

Ultrafast Electron Microscopy: The Problem of Spatial Coherence

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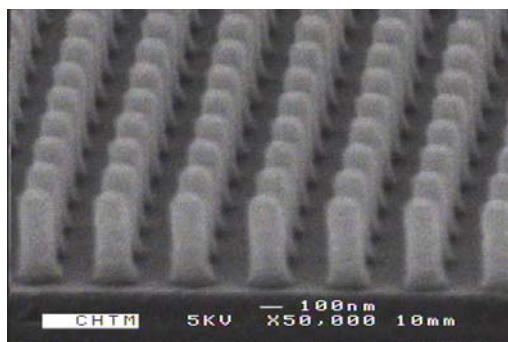
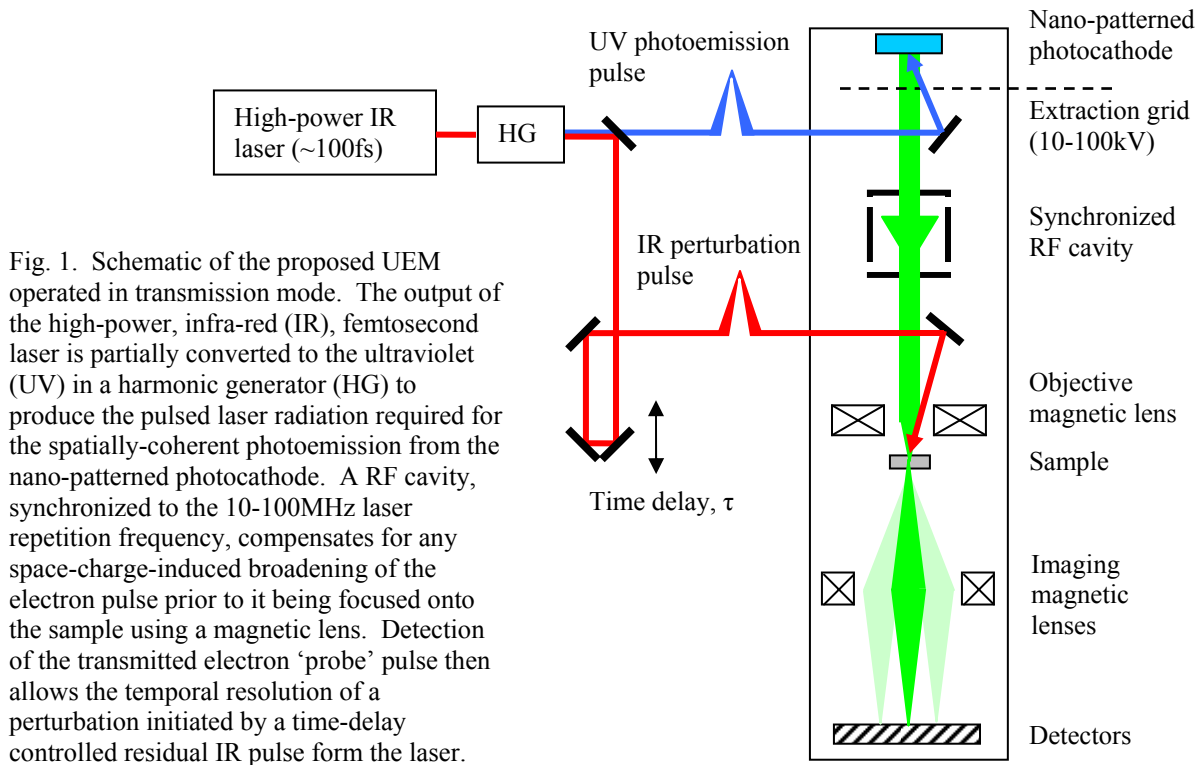
Several efforts [1-3] are currently underway to develop an ultrafast electron microscope (UEM): that is, to combine the atomic-scale resolution of electron microscopy with the sub-picosecond ($<10^{-12}$ s) temporal resolution now standard in ultrafast laser spectroscopy. In such an instrument, an ultrashort (ideally femtosecond) pump laser pulse initiates a dynamic transient event in a sample that is observed and time-resolved by a synchronized sub-ps electron pulse (Figure 1). The resulting unprecedented spatio-temporal resolution could allow for the direct visualization of the dynamic properties of *individual* nanoparticles and, in the future, individual atomic motions, thus providing an important new tool for sensing materials and molecular dynamics on the nanoscale [3]. To date, however, UEMs have operated almost exclusively as a dynamic transmission electron microscope (DTEM) in the diffraction mode; i.e., monitoring ensemble atomic motions in the Fourier plane as in ultrafast electron diffraction [4]. The inability to produce high spatial resolution images in a UEM can be traced to the lack of adequate spatial coherence in the electron pulse.

