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Effects of Quantized, Transient Chromatic Aberrations in Ultrafast Electron Microscopy

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High-resolution *in situ* electron microscopy is currently an extremely active area of research owing, in part, to the development and advancement of new instrumentation and methods for studying specimens at the atomic scale in liquids, at elevated temperatures and pressures, and under electrical biasing and during mechanical deformation. Notably, the increased experimental flexibility associated with employment of spherical aberration-correction systems – that is, relaxed requirements on accelerating voltages and pole-piece gaps – has facilitated studies of atomic-scale dynamic processes occurring under *operando* conditions with millisecond temporal resolution [1,2]. However, wide ranges of atomic-scale structural dynamics occur on timescales much shorter than one millisecond, the motions of which therefore cannot be resolved with current digital detector technologies.

Extension of the capabilities of electron microscopy to the ultrafast temporal regime is made possible by application of the pump-probe methodology in which a pulsed-laser is used to generate discrete photoelectron packets from the emission source to probe the specimen at a precisely controlled delay after optical excitation (pump). This approach, known as ultrafast electron microscopy (UEM), has been used to investigate dynamic phenomena via sub-picosecond temporally-resolved imaging, diffraction, and spectroscopy [3]. As in conventional TEM, the spatial resolution of UEM is dependent upon the coherence and current of the beam emanating from the electron source. One strategy for optimizing the coherence of photo-generated beams relies on populating each packet with, on average, a single electron. This method circumvents Coulombic space-charge broadening at the electron source (i.e., the Boersch effect), and the resulting beam coherency can be comparable to that observed during conventional thermionic emission [4]. This has been demonstrated via pulsed imaging (i.e., employing discrete photoelectron packets without pumping the specimen) of lattice fringes in various specimens and illustrates the potential for resolving real-space, angstrom-scale ultrafast dynamics with UEM [5,6].

While the single-photoelectron approach circumvents deleterious space-charge effects, the narrow energy spread can be compromised when the photon pulses and the electron packets are spatiotemporally overlapped at the specimen; light absorption and emission can occur in the specimen near-field, thus generating large populations of photoelectrons (relative to the elastically-scattered electrons) having discrete energies differing by integer multiples of photon quanta [7]. This results in a radially symmetric projection of the discrete electron energy distribution in the image plane due to the velocity dependence of the Lorentz force and, consequently, produces annular chromatic aberrations in the real-space intensity distributions. Here, we describe how the induced electron-energy envelope resulting from absorption and emission of photons in the optically-pumped specimen near field becomes the dominant effect limiting spatial resolution of UEM images during initial excitation. Moreover, select spatial frequencies become exaggerated due to the quantized nature of the near-field interaction. Application of the effect to representative lattice-fringe images of model nanostructures indeed limits resolving power and suggests that this transient, quantized chromatic aberration will need to be addressed in order to achieve angstrom-scale, real-space ultrafast imaging with UEM in the initial

moments of specimen excitation.

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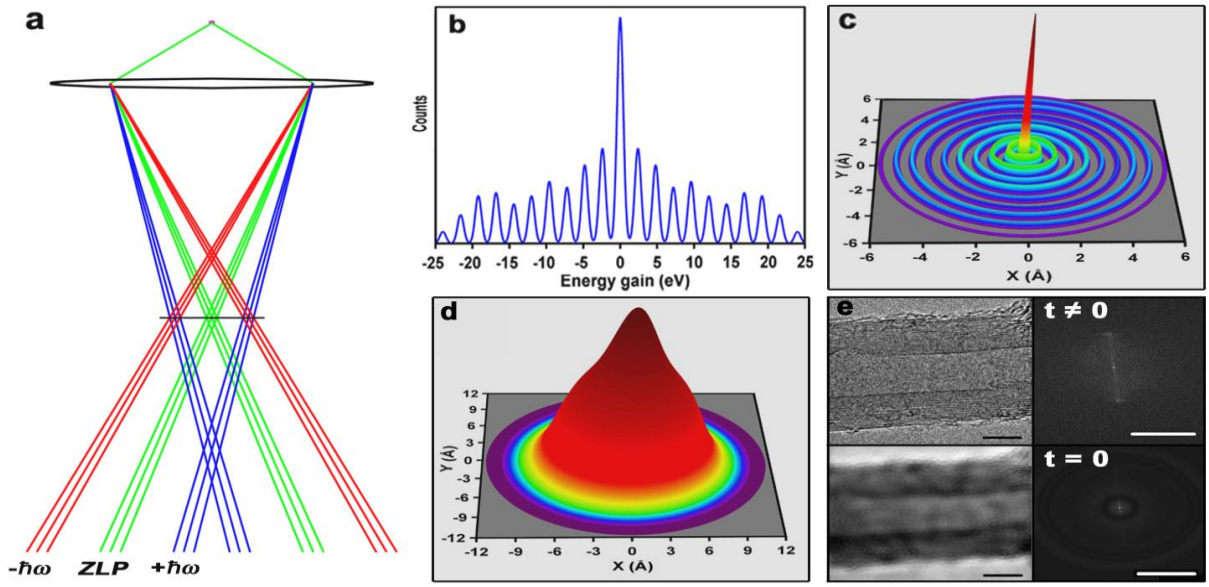

Figure 1. (a) Conical ray diagram depicting focusing of electrons of quantized energies ($\pm\hbar\omega$) arising from absorption and emission of photons in the specimen near field. Rays corresponding to elastically-scattered electrons (ZLP) are shown for reference. **(b)** Calculated low-loss EEL spectrum occurring at $t = 0$ (i.e., precise spatiotemporal overlap of the laser pulse and electron packet at the specimen). Each sideband occurs at integer multiples of the incident photon energy. **(c)** Resulting annular chromatic point-spread function (PSF) at $t = 0$. **(d)** Overall PSF shown in **(c)** and the effects of spherical aberration. The observed shoulder arises from the annular chromatic PSF. **(e)** Result of applying the annular chromatic PSF to a TEM bright-field image wherein lattice fringes are observed. The image (with FFT) is representative of an expected UEM image at $t \neq 0$, while the $t = 0$ image is blurred due to the quantized, chromatic aberration (scale bars: 5 nm, 2.5 $Å^{-1}$).