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# Ultrafast Electron Mircoscopy: 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 $\Psi$ for $N$ individual electrons propagating in different directions $\mathbf{k}_j$ with different energies $E_j = \hbar\omega_j$ and phases $\phi_j$, 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

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

The problem is that the electrons photoemitted from the laser-driven photocathode, in general, will not be generated with the same energy, wave vector **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 form 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].

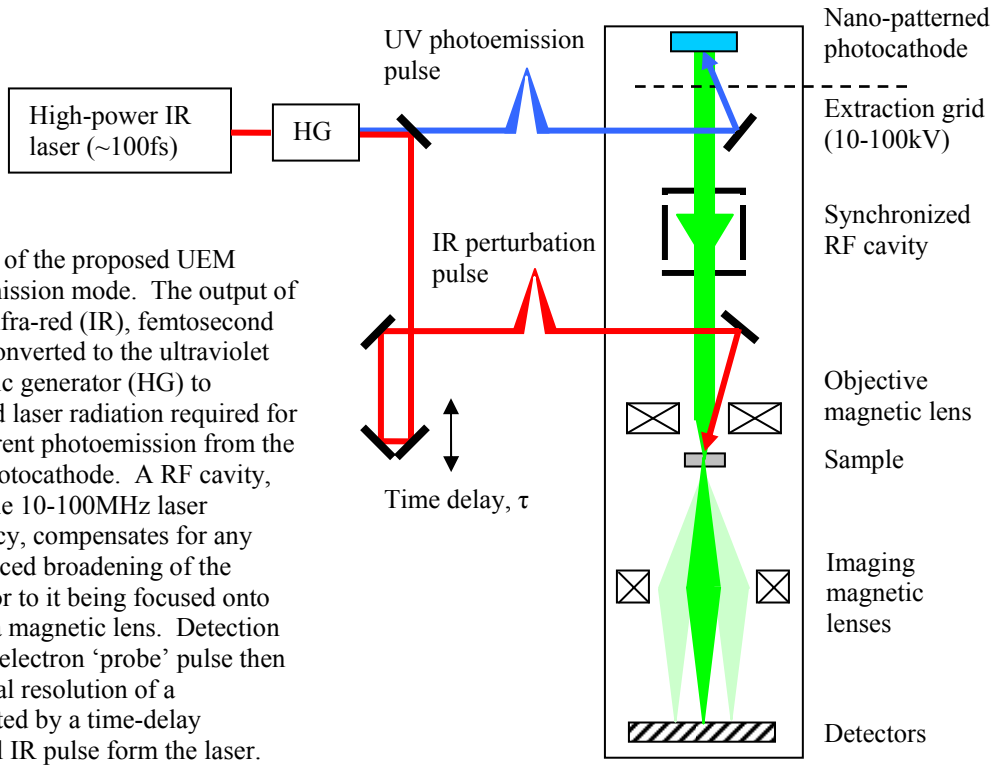
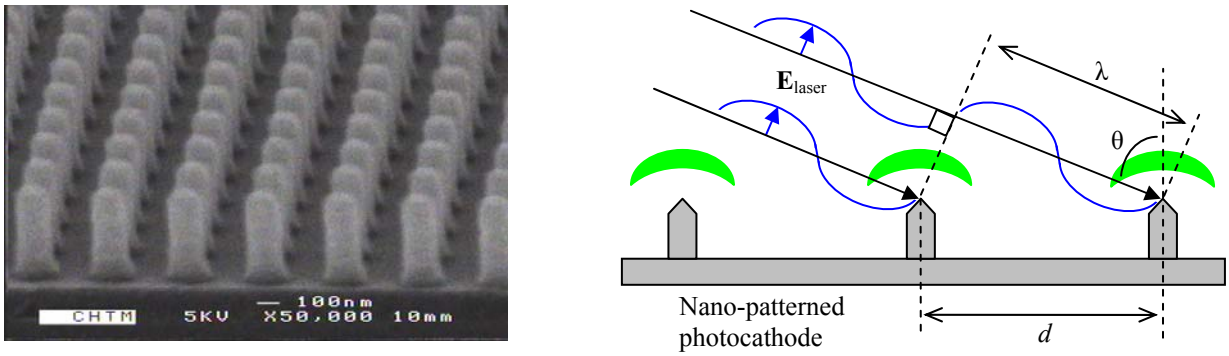



Fig. 1. Schematic of the proposed UEM operated in transmission mode. The output of the high-power, infra-red (IR), femtosecond laser is partially converted to the ultraviolet (UV) in a harmonic generator (HG) to produce the pulsed laser radiation required for the spatially-coherent photoemission from the nano-patterned photocathode. A RF cavity, synchronized to the 10-100MHz laser repetition frequency, compensates for any space-charge-induced broadening of the electron pulse prior to it being focused onto the sample using a magnetic lens. Detection of the transmitted electron 'probe' pulse then allows the temporal resolution of a perturbation initiated by a time-delay controlled residual IR pulse form the laser.




(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, Universty of New Mexico). (b) Schematic of the irradiance condition ($\lambda = d\sin\theta$) at wavelength $\lambda$ proposed to achieve laser-driven phase-coherent electron photoemission (green wavelets) from a nano-patterned photocathode with spatial period $d$.

## References

[1] O. Bostanjoglo et al., *Ultramicroscopy* **81** (2000) 141.
[2] V.A. Lobastov et al., *PNAS* **102** (2005) 7069.
[3] W.E. King, et al., *J. Appl. Phys*. **97** (2005) 111101.
[4] *See, for example,* J. Cao et al., *Appl. Phys. Lett*. **83** (2003) 1044.
[5] H. Niu and W. Sibbett, *Rev. Sci. Instrum*. **52** (1981) 1830.
[6] B.J. Siwick et al., *J. Appl. Phys*. **92** (2002) 1643.
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