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ALPHA-NULL DEFOCUS: AN OPTIMUM DEFOCUS CONDITION WITH RELEVANCE FOR FOCAL-SERIES RECONSTRUCTION

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Two optimum defocus conditions are well known to users of high-resolution transmission electron microscopes. Scherzer[1] defocus is useful in high-resolution electron microscopy (HREM) because it produces an image of the specimen "projected potential" to the resolution of the microscope[2]. Lichte[3] defocus is useful in electron holography because it optimizes sampling in frequency-space by minimizing the slope of the microscope objective lens phase change out to the highest spatial frequency in the hologram, consequently minimizing dispersion. For focal-series reconstruction, the requirement to maximize transfer into the image of high-frequency diffracted beam amplitudes leads to a third optimum defocus condition[4].

Image reconstruction methods allow the achievement of super-resolution – resolution beyond the native (Scherzer) resolution of the microscope – by correction of the phase changes introduced by the microscope objective lens[5]. One such method is focal-series reconstruction, in which diffracted-beam information obtained at several different focus values is combined[6]. To produce a valid super-resolution result, it is necessary to ensure that every spatial frequency is represented appropriately. Suitable choice of an optimum defocus produces optimum transfer of diffracted-beam amplitudes at any chosen spatial frequency.

Same-phase transfer of diffracted-beam amplitudes is maximized at Scherzer defocus, $\varepsilon_S = -\sqrt{(3C_S\lambda/2)}$, but only extends to the Scherzer resolution of $d_S = 0.64C_S^{1/4}\lambda^{3/4}$, where λ is electron wavelength and C_S is the objective lens spherical aberration coefficient. Mixed-phase transfer of diffracted-beam amplitudes can occur beyond Scherzer resolution, out to the microscope information limit[7] of $d_\Delta = \sqrt{(\pi\lambda\Delta/2)}$. Here Δ is the standard deviation of microscope spread of focus occasioned by partial temporal coherence of the electron beam, $\Delta = C_C\sqrt{\{(\sigma^2(V)/V^2 + 4\sigma^2(I)/I^2 + \sigma^2(E)/E^2\}}$, where $\sigma^2(V)$, $\sigma^2(I)$, $\sigma^2(E)$ are the variances in high-voltage, lens current and energy spread over the time scale of image acquisition, and C_C is the chromatic aberration coefficient of the objective lens. Although partial spatial coherence does not contribute to the information limit of the microscope, it does limit transfer of diffracted-beam amplitudes into any single image. The envelope for incident beam convergence has the form $E_\alpha(\mathbf{u}) = \exp\{-\pi^2\alpha^2(\varepsilon+\lambda^2C_S\mathbf{u}^2)^2\mathbf{u}^2\}$. For an incident beam convergence semi-angle[8] of 1.3α, the cutoff spatial frequency is given by[9]

$$|\mathbf{u}|_\alpha = S_+^{1/3} + S_-^{1/3} \qquad \text{where} \qquad S_\pm = \left[\frac{3.3}{4\pi\alpha} \pm \sqrt{\frac{\varepsilon^3}{27C_S\lambda^2} + \left(\frac{3.3}{4\pi\alpha}\right)^2}\right] / C_S\lambda^2$$

Because the import of the above expression is obscure, it is more useful to consider the action of incident-beam convergence as a function of defocus, ε, for any particular spatial frequency, $|\mathbf{u}|$. The phase imposed on diffracted-beam amplitudes is $\chi(\mathbf{u}) = \pi\varepsilon\lambda|\mathbf{u}|^2 + \pi C_S\lambda^3|\mathbf{u}|^4/2$. Transfer of beam hkl varies with defocus as $\sin\{\pi\varepsilon\lambda|\mathbf{u}_{hkl}|^2\}$, with an offset from zero defocus of $\sin\{\pi C_S\lambda^3|\mathbf{u}_{hkl}|^4/2\}$. Incident-beam convergence confines this transfer to a Gaussian "packet" of defocus values centered on the alpha-null defocus. For a diffracted beam hkl, with a spatial frequency of u_{hkl}, the envelope for incident beam convergence has null damping effect when defocus is $\varepsilon_\alpha^o(u_{hkl}) = -C_S(\lambda u_{hkl})^2$. On either side of this alpha-null defocus value, the damping effect of incident-beam convergence reduces the diffracted-beam transfer to exp(-2), or 13.5%, when defocus reaches a value of $\varepsilon_\alpha^o(u_{hkl}) \pm \varepsilon_\alpha^{2\sigma}(u_{hkl}) = -C_S(\lambda u_{hkl})^2 \pm \sqrt{2}/(\pi\alpha u_{hkl})$. Thus the position of alpha-null defocus for any particular spatial frequency depends only on the value of C_S – on the other hand, defocus-packet width around the alpha-null defocus depends only on the convergence semi-angle.

Focal-series reconstruction is facilitated when defocus-packets for all diffracted beams overlap. To image carbon atoms at the correct positions in a [110] "dumbbell" image from diamond, the super-resolution image must include contributions from four sets of diffracted beams out to the 004 spacing of 0.89Å[10]. Transfer at the four diffracted-beam frequencies is confined to four defocus packets centered on the alpha-null defocus of each spatial frequency. Over a defocus range of 0 to –5000Å (fig.1), offsets of the defocus packets to their alpha-null positions are larger and their packet widths narrower (making overlap more difficult) for smaller spacings (higher frequencies). Reduction in C_S from 0.9mm (left column) to 0.6mm (right) reduces the alpha-null offset and improves overlap. Reduction in convergence from 1.0 milliradian (top row) to 0.5milliradian (center) to 0.25milliradian (bottom) increases packet widths and improves overlap.

