9507	137.0748	2.5452	22.4017	1.9795	4.5319	0.4817	0.4340
Ca	20	4.4696	99.5228	2.9708	22.6958	1.9696	4.1954	0.4818	0.4165
Sc	21	3.9659	88.9597	2.9169	20.6061	1.9254	3.8557	0.4802	0.3988
Ti	22	3.5653	81.9821	2.8181	19.0486	1.8930	3.5904	0.4825	0.3855
V	23	3.2449	76.3789	2.6978	17.7262	1.8597	3.3632	0.4864	0.3743
Cr	24	2.3066	78.4051	2.3339	15.7851	1.8226	3.1566	0.4901	0.3636
Mn	25	2.7467	67.7862	2.4556	15.6743	1.7923	2.9998	0.4984	0.3569
Fe	26	2.5440	64.4244	2.3434	14.8806	1.7588	2.8539	0.5062	0.3502
Co	27	2.3668	61.4306	2.2361	14.1798	1.7243	2.7247	0.5148	0.3442
Ni	28	2.2104	58.7267	2.1342	13.5530	1.6891	2.6094	0.5238	0.3388
Cu	29	1.5792	62.9403	1.8197	12.4527	1.6576	2.5042	0.5323	0.3331
Zn	30	1.9418	54.1621	1.9501	12.5177	1.6192	2.4164	0.5434	0.3295
Ga	31	2.3205	65.6019	2.4855	15.4577	1.6879	2.5806	0.5992	0.3510
Ge	32	2.4467	55.8930	2.7015	14.3930	1.6157	2.4461	0.6009	0.3415
As	33	2.3989	45.7179	2.7898	12.8166	1.5288	2.2799	0.5936	0.3277
Se	34	2.2980	38.8296	2.8541	11.5359	1.4555	2.1463	0.5895	0.3163
Br	35	2.1659	33.8987	2.9037	10.4996	1.3951	2.0413	0.5886	0.3070
Kr	36	2.0338	29.9992	2.9271	9.5977	1.3425	1.9520	0.5888	0.2986
Rb	37	4.7760	140.7821	3.8588	18.9910	2.2339	3.7010	0.8683	0.4194

Commands Menu

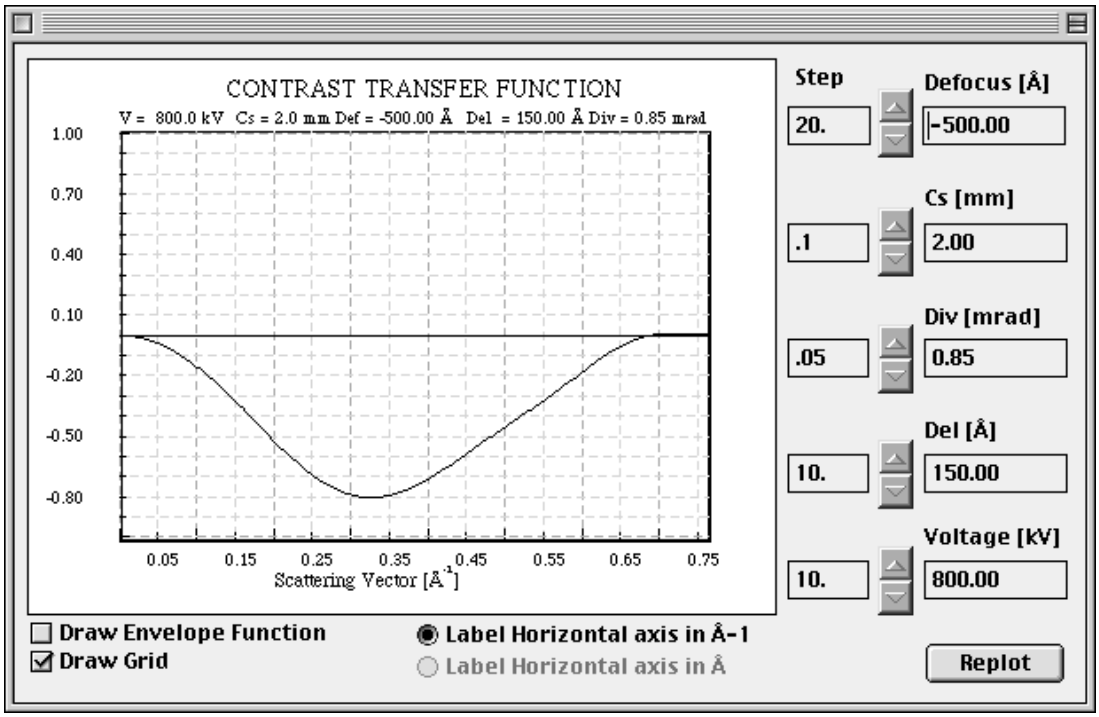


Erase
Erases the selection made by the selection tool.

Draw the unit Cell...
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.



