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Diffraction imaging and diffuse scattering by small dislocation loops

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Abstract

Defining the limits of visibility of small defect clusters and dislocation loops, and optimal diffraction conditions for electron microscope imaging remains one of the central problems of electron microscopy of irradiated materials. Using computer image simulations based on the propagation–interpolation algorithm for solving the Howie–Basinski equations, we investigate the relation between the actual and the 'observed' size of small loops, the part played by many-beam dynamical diffraction effects, and limitations of electron microscope imaging in identifying the structure of small defects. We also discuss the link between real-space imaging and diffuse scattering by small dislocation loops.

1. Introduction

Electron microscopy of irradiated materials is arguably the only available method of visualizing defect structures formed under irradiation. For example, small dislocation loops and point-defect clusters in crystals are usually investigated using diffraction contrast images produced by transmission electron microscopy. For relatively large defects a combination of dynamical imaging and image contrast simulations has proven very successful for determining defect structures [1]. At the same time very small clusters are usually better seen under weak-beam diffraction conditions. Image simulations are necessary for a full analysis of such images [2,3].

In this paper we give a brief review of a recently developed propagation–interpolation algorithm for solving the Howie–Basinski equations [4] and its applications to simulating electron microscope images of small dislocation loops. We also outline the principles of observation of electron diffuse scattering by *individual* defects [6].

2. The propagation–interpolation algorithm

In many-beam dynamical diffraction theory the wave function $\psi(\mathbf{r}) = \psi(x,y,z)$ of high-energy electrons propagating through a thin foil is approximated by a sum of plane waves with slowly varying amplitudes $\phi_{\mathbf{g}}(\mathbf{r})$ as

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$$\psi(\mathbf{r}) = \sum_{\mathbf{g}} \phi_{\mathbf{g}}(\mathbf{r}) \exp[2\pi \mathrm{i}(\mathbf{k}+\mathbf{g}+\mathbf{s}_{\mathbf{g}}) \cdot \mathbf{r}]. \quad (1)$$

Here $\mathbf{k}$ is the wave vector of electrons incident on the foil and $\mathbf{s}_{\mathbf{g}}$ is the excitation error for the beam with diffraction vector $\mathbf{g}$. Vector $\mathbf{s}_{\mathbf{g}}$ is parallel to the zone axis $\mathbf{z}$ and its length is defined by the condition of energy conservation $\mathbf{k}^2 = |\mathbf{k}+\mathbf{g}+\mathbf{s}_{\mathbf{g}}|^2$. The potential of interaction between the high-energy electrons and the crystal is evaluated using the deformable ion approximation

$$V(\mathbf{r}) = \sum_{\mathbf{g}} V_{\mathbf{g}} \exp[2\pi \mathrm{i}\mathbf{g} \cdot (\mathbf{r} - \mathbf{R}(\mathbf{r}))], \quad (2)$$

where $\mathbf{R}(\mathbf{r})$ is the field of atomic displacements around a defect. In our simulations this field is assumed to be continuous and is evaluated either by using linear anisotropic elasticity [5] or by interpolating between discrete atomic positions found using molecular statics or molecular dynamics.

By inserting (1) and (2) into the Schrödinger equation and neglecting the second order derivatives, we arrive at the Howie–Basinski equations [4]

$$\begin{aligned}(\mathbf{k}+\mathbf{g}+\mathbf{s}_{\mathbf{g}}) \cdot \nabla\phi_{\mathbf{g}} = {} & -\mathrm{i}\pi U_0 \phi_{\mathbf{g}} - \mathrm{i}\pi \sum_{\mathbf{g}'} (1-\delta_{\mathbf{g}\mathbf{g}'}) \\ & \times U_{\mathbf{g}-\mathbf{g}'} \exp[2\pi \mathrm{i}(\mathbf{g}'-\mathbf{g})\mathbf{R}(\mathbf{r}) \\ & + 2\pi \mathrm{i}(\mathbf{s}_{\mathbf{g}'} - \mathbf{s}_{\mathbf{g}}) \cdot \mathbf{r}]\phi_{\mathbf{g}'}, \end{aligned} \quad (3)$$

where $U_{\mathbf{g}} = -(2m/h^2)V_{\mathbf{g}}$ and $h = 2\pi\hbar$ is the Planck constant. To eliminate the phase factors in Eq. (3) we apply a gauge transformation

$$\phi_{\mathbf{g}}(\mathbf{r}) = \Phi_{\mathbf{g}}(\mathbf{r}) \mathrm{e}^{-2\pi \mathrm{i}\mathbf{g}\cdot\mathbf{R}(\mathbf{r})} \mathrm{e}^{-2\pi \mathrm{i}\mathbf{s}_{\mathbf{g}}\cdot\mathbf{r}} \mathrm{e}^{-\mathrm{i}\pi \frac{U_0}{(\mathbf{k}+\mathbf{g}+\mathbf{s}_{\mathbf{g}})_z} z}. \quad (4)$$

