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Prospects for Ultrafast Electron Mircoscopy: Electron Pulse Compression and Spatial Coherence

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The prospect of combining today's atomic-scale resolution of electron microscopy [1] with the sub-picosecond ($<10^{-12}$s) temporal resolution now standard in ultrafast laser spectroscopy [2] promises tremendous scientific advances by providing an important new tool for sensing materials and molecular dynamics on the nanoscale [3]. An ultrafast electron microscope (UEM) (e.g., a dynamic transmission electron microscope (DTEM)) with such unprecedented spatio-temporal resolution could allow the direct visualization of individual atomic motions, which should have a major impact on our understanding of fundamental ultrafast processes in nature; such as, single molecular bond dissociation in catalysis (biological or industrial) and atomic re-arrangements in phase transformations. An electron microscope with ultrafast observation capabilities is also expected to revolutionize the burgeoning field of nanotechnology by enabling the dynamic properties of an *individual* nanoparticle to be investigated for the first time.

Prior work, including the demonstration of the first UEM with ~100nm spatial and ~1ns temporal resolution [4], over 30 years of research into streak camera technology for ultrashort laser pulse measurement [5], and recent ground-breaking ultrafast electron diffraction studies of matter [6,7], has indicated that the development of a functional UEM faces two key challenges. First, although sub-ps electron pulses can be readily produced from a femtosecond laser-driven photocathode [7], inherent and detrimental electron-electron scattering (i.e., Coulomb or space-charge) effects will both broaden the electron pulse duration [8] and decrease its spatial coherence upon propagation. For a given average current down the electron microscope column, the linear density dependence of the electron-electron scattering rate implies that deleterious space-charge effects can be significantly reduced by simultaneously (*i*) eliminating beam-crossing (or focal) points, (*ii*) employing a large-area (~mm^2) photocathode driven by a high-power and high repetition-rate (~MHz) laser (e.g., a diode-pumped femtosecond Yb:fiber laser [9]), and (*iii*) minimizing the time-of-flight from the photocathode to the sample. On the other hand, it should also be possible to compensate for space-charge-induced intra-pulse energy chirp [8], and hence reverse the electron pulse broadening, through the use of a synchronized and suitably phased RF acceleration cavity – a standard technique in high-energy physics particle accelerators. These considerations lead to the proposed short-column (~20cm) DTEM design shown schematically in Figure 1(a), where a 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.

The second challenge concerns the degree of spatial coherence in the electron pulse required for the UEM to achieve the desired sub-nm spatial resolution. Ideally, to offset the decoherencing effect of electron-electron scattering and the lack of 'spatial filtering' down the proposed UEM column, the electron pulse initially produced by the laser-driven photocathode should be perfectly spatially coherent. This is difficult to achieve since the electronic state from which the photoemission occurs is generally not coherent over the spatial extent of a large-area photocathode. Fortunately, today's nanotechnology may offer a solution to this problem – a nanopatterned photocathode, such as a suitably designed array of quantum dot photoemitters. Nonlinear optical techniques (e.g., multi-photon-assisted field emission (Figure 1(b)) could then be used to phase-control the photoemission from such a nanopatterned photocathode to yield the initially spatially coherent electron pulse that is essential for the UEM to achieve sub-nm resolution.

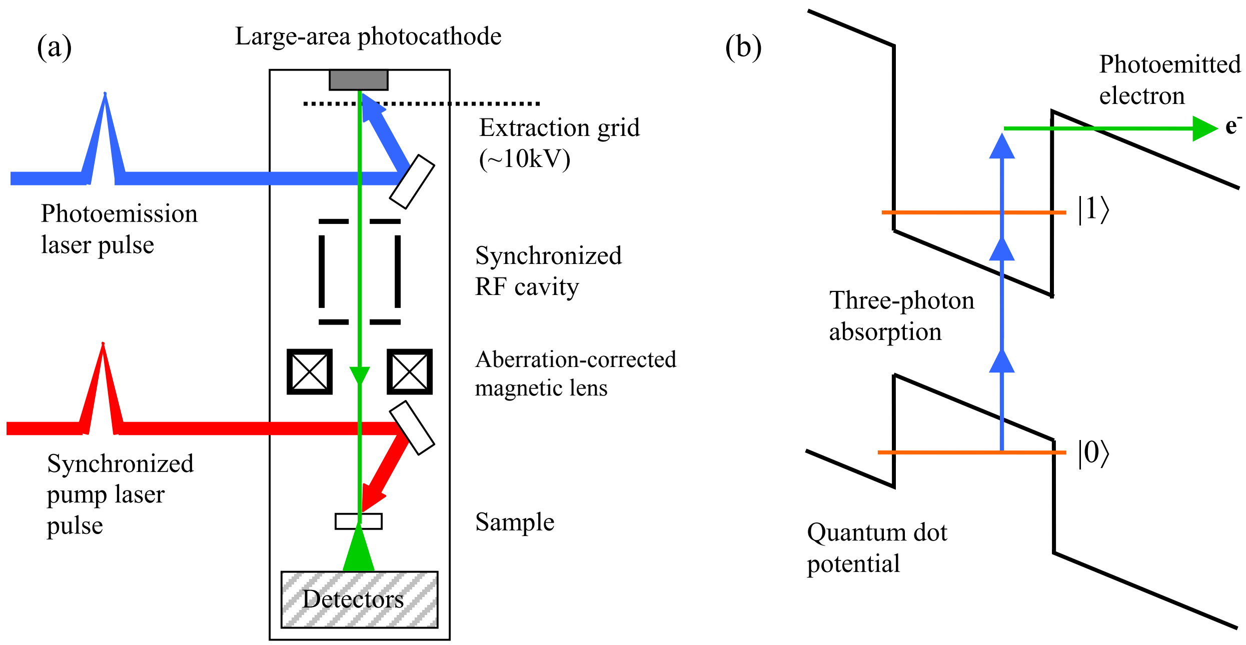

Fig. 1. (a) Schematic of the proposed short-column DTEM and (b) an example of three-photon-assisted field emission from the ground state |0⟩ of a quantum dot.

To determine the effectiveness of the proposed UEM column design, an electron pulse propagation model has been developed to investigate the limits imposed by space-charge effects. The model, which is based on a Gaussian-shell model optical beam propagation analysis, connects both the spatial and temporal (i.e., pulse broadening) beam coherence to the electron-electron scattering. Preliminary results from this four-dimensional electron pulse propagation model will be presented.

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