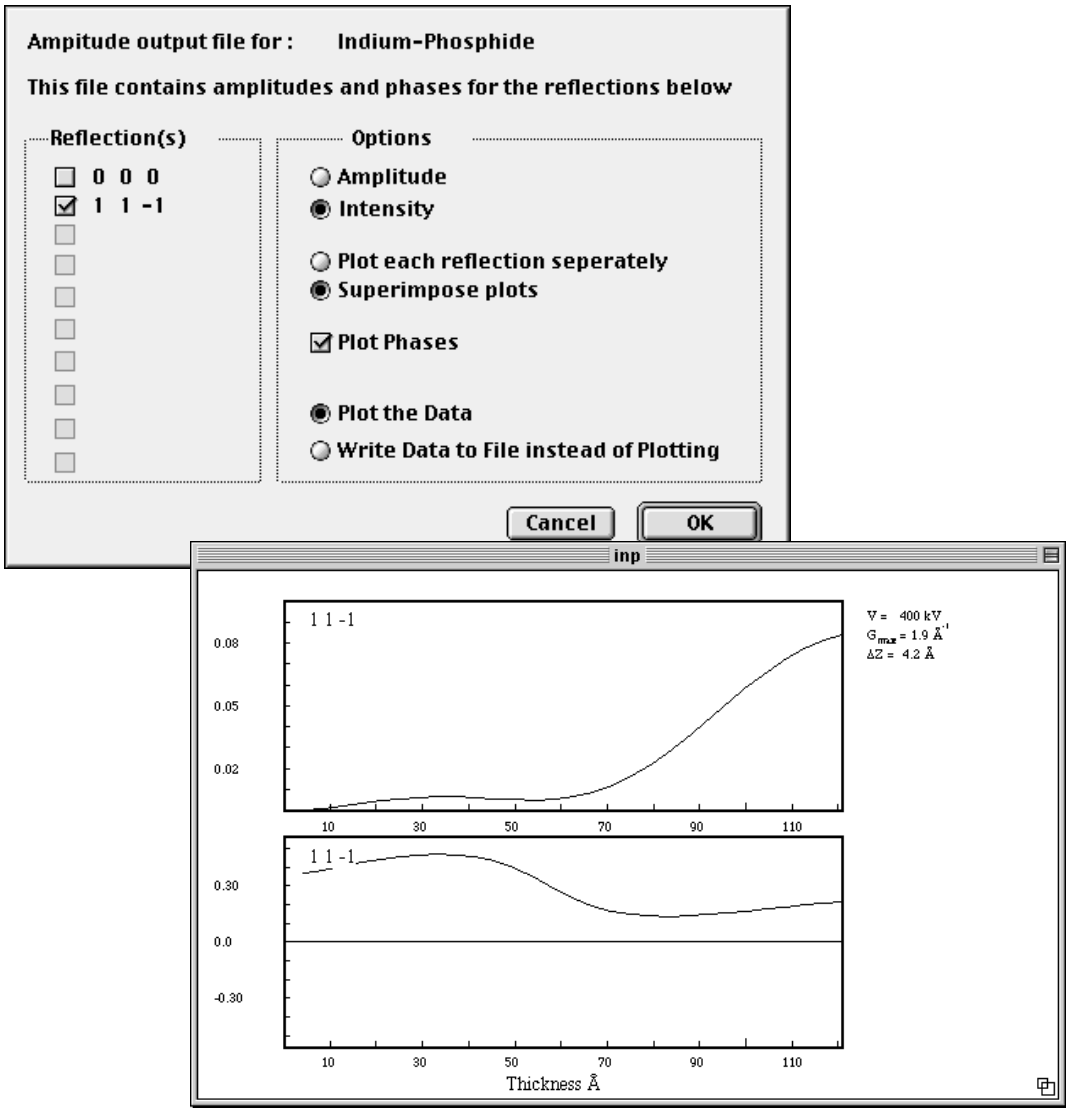
Draw the CTF
Draws the Contrast Transfer Function for the current microscope values. The original microscope values are taken from the structure data, but the user is free to change the values associated with the CTF independent of the values used in calculating the image. By selecting to print with this window being the



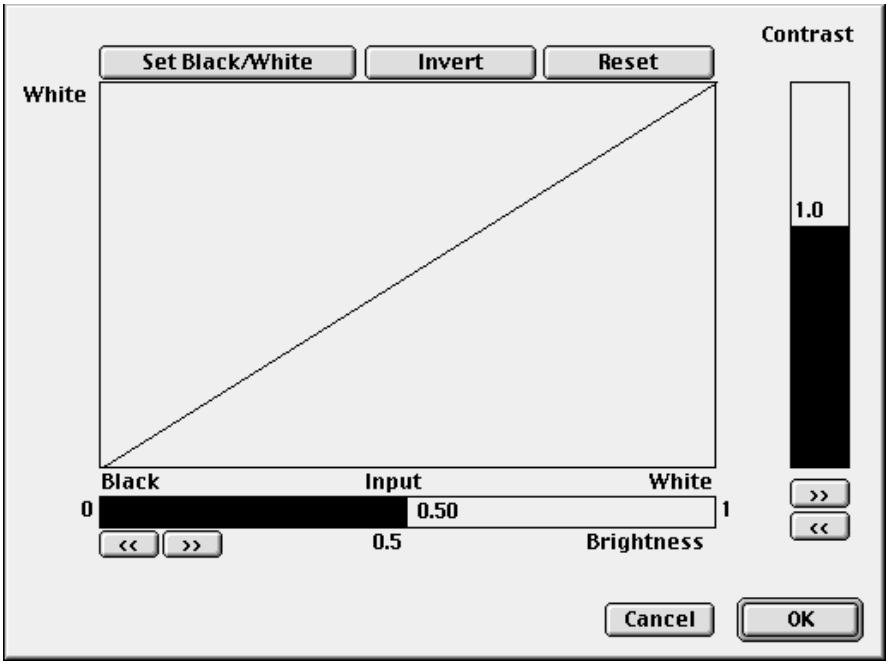
front window, the CTF is printed.

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

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.