Ideally, in a UEM, the quantum state function Ψ for N individual electrons propagating in different directions \mathbf{k}_j with different energies $E_j = \hbar\omega_j$ and phases ϕ_j , should tend to that of a pulse propagating in one direction k , with one energy $E = \hbar\omega$, and with a well defined (i.e., spatially phase-coherent) amplitude $\Psi_0(\mathbf{r}, t)$; that is

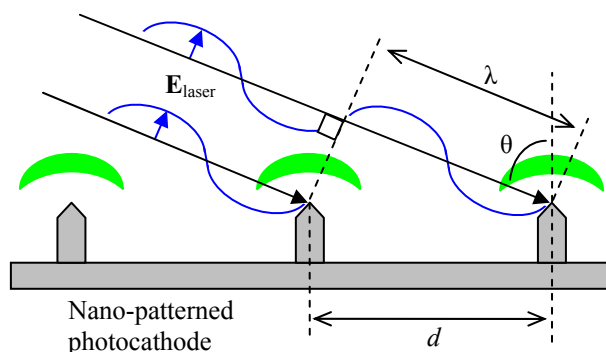
$$\Psi = \sum_{j=1}^N \exp[i(\mathbf{k}_j \cdot \mathbf{r} - \omega_j t + \phi_j)] \approx \Psi_0(\mathbf{r}, t) \exp[i(kz - \omega t)] .$$

The problem is that the electrons photoemitted from the laser-driven photocathode, in general, will not be generated with the same energy, wave vector \mathbf{k} , and phase. Moreover, even if a spatially-coherent electron pulse is emitted from the photocathode, space-charge effects (i.e., repulsive electron-electron scattering) will destroy the spatial coherence [2] and temporally broaden the pulse [5,6]. While the latter effect could be compensated for by the use of a synchronized RF cavity (Figure 1), the destruction of the spatial coherence will be detrimental to the spatial resolution of a UEM. Fortunately, a detailed numerical investigation into the space-time dynamics of electron pulses has revealed that for 'disk-like' pulses (i.e., transverse radial dimension much larger than the length of the pulse in the propagation direction) space-charge effects predominantly affect the temporal duration of the electron pulse upon propagation down the column. Thus, if the electron pulse can be photoemitted with a degree of spatial coherence, there is a good chance that electron-electron scattering will not substantially degrade the spatial coherence over the first few nanoseconds of propagation time.

Figure 2 illustrates the method that is currently being investigated as a means to generate a spatially coherent electron pulse from a nano-patterned photocathode. An array of silicon nanoposts (field emitters) is irradiated by an intense ultrashort laser pulse in such a manner to ensure that the phase of the incident laser field is the same at each nano-emitter. The resulting 'phase-control' of the photoemission could yield a set of coherent Huygen's electron wavelets to constitute an initially spatially coherent electron pulse required for the UEM (Figure 1) to achieve sub-nm resolution [7].



(a)



(b)

Fig. 2. (a) Cross-section SEM image of an array of 110nm silicon nanoposts with a pitch of 360 nm manufactured using laser interference lithography (courtesy of R. Bommena, University of New Mexico). (b) Schematic of the irradiance condition ($\lambda = d \sin \theta$) at wavelength λ proposed to achieve laser-driven phase-coherent electron photoemission (green wavelets) from a nano-patterned photocathode with spatial period d .

References

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