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Defining HRTEM Resolution: Why Young's Fringes Don't Determine Resolution

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Aberration correction of the objective lens in the high-resolution transmission electron microscope (HRTEM) can extend microscope resolution to the linear information limit [1], the upper bound of a HRTEM's structural resolution. HRTEM resolution is demonstrated canonically in real space when two image peaks (representing atoms) are distinguishable from a single peak – just as Rayleigh's criterion establishes when two sources of light (stars) are distinguishable from a single source [2]. The "A-OK" set of specimens can be employed to measure HRTEM resolutions from 1.6Å to 0.2Å unequivocally in real space [3] and to characterize image resolution quality [4]. Such real-space measurements have established sub-Ångström resolutions of 0.78Å [5-6] and 0.63Å [7]. Despite the straightforward approach of resolution quantification in real space, resolution is often gauged using image intensity spectrum measurements in reciprocal space, probably because resolution $|\mathbf{d}|$ requires the corresponding spatial frequency $1/|\mathbf{d}|$ in the TEM image intensity spectrum; however, the mere presence of a $1/|\mathbf{d}|$ frequency does not establish a resolution of $|\mathbf{d}|$ [8].

Spatial frequency transfer from specimen exit surface to microscope image can be determined in reciprocal space from diffractograms of images of amorphous specimens. To distinguish signal from noise in such diffractograms, it is possible to map the signal fadeout with spatial frequency by adding two successive images, obtained under the same conditions except for a small relative shift of one to the other. The forward Fourier transform (FFT) of these added images contains Young's fringes that fade out at the same spatial frequency as the correlated signal – the signal is the same in both images, but the noise is different. Image formation theory confirms that Young's fringes track both linear and non-linear intensity components in the image, so Young's fringes measure the extent of the information that is common to both images, but do not discriminate between linear contributions (present in the image out to its resolution) and non-linear contributions that may extend to much higher spatial frequency [8]. Thus, the assumption that the range of fringes in Young's-fringe diffractograms is a dependable measure of the microscope's linear information limit can lead to significant overestimates of HRTEM information limit [9].

Image intensity, $I(\mathbf{x}) = \psi(\mathbf{x}).\psi^*(\mathbf{x})$, gives an image intensity spectrum $I(u) = \Psi(u) \odot \Psi^*(-u)$, where $\odot$ represents convolution and $\Psi(\mathbf{u}) = \mathcal{F}\{\psi(\mathbf{x})\}$ is the FFT of $\psi(\mathbf{x})$. Limited temporal and spatial coherence (focus spread Δ and beam convergence α) produce a transmission cross-coefficient (TCC) of $D(u',u'-u) = B(u',u'-u).C(u',u'-u)$, where $B(u',u'-u) = \exp\{-\frac{1}{2}\pi^2\lambda^2\Delta^2(u'^2-(u'-u)^2)^2\}$ is the TCC due to limited temporal coherence; $C(u',u'-u) = \exp\{-\pi^2\alpha^2[\varepsilon u + C_S\lambda^2(u'^3-(u'-u)^3)]\}$ is the TCC due to limited spatial coherence; here λ is electron wavelength, ε is defocus, and spherical aberration coefficient is C_S [8]. The image intensity spectrum is then $I_{\alpha\Delta}(u) = \sum_{u'} \Psi(u') \cdot \Psi^*(u'-u) \cdot D(u',u'-u)$.

The function $D(u', u'-u)$ reveals differences between the linear information limit and the non-linear Young's fringe limit. For linear image contributions, u' or $(u'-u)$ is zero, the spread-of-focus TCC function collapses to the spread-of-focus envelope function $B(u) = \exp\{-\frac{1}{2}\pi^2\lambda^2\Delta^2u^4\}$, and temporal coherence determines the upper (information) limit to linear transfer. When neither term has zero spatial frequency, the resultant interference in the intensity spectrum will contribute at a spatial frequency equal to the vector difference of the contributing terms. For non-linear image contributions where $u'+(u'-u) = 0$, Young's fringes are undamped by the temporal coherence TCC, the frequency u in the intensity spectrum is twice that of the specimen frequency, and transfer is damped by the convergence envelope function $C(u) = \exp\{-\pi^2\alpha^2(\varepsilon u + C_S\lambda^2u^3)\}$. The two limits are independent. Microscope information limit (MIL) is set by temporal coherence spread of focus. Young's fringe limit (YIF limit) is set by spatial coherence incident beam convergence.