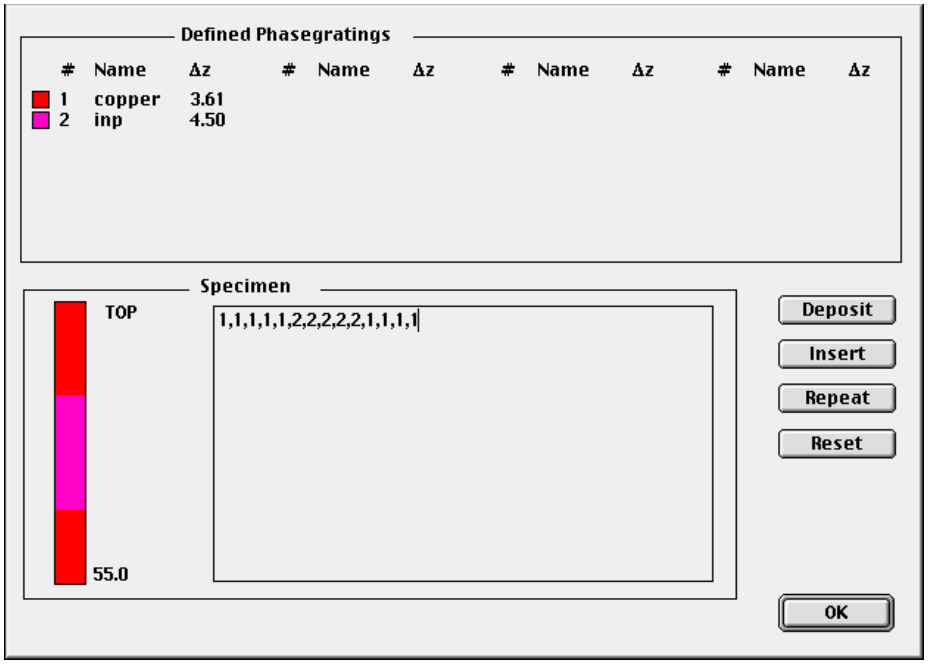


Set Contrast Curve...
Instead of changing the hardware lookup tables to effectively change brightness and contrast values, the brightness and contrast can be set before the images are displayed on to the screen. This is performed from this menu command. Set Black / White

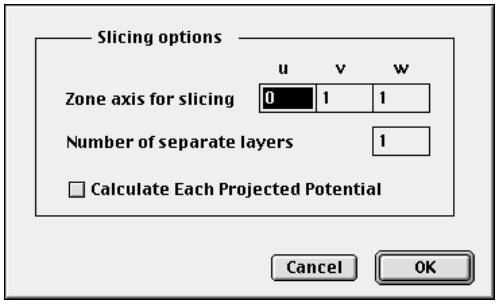


allows the user to specify the curve by selecting two points on the input/output curve.

Stack Phasegratings...
This allows the user to specify a sequence of phasegratings that should be used in the multislice calculation. This applies only to layered structures. See Chapter 9 for a more detailed instruction on how to create a layered structure.

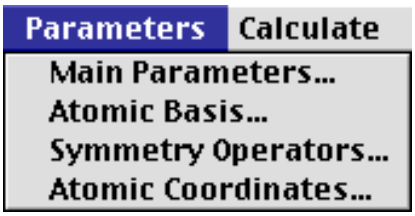


Slice Unit Cell...
Use this option to subdivide a structure into separate layers for use in a layered structure calculation. The direction perpendicular to the slices and the number of slices must be specified.

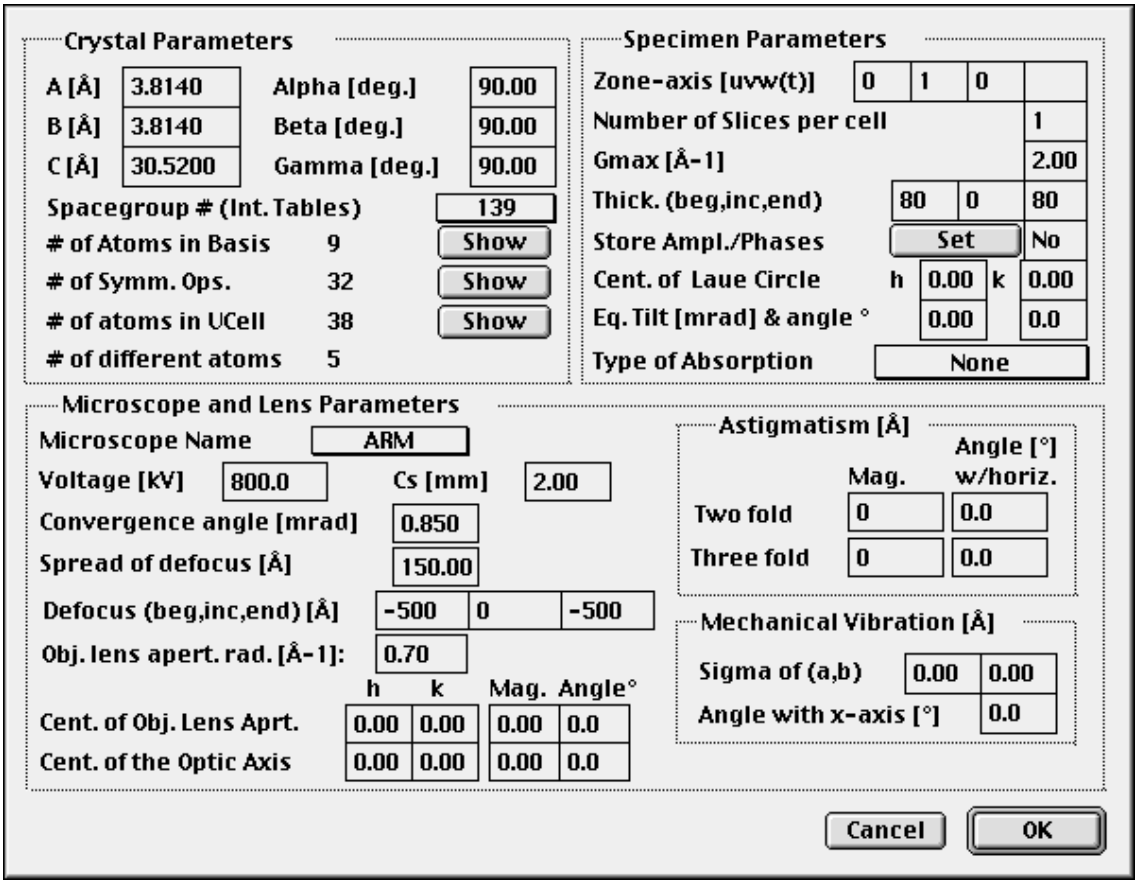


Parameters Menu

Main Parameters...

