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ULTRAFAST TIME-RESOLVED ELECTRON DIFFRACTION WITH MEGAVOLT ELECTRON BEAMS

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An rf photocathode electron gun is used as an electron source for ultrafast time-resolved pump-probe electron diffraction. We observed single-shot diffraction patterns from a 160 nm Al foil using the 5.4 MeV electron beam from the Gun Test Facility at the Stanford Linear Accelerator. Excellent agreement with simulations suggests that single-shot diffraction experiments with a time resolution approaching 100 fs are possible.

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Our understanding about dynamical processes in chemistry, materials science and biology on the picosecond and sub-picosecond time scale stems almost exclusively from time-resolved spectroscopy. Structural changes, on atomic length scales, can only be inferred indirectly from the analysis of spectra. Both x-ray and electron diffraction share the goal of 'imaging' molecular structures with a time resolution that captures the motions as systems evolve, whether they be solids, liquids or gases. Lab scale experiments in both electron diffraction[1,2] and x-ray scattering[3] have produced impressive results. Recently, in anticipation of the construction of the Linac Coherent Light Source (LCLS) at the Stanford Linear Accelerator Center (SLAC), an experiment using the electron bunch from the SLAC Linac to produce spontaneous undulator radiation[4] has shown the possibilities for ultrafast x-ray scattering from condensed systems with 100 fs time resolution.[5] This has encouraged us to approach ultrafast electron diffraction (UED) using experimental techniques based on electron sources developed for particle accelerators, with the aim of obtaining single-shot diffraction patterns on a 100 fs time scale.

Electron diffraction is complementary to x-ray scattering, but features much larger cross sections that allow the study of surface phenomena, the bulk structures of thin foils and membranes, as well as molecular structures of gas phase samples.[6] As with linac based x-ray sources there has been significant development of electron sources for UED based on the use of photocathodes.[7] Unfortunately, the space-charge interactions of the electrons within a pulse, and the initial kinetic energy distribution with which the electrons are generated, have made it difficult to obtain pulses much shorter than 1 ps[8,9,10],in 'conventional' UED experiments using ≈30 keV electron beams. To improve the time resolution one could use fewer electrons per pulse, but that requires longer data acquisition times to obtain the necessary signal-to-noise ratio.[11] Alternatively, it is possible to increase the electric field inside the electron gun, while reducing the flight distance between the gun and the target.[12] Both tend to reduce the time of flight of the electron pulse, thereby giving the electron pulse less time to spread. Even so, this approach is limited because the maximum DC and pulsed electric fields are 12 MV/m and 25 MV/m, respectively.[13,14]

In the present work we take a fresh approach to ultrafast time-resolved pump-probe diffraction by using MeV electron beams generated in a gigahertz RF photocathode electron gun.[15,16] The concept stems from the development of RF electron sources for free electron lasers. The physics of these sources is well understood and combines the following advantages:

1) Electron energies of 5-6 MeV are easily accessible. Intense, well-collimated electron beams can be generated.[2]

2) The electric fields in the gun can exceed 100 MV/m,[17] so that the electrons reach relativistic speeds within a few millimeters. Once relativistic, the Coulomb repulsion stops playing an important role in longitudinal pulse broadening, because the impulse required to change the speed of a relativistic electron is much higher than that for a non-relativistic electron. In addition, relativistic beams have little velocity spread, implying minimal pulse broadening between the gun and the target.

3) With proper timing of the laser pulse with respect to the field in the RF gun, the electron bunch can be significantly shorter than the laser pulse used to generate the electrons.[18]

Our experiments use the SLAC gun test facility (GTF),[19] shown schematically in figure 1, which was designed to deliver a high brightness electron beam for the LCLS project. The GTF gun routinely produces low divergence, short duration electron bunches. The GTF beam line configuration consists of a 1.6 cell photocathode rf gun and magnetic solenoid, followed by a 3 m linac, both operating at the s-band radiofrequency of 2.856 GHz. For this experiment, the linac is not powered and serves as an electron drift tube. The gun rapidly accelerates photoelectrons in an rf field with peak amplitude on axis of 104 MV/m to an energy of 5.4 MeV, preserving the short pulse lengths and transverse beam quality. The copper photocathode of the RF gun is illuminated by a quadrupled Nd:glass laser, at 263 nm, with a pulse duration of 2 ps FWHM, and a flat-top spatial profile with 2 mm diameter. The 160 nm thick Al sample is placed 0.75 meters downstream of the cathode, and a view screen is 3.95 m downstream of the sample. A solenoid at the gun exit is used to minimize the divergence at the Al sample, and a magnetic quadrupole lens doublet produces a parallel-to-point imaging between the sample and the detector.

The transverse[20] and longitudinal[21] beam quality produced by the GTF gun have been previously reported. Measurements are typically performed with the linac on, accelerating the beam to approximately 30 MeV. The beam parameters determined from these measurements can be translated upstream to the position of the Al sample, where the beam energy is only 5.4 MeV. Although measurements were not made with the exact accelerator settings used in the diffraction experiment, the estimated beam parameters are listed in Table I based on previously reported data with similar accelerator settings.

The estimated rms bunch length is 560 fs and the total energy spread is 36 keV or 0.65%. The pulse length is much shorter than the roughly 800 fs rms (2 ps FWHM) laser pulse that generates the electron beam. The electron bunch length can be controlled by adjusting the laser arrival time at the cathode with respect to the applied rf field. Additional compression can be achieved by optimizing the laser pulse arrival time at the cathode.