We report experiments on a JEOL JEM-2100F confirming our previous theoretical examination of Young's-fringe limits [10]. Figure 1 compares fringes under non-linear (a) and linear (b) imaging conditions. YIFs created by non-linear imaging of thick amorphous carbon film extend to 0.6Å (a), but YIFs from weak-phase-object images of thin amorphous Ge film fade out at the 1Å information limit; the presence of Thon CTF rings confirms linear imaging (b). The 60% overestimate of information limit in (a) is significantly larger than previously found at 200kV [11]. Belief that "Young's fringes determine information limit" is as mistaken as the myths that HRTEM images "do not show atom positions" [12], that to find Scherzer imaging conditions "requires knowledge of the atom species and positions" [13], and that information limit is not determined by coherence [14]. After improving the coherence of the CM300/OÅM and demonstrating the ensuing increase in information limit to 0.78Å [5,6,15], one of us discovered a coworker's report claiming to have "extended the information limit from 1.05Å to 0.78Å by developing software for focal series reconstruction" [14]. In real life, true improvements to information limit require actual physical modifications [16].

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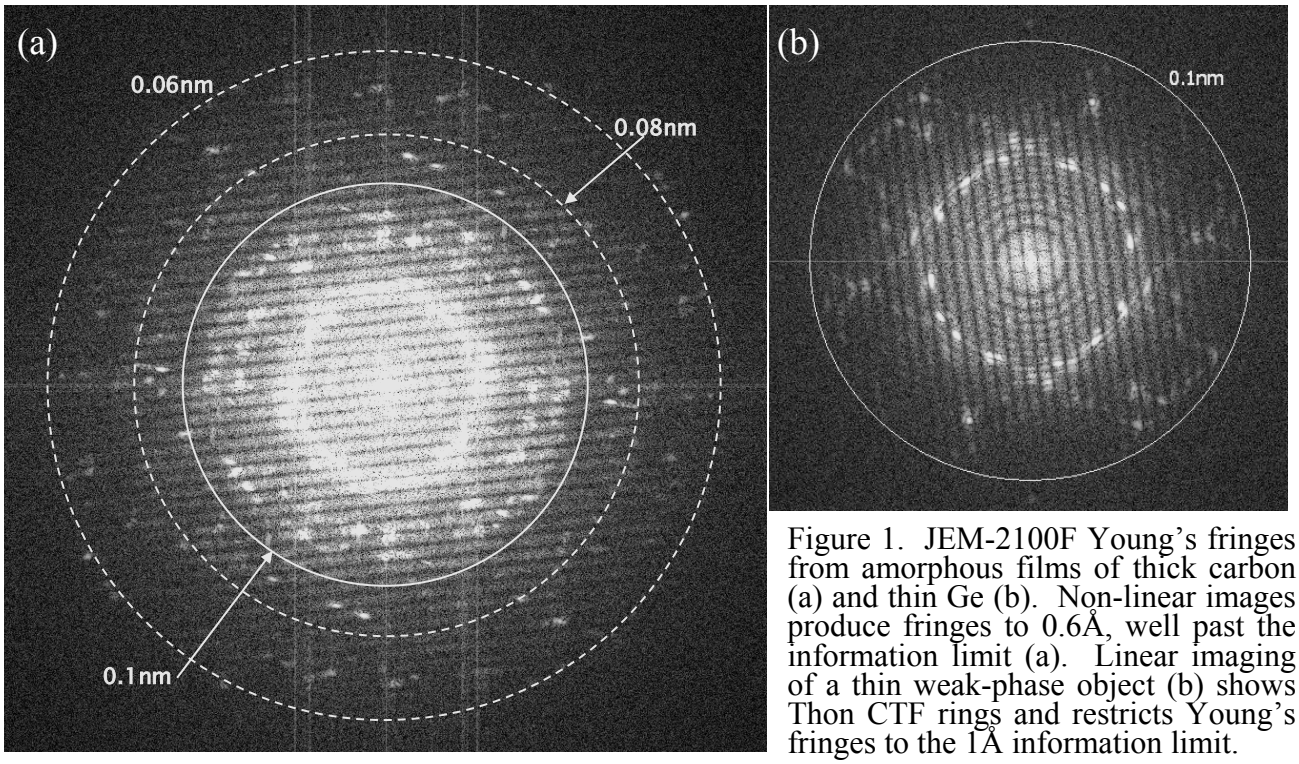

Figure 1. JEM-2100F Young's fringes from amorphous films of thick carbon (a) and thin Ge (b). Non-linear images produce fringes to 0.6Å, well past the information limit (a). Linear imaging of a thin weak-phase object (b) shows Thon CTF rings and restricts Young's fringes to the 1Å information limit.

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