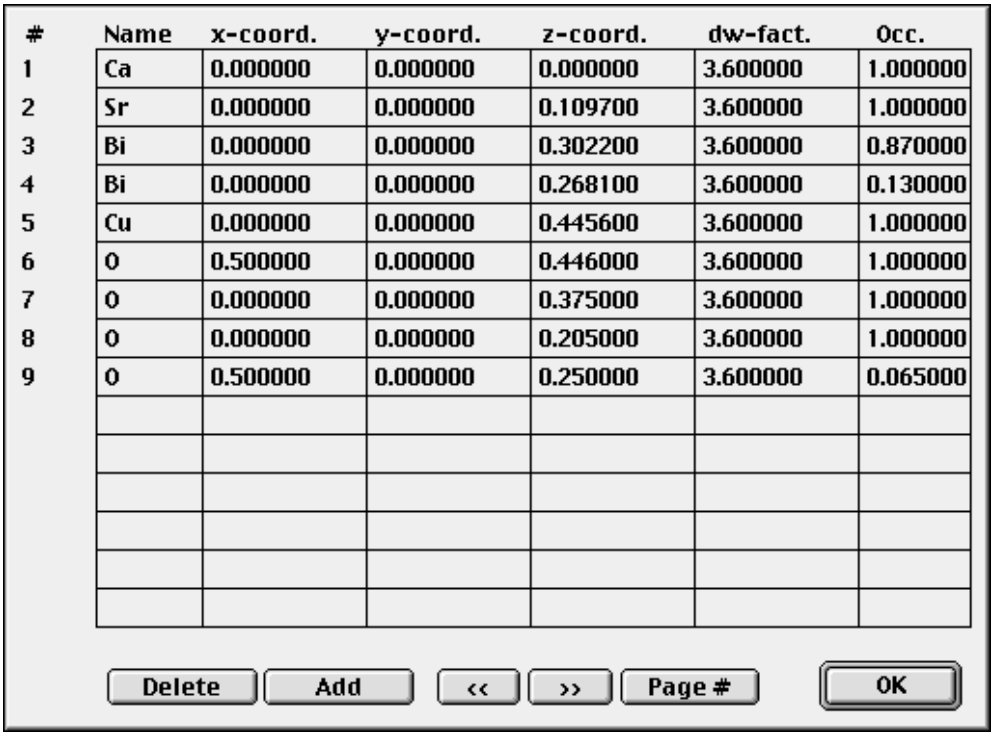


This brings up a dialog box showing the current conditions for the simulation. The values are taken from the input given to the



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

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.

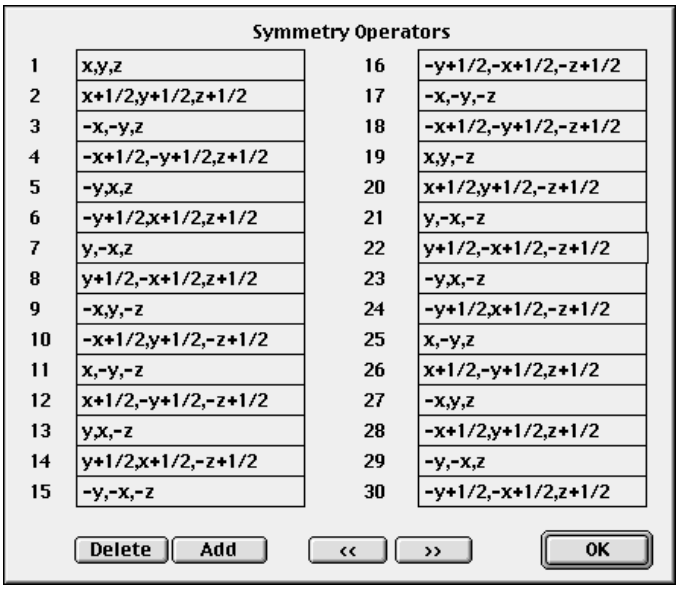


#	Name	x-coord.	y-coord.	z-coord.	dw-fact.	Occ.
1	Ca	0.000000	0.000000	0.000000	3.600000	1.000000
2	Sr	0.000000	0.000000	0.109700	3.600000	1.000000
3	Bi	0.000000	0.000000	0.302200	3.600000	0.870000
4	Bi	0.000000	0.000000	0.268100	3.600000	0.130000
5	Cu	0.000000	0.000000	0.445600	3.600000	1.000000
6	O	0.500000	0.000000	0.446000	3.600000	1.000000
7	O	0.000000	0.000000	0.375000	3.600000	1.000000
8	O	0.000000	0.000000	0.205000	3.600000	1.000000
9	O	0.500000	0.000000	0.250000	3.600000	0.065000

Delete | Add | << | >> | Page # | OK

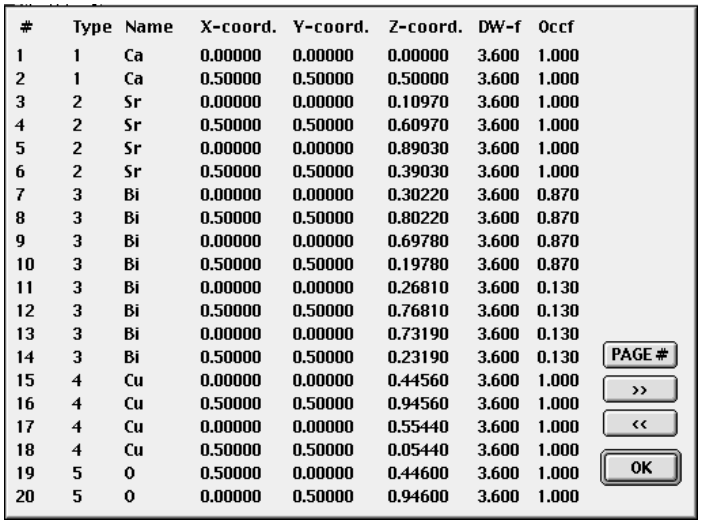
Symmetry Operators...
This brings up the list of symmetry operators either associated by the space group or entered manually by the user. The symmetry operators can be edited, and new ones may be added to

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.