The new amplitudes $\Phi_{\mathbf{g}}(\mathbf{r})$ satisfy equations

$$\begin{aligned}(\mathbf{k}+\mathbf{g}+\mathbf{s}_{\mathbf{g}}) \cdot \nabla\Phi_{\mathbf{g}} = {} & 2\pi \mathrm{i}(\mathbf{k}+\mathbf{g}+\mathbf{s}_{\mathbf{g}}) \cdot \mathbf{s}_{\mathbf{g}}^{(\mathbf{R})} \Phi_{\mathbf{g}} \\ & - \pi \mathrm{i} \sum_{\mathbf{g}'} (1-\delta_{\mathbf{g}\mathbf{g}'}) U_{\mathbf{g}-\mathbf{g}'} \Phi_{\mathbf{g}'}, \end{aligned} \quad (5)$$

where $\mathbf{s}_{\mathbf{g}}^{(\mathbf{R})} = \mathbf{s}_{\mathbf{g}} + \nabla[\mathbf{g} \cdot \mathbf{R}(\mathbf{r})]$ is an effective excitation error that varies spatially as a function of the distortion field $\partial R_i/\partial x_j$, where $i,j = 1,2,3$. Since $\phi_{\mathbf{g}}$ and $\Phi_{\mathbf{g}}$ in Eq. (4) differ only by a phase factor, the gauge transformation does not affect the intensities of the transmitted and diffracted beams and the simulated images. The lattice distortion introduced by a defect appears only in the local excitation error $\mathbf{s}_{\mathbf{g}}^{(\mathbf{R})}$. If the crystal undergoes a homogeneous (affine) transformation then $\mathbf{s}_{\mathbf{g}}^{(\mathbf{R})}$ is a constant, and Eq. (5) describes the diffraction from a homogeneously deformed crystal. This suggests that Eq. (5) may be solved numerically for an arbitrarily deformed crystal by dividing it into small cells and taking $\mathbf{s}_{\mathbf{g}}^{(\mathbf{R})}$ as a constant within each cell. Anomalous absorption is introduced phenomenologically by adding an imaginary part to the Fourier components of the potential [7,8].

If the column approximation is applied to the transformed equations (5) we neglect the components of $\nabla\Phi_{\mathbf{g}}$ perpendicular to the zone axis $\mathbf{z}$. In this case we arrive at the *modified* Howie–Whelan equations

$$\begin{aligned}\frac{\partial\Phi_{\mathbf{g}}}{\partial z} = {} & \frac{2\pi \mathrm{i}}{\beta_{\mathbf{g}}} (\mathbf{k}+\mathbf{g}+\mathbf{s}_{\mathbf{g}}) \cdot \mathbf{s}_{\mathbf{g}}^{(\mathbf{R})} \Phi_{\mathbf{g}} \\ & - \pi \mathrm{i} \sum_{\mathbf{g}'} (1-\delta_{\mathbf{g}\mathbf{g}'}) \frac{U_{\mathbf{g}-\mathbf{g}'}}{\beta_{\mathbf{g}}} \Phi_{\mathbf{g}'}, \end{aligned} \quad (6)$$

where $\beta_{\mathbf{g}} = (\mathbf{k}+\mathbf{g}+\mathbf{s}_{\mathbf{g}})_z$.

In principle, solving Eq. (5) numerically requires integrating these equations along the characteristics defined by the directions of propagation of diffraction beams $\mathbf{k}+\mathbf{g}+\mathbf{s}_{\mathbf{g}}$. The algorithm developed here replaces propagating solutions along the characteristics by a sequence of two-step events, where the first step involves solving the modified Howie–Whelan equations (6) for a thin slice within a set of adjacent narrow columns, and the second step corrects the solution for the effect of inclined propagation of the beams by means of interpolating between values found at the first step for the adjacent columns. It can be proven [10] that in the limit of small slice thickness and small column width a solution found using the propagation–interpolation algorithm is equivalent to the solution found by integrating the Howie–Basinski equations.

What are the advantages of the approach described above over the existing methods of image simulations [9,3]? On the one hand, the new algorithm makes it possible to simulate images of three-dimensional defect structures (see, e.g. [2]) while the earlier solutions of the Howie–Basinski equations only addressed the case of infinite straight dislocations [9]. In comparison with the multislice algorithms [3] the method is more flexible and is able to use as input the distortion field $\partial R_i/\partial x_j$ evaluated using either linear elasticity or atomistic simulations. Also, the structure of Eq. (5) makes it possible to select, at the start of a simulation, a set of $\mathbf{g}$-vectors that contribute to the formation of the image, therefore avoiding using a large number of virtual reflections required for carrying out a multislice simulation. Last but not least, the simplicity of the propagation–interpolation algorithm