Under NCEM OÅM (one-Ångstrom microscope[11]) conditions, reconstruction gives a [110] diamond image with the correct 0.89Å spacing (fig.2a). With the effects of 20Å focus spread included (fig.2b), the alpha-null defocus (arrow) is seen as the optimum defocus for transfer of all diffracted information out to 0.89Å. The alpha-null defocus should be included as the (furthest underfocus) limit for all focal series reconstruction. [12]

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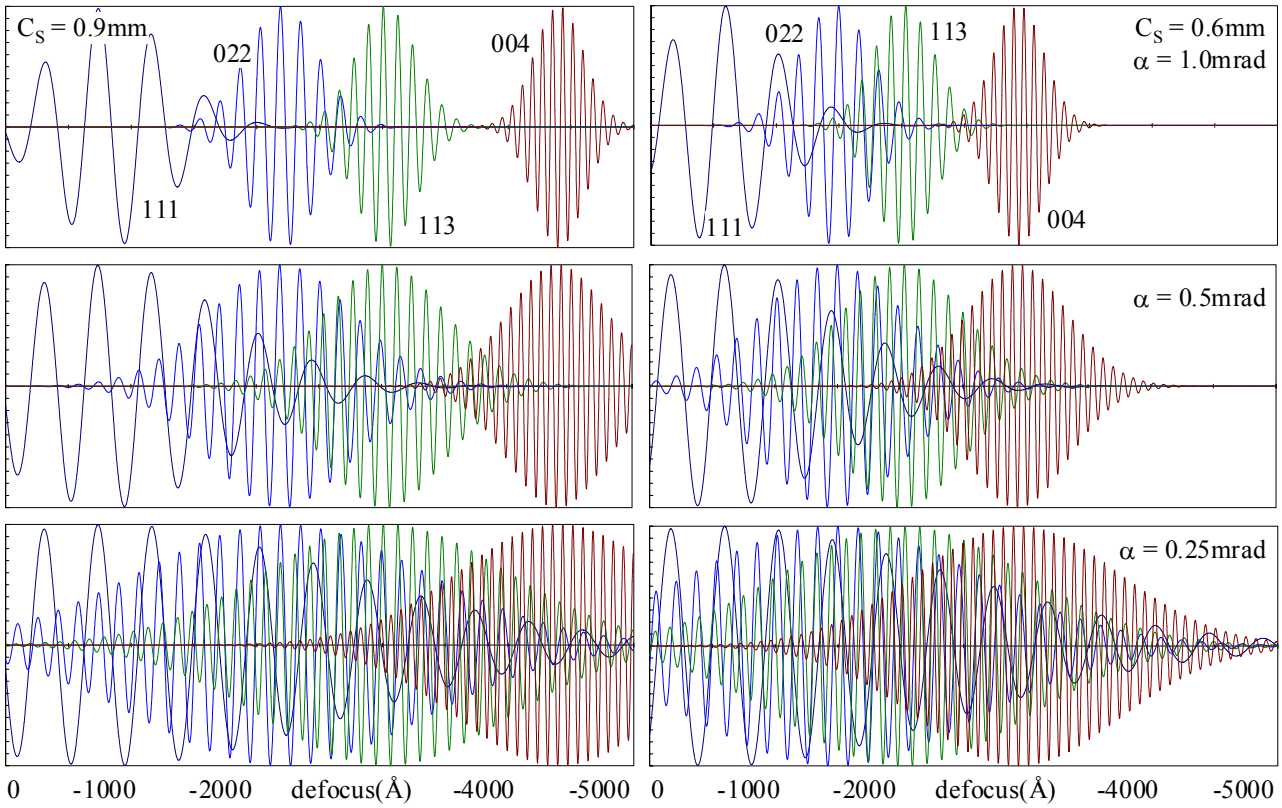

Fig.1 Defocus packets for decreasing spherical aberration C_S (across) and decreasing convergence α (down) show increasing transfer of [110] diamond diffracted beams. Plotted over the defocus range 0 to –5000Å.

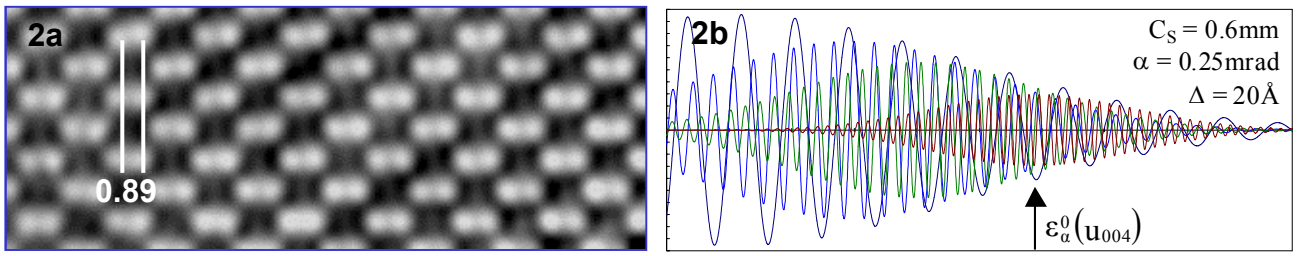

Fig.2. (a) Reconstruction from experimental OÅM focal series shows 0.89Å spacing of C-C "dumbbells". (b) Under OÅM conditions transfer is optimized at the alpha-null defocus for the diamond 004 beam (arrow).