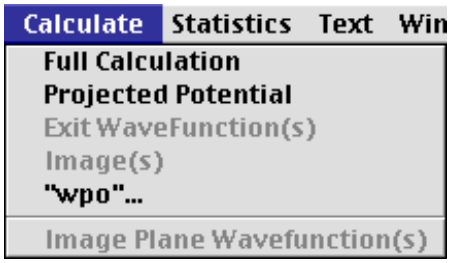


#	Type	Name	X-coord.	Y-coord.	Z-coord.	DW-f	Occf
1	1	Ca	0.00000	0.00000	0.00000	3.600	1.000
2	1	Ca	0.50000	0.50000	0.50000	3.600	1.000
3	2	Sr	0.00000	0.00000	0.10970	3.600	1.000
4	2	Sr	0.50000	0.50000	0.60970	3.600	1.000
5	2	Sr	0.00000	0.00000	0.89030	3.600	1.000
6	2	Sr	0.50000	0.50000	0.39030	3.600	1.000
7	3	Bi	0.00000	0.00000	0.30220	3.600	0.870
8	3	Bi	0.50000	0.50000	0.80220	3.600	0.870
9	3	Bi	0.00000	0.00000	0.69780	3.600	0.870
10	3	Bi	0.50000	0.50000	0.19780	3.600	0.870
11	3	Bi	0.00000	0.00000	0.26810	3.600	0.130
12	3	Bi	0.50000	0.50000	0.76810	3.600	0.130
13	3	Bi	0.00000	0.00000	0.73190	3.600	0.130
14	3	Bi	0.50000	0.50000	0.23190	3.600	0.130
15	4	Cu	0.00000	0.00000	0.44560	3.600	1.000
16	4	Cu	0.50000	0.50000	0.94560	3.600	1.000
17	4	Cu	0.00000	0.00000	0.55440	3.600	1.000
18	4	Cu	0.50000	0.50000	0.05440	3.600	1.000
19	5	O	0.50000	0.00000	0.44600	3.600	1.000
20	5	O	0.00000	0.50000	0.94600	3.600	1.000

PAGE #
>>
<<
OK

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.

“Wpo”
This will calculate the weak phase object images for a range of

resolutions specified by the user. See the introduction and the later chapter for information on the WPO Approximation and its use.

Image Plane Wavefunction{s)
This will calculate the complex wavefunction at the image plane based on only linear terms in the contrast transfer function, i.e. only interference between the central beam and scattered beams.

Statistics Menu

The current operations in this menu are:

Histogram
Displays the histogram distribution of an area selected by the selection tool, or of the entire image screen if selected through the Select All command.

Column Average
Shows an average intensity along the horizontal line defined by the rectangle chosen by the selection tool. The trace being calculated by the program corresponds to the average of the pixels defined by the width of the rectangle.
This menu determines the appearance of text drawn in the MacTempas image window. The following text attributes can be set:

Text Menu

Font
Size
Style
Left Justified
Center Justified
Right Justified
These are the standard Macintosh menu items for formatting the text produced by the Text Tool.

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. The use of the Option Key can make the windows invisible/visible.

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 the editor EDIT. 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 <structurename>.at is given below:

Line #	Parameter(s)	Meaning
1	Title	Arbitrary description of this structure
2	SpaceGroupNumber	Just as is says, one of the 230 spacegroups, (1-230) including 0.
3	a b c a b g	The lattice parameters and angles
4	Gmax	The maximum reciprocal lattice vector in the multislice calculation. 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 associated 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 symbol, x-,y-,z coordinates in relative units of the lattice vectors, Debye-Waller factor and occupancy factor.
9	The same as line 8 for atom number 2.	
10	The same as line 8 for atom number 3.	
.		
8+NBasis	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+NBasis	Voltage	Accelerating voltage (kVolt).
10+NBasis	Lh Lk	The center of the Laue circle in units of the h and k of the transformed reciprocal unit cell. (Real numbers).

Line #Parameter(s)	Meaning
11+NBasis	
Thickness	The specimen thickness or T1,T2,DT First thickness, last thickness, increm. The commas are required.
12+NBasis	
IPlot	Amplitudes to be stored as for possible plotting, (YES/NO).
13+NBasis	
ih ik il	The indices of the reflection to be stored, or if IPlot == NO then :
Defocus	Objective lens defocus or
D1,D2,DD	First defocus, last defocus, increment. The commas are required.
14+NBasis +NAmp	
ApertureRad.	Radius of the objective lens Aperture in units of Å-1.
15+NBasis +NAmp	
Ah Ak	The center of the objective lens aperture in units of h,k of the transformed reciprocal unit cell.
16+NBasis +Namp	
Oh Ok	The center of the optic axes in the same units as Ah,Ak.

Line #Parameter(s)	Meaning
17+NBasis +Namp s1,s2,s3	Symmetry operator number 1. An example is x+1/3,y+5/6,z+1/3. The commas are required.
.	
17+NBasis +Namp +NSymop istat	The calculation status of this structure. For a new structure this should be 1
18+NBasis +Namp +NSymop Vibration	Halfwidth of mechanical Vibration 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	x	y	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

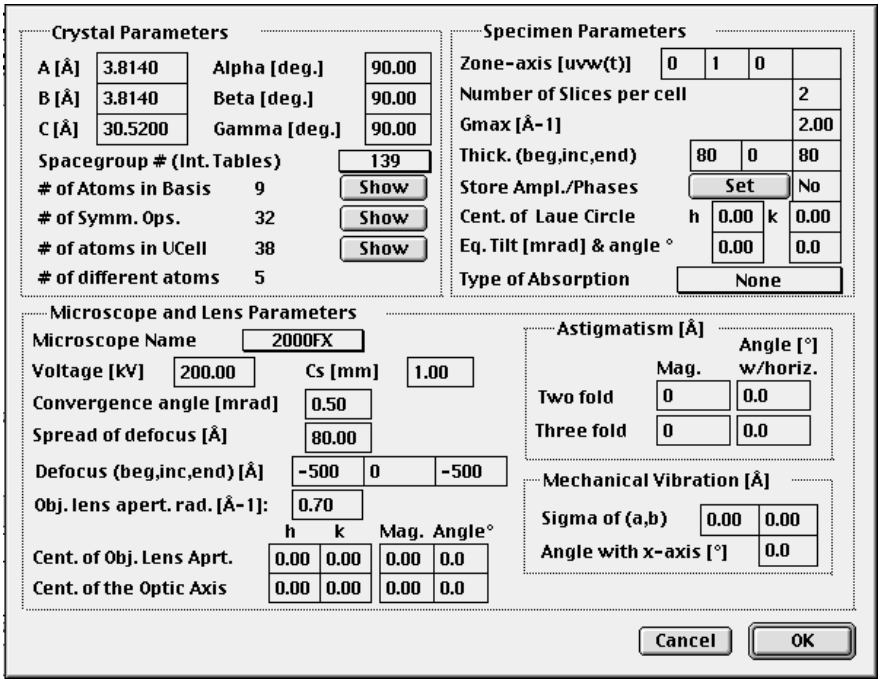
Isotropic thermal parameters for all atoms are fixed at 3.6 $Å^2$.

Entering the Structure

To enter a new structure into MacTempas, we first go to the **FILE menu** (Section 3.3), and select **New...** 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 many of the input parameters have default values. The only data that must be entered in ordered to create a valid file is the atoms in the basis.

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.



Comment BCSCO by Tarascon et al

MacTempas has a field for an optional comment or title. The

user should enter a comment such as the above, short enough to fit, yet detailed enough to jog the memory when referred to six months hence.

Space group # 139

From the structure information, we know that the cell is tetragonal 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 space-group from the popup menu.

a 3.814

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.

c 30.52

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. Note that MacTempas may be forced to choose a lower value of $\mathbf{G}_{max}$ if a

large unit cell is used. This occurs because MacTempas has a limit on the array dimensions used in the multislice calculations. Although this limit can be changed under **Preferences** , the current limit is in effect until the program is restarted. The array dimensions are required to be powers of two. An upper limit of 256 by 256 is limited by array dimensions to a parameter-16 multislice calculation (like 256 by 256, or 512 by 128, or 1024 by 64...) so that $\mathbf{G}_{max}$ for the diffracted beams is limited to 2n-2a* along one reciprocal-lattice coordinate and to 2m-2b* along the other, where n+m=16 (hence “parameter-16”). Note that MacTempas will choose values of n and m to maximize $\mathbf{G}_{max}$ up to the default value of 2.

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

Set Basis 9
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.

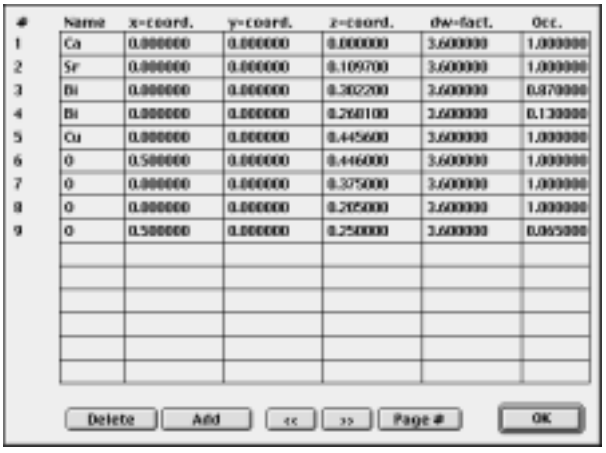
The second atom position is entered in the same way with responses of -

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
x,y,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.



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 half-width of depth of focus to 80Å, and the semi-angle of beam convergence to 0.5milliradian.

Specimen Thickness 40 80 20

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 No

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 (400)

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 0,0

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 -800 -200

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 imaging-condition 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 condition; that is, underfocus is negative.

Aperture Radius 0.67

The value for the radius of the objective aperture should correspond to the radius in reciprocal Ångstrøm units, as measured from a diffraction pattern exposed with the aperture superimposed. We will enter 0.67 to represent a typical value.

Center of the Objective Aperture 0,0

In order to simulate dark-field images, MacTempas provides for an objective aperture displaced from the center of the diffraction pattern. As for the Laue circle center, the aperture center is defined in units of h and k. We leave the default values of 0,0.

Center of the Optic Axis 0,0

To provide for microscope misalignment, or for conditions of tilted-beam imaging, the coordinates of the diffraction pattern at which the optic axis lies can be specified in the same manner as the center of the aperture. Again, we use default values of 0,0.

Verifying the Input

After the response to the last data-entry prompt, MacTempas draws the windows it uses. To re-display the input information

click on the "Main Parameters" in the **Parameters** menu. At this stage any desired changes can be made by using the mouse to move the cursor to the desired parameter, and making the change.

When all the data in the top field are satisfactory, we go to "Atomic Basis" in the **parameters** menu to check that all atom parameters have been entered correctly. At this stage it is also worthwhile getting MacTempas to display a model of the structure by.going to the **Commands** menu and clicking on "Draw the Unit Cell".

Running the Calculation

When we are satisfied that all data are correct, we run the simulation by.clicking on "Full Calculation" in the **calculate** menu. Note that MacTempas displays the current status of the calculation 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 (diffraction 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 **source** 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. To increase the area of image, keeping the magnification the same, select "#CELLS" in the **source** window, and double the size in the x direction by choosing 2 unit cells in this direction and one in the y direction; then

select "DISPLAY".

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 "DISPLAY".

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". Notice that the current values of "ZOOM" and "#CELLS" remain set from the last update.

To display the diffraction patterns at the stored specimen thicknesses, select "DIFFR.PATT" in the **source** window, then "DISPLAY". 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.

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

$$\Psi(x, y) = 1 - i\sigma t\Phi(x, y)$$

where $\Psi(x, y)$ is the electron wavefunction at a point (x,y) and $\Phi(x, y)$ 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 Scherzer Lens

An 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 $\Psi(x, y)$ above) causes all scattered beams that pass through the lens to be 180 degrees out of phase

with the central beam.

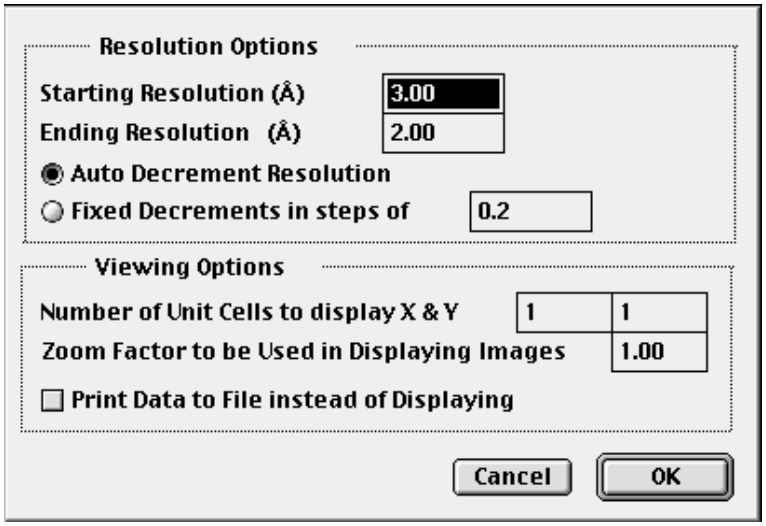
Under the two assumptions above, the image intensity in the WPO approximation can be written as

$$\Psi(x, y) = 1 - i\sigma t\Phi(x, y)$$

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

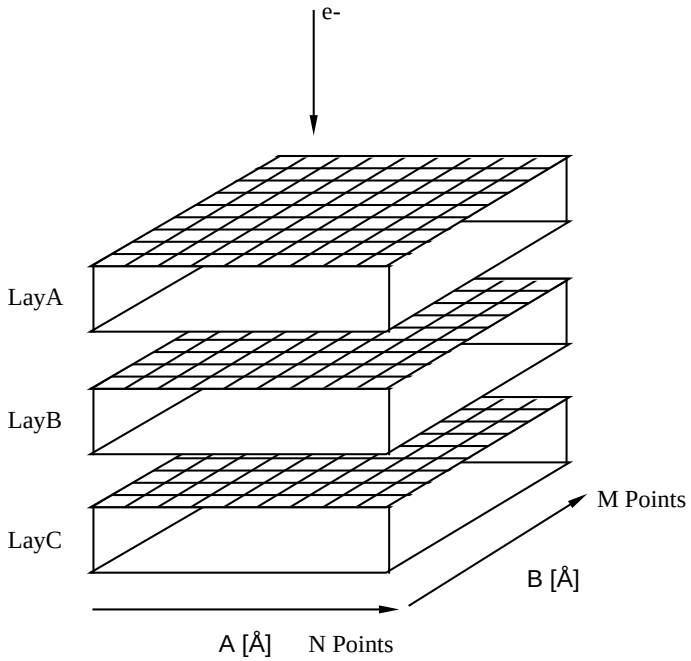
reached.


Resolution Options
Starting Resolution (Å)
3.00
Ending Resolution (Å)
2.00
Auto Decrement Resolution
Fixed Decrements in steps of
0.2
Viewing Options
Number of Unit Cells to display X & Y
1
1
Zoom Factor to be Used in Displaying Images
1.00
Print Data to File instead of Displaying
Cancel
OK

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 slice-thickness and the unit cell must be sub-divided into several



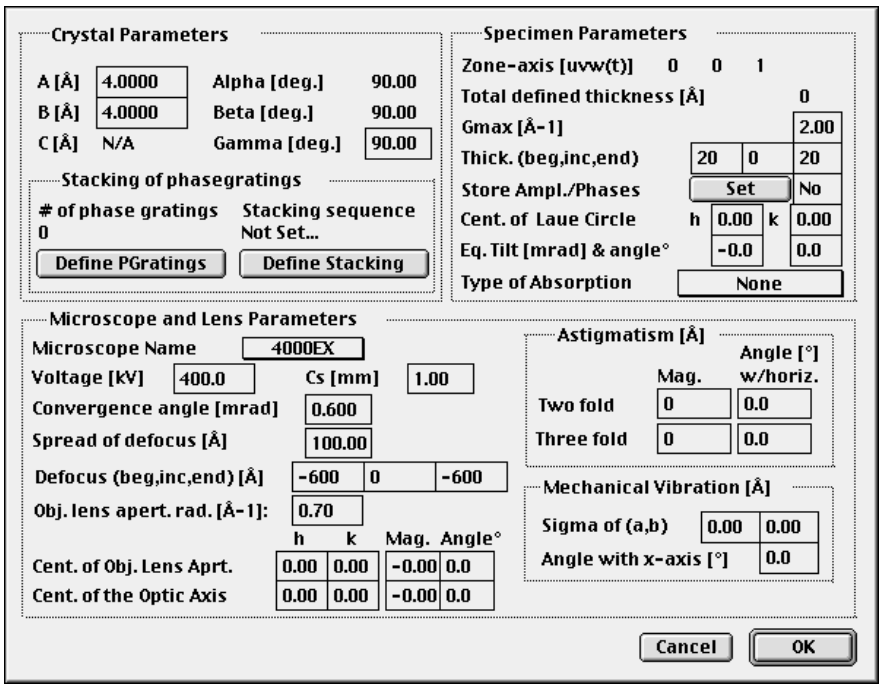
slices with different atomic content. As an example we will work with three layers which we will call LayA, LayB and LayC. Each of these "layers" are what we would call a "single" structure. That means they are defined as a unit cell with lattice parameters and atomic content. The one thing they have 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).

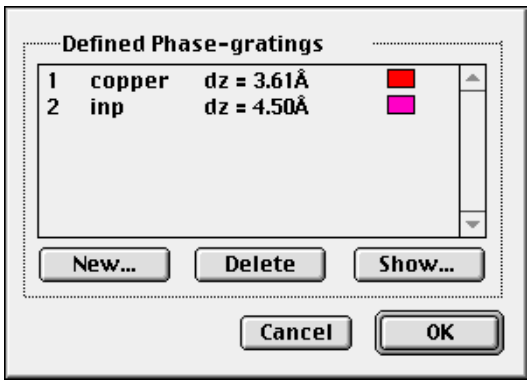
2) Calculate the phasegrating for each structure LayA, LayB and LayC using the same value for Gmax.

3) Now create a "**New** " Structure in MacTempas using the

option "**LayeredStructure** ". You will be asked to fill out infor-

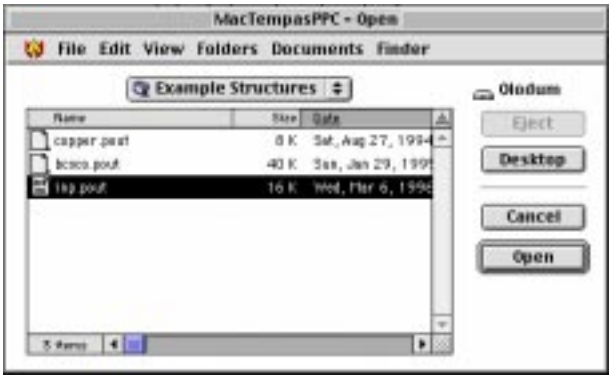


mation regarding the lattice parameters A and B etc. There are no input for atoms, because a layered structure has no atom information per se. Even though you are asked to fill out a specimen thickness, this value has no meaning at this time, because the content of the structure has not been defined.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

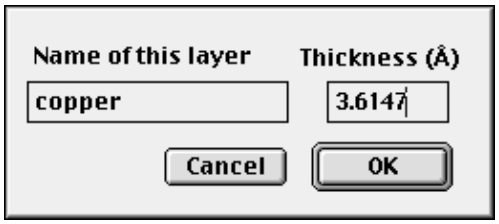


LayC. When you create the layered structure, a default value of 2 $Å^{-1}$ is supplied and you must change it in the main parameters if a different value was used in calculating the phase-gratings for LayA, LayB and LayC.

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 Phasegratings** ". 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-



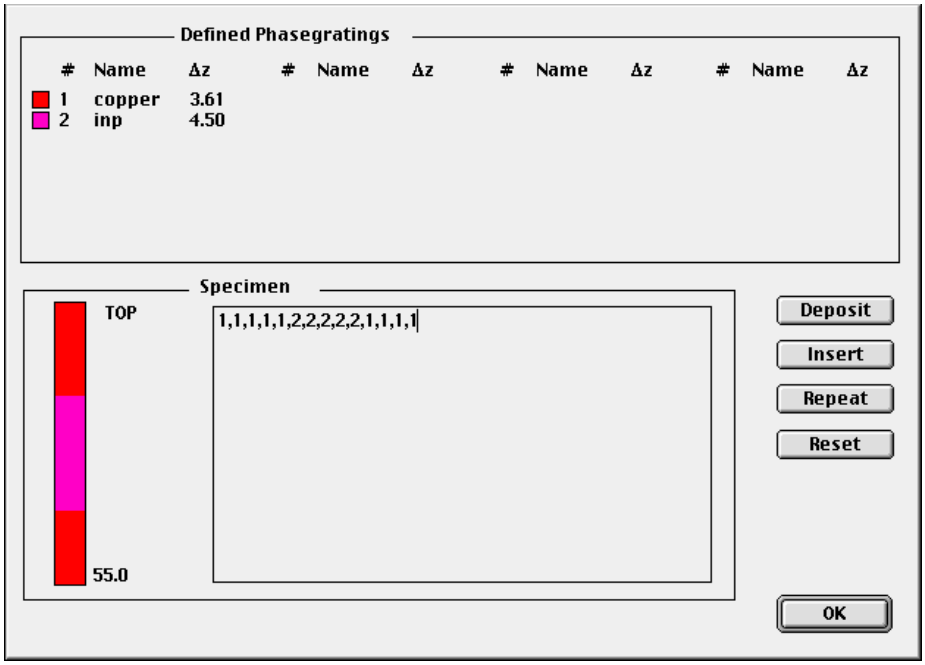
rating files (—.**pout**). Double Click on LayA.pout and fill in the value for the slice-thickness that was used in the calculation of LayA.pout. Continue and do the same for LayB and LayC.



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

1,1,1,1,1,1,1,2,2,2,2,2,2,23,3,3,3,3,3,1,1,1 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.



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

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 non-zero (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 "sub-slices". 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

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

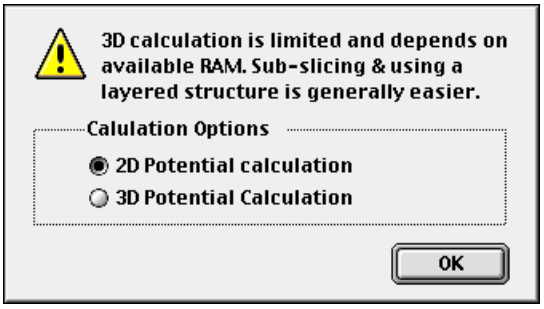
An improvement on sub-dividing the projected potential is to sub-divide the unit cell atom positions. In this procedure the list of atom positions within the unit cell is divided into n groups depending upon the atom position in the incident beam direction. From these sub-sliced groups, different projected 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 sub-slice $z_0 \pm \partial z$ 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-layer-lines by producing n identical sub-slices.

When a three-dimensional calculation is selected, (3D-Potential Calculation activated), MacTempas uses a sub-divided three-dimensional 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 N-beam 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.