The electrons were detected with a thin, phosphor coated Al substrate. The screen is mounted at 45 degrees with respect to the incident electron beam and photons are emitted at 90 degrees through a standard quartz viewport. The video system consisted of a Pulnix TM7EX analog camera with a C mount lens adjusted to image the screen onto the CCD. The working distance was 182 mm and the numerical aperture was approximately 0.15. Images were captured with an 8 bit frame grabber. The DQE of the detector used in the present study is estimated to be between 0.01 and 0.1.

Simulations of the complete GTF system were carried out in order to model the diffraction experiments. We used the General Particle Tracer program[21] (GPT) to calculate the trajectories of a swarm of 832,195 ‘particles’ with a total charge adjusted to match that of the electron pulse.[22] Figure 2 shows both the experimentally measured and the simulated diffraction patterns from the polycrystalline foil. The simulations include the full six dimensional phase space of the electron beam from GPT. The inset shows the observed image together with a false color map of the simulation results. It is clear that even the single, ultrashort electron pulse is sufficient to record the powder pattern of the foil. However, time resolved images will require thinner foils to match the typical laser penetration depth (20 nm for Al as used by Siwick and collaborators[12] for example) and the number of electrons scattered will scale with the thickness. The reduction in signal strength should be compensated by a detector with a DQE approaching one.

In general, there is a good agreement between the simulations and the experiment. All peak positions and the overall intensity profile are well reproduced. The experimentally observed width of the diffraction peaks is broader than calculated. This is due to differences in the actual versus simulated electron beam phase space. These differences arise from differences between the simulation input and the actual parameters for the experiment ranging from non-uniformity of the laser illumination of the photocathode, errors in rf phase, magnetic field errors to name a few. The simulations clearly indicate that with better control of these parameters the (111) and (200) peaks can be easily resolved. These improvements are beyond the scope of this proof of principle experiment. Also, the differences between the measurement and simulation can be used to extract the actual electron beam phase space as pointed out by Wang, Xiang, Kim Ihee[16].

In summary, we have shown that diffraction patterns are obtainable from a single, ultrashort pulse with MeV energy. Use of relativistic electrons greatly reduces the space-charge limitations that constrain diffraction with smaller beam energies. It is possible to maintain a 500 femtosecond electron pulse with a charge that is sufficient to obtain a single shot diffraction pattern. Finally, pulse compression within the RF gun counters temporal broadening effects, and simulations[23] suggest that a time resolution exceeding 100 fs is possible. Atomic scale 'movies' of molecular structures, phase transitions in solid films and liquid dynamics are thus within reach.

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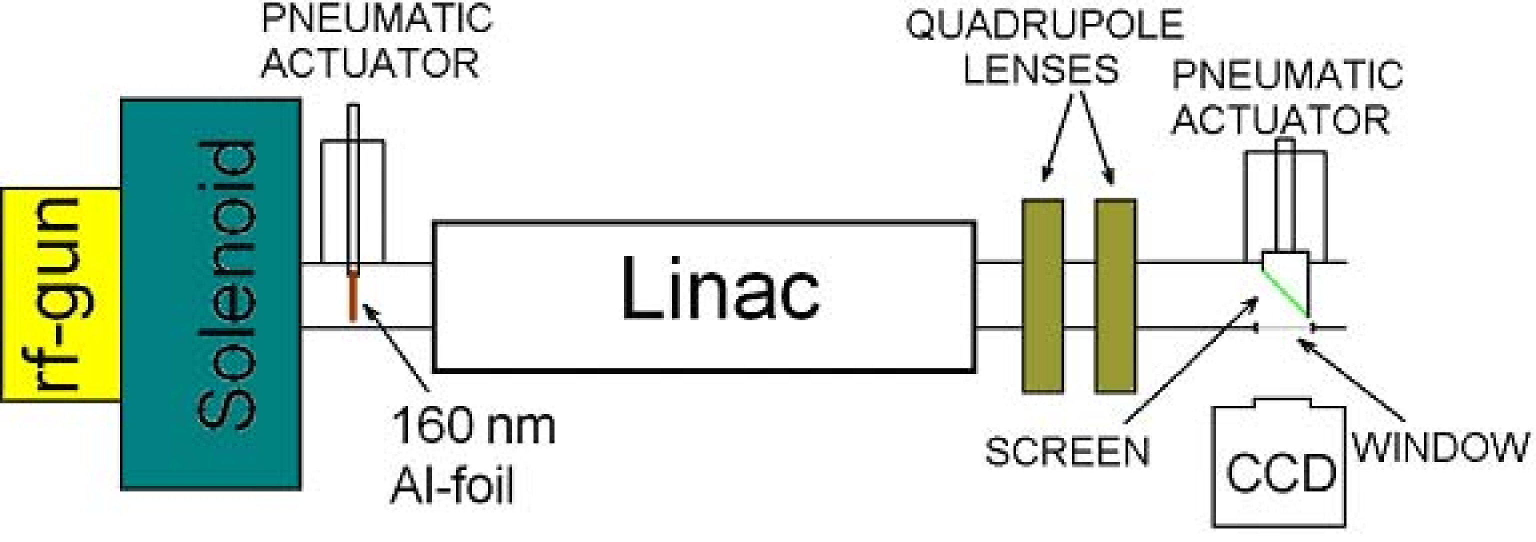
Table I: The estimated electron beam parameters from an S-band photo-cathode rf gun used in the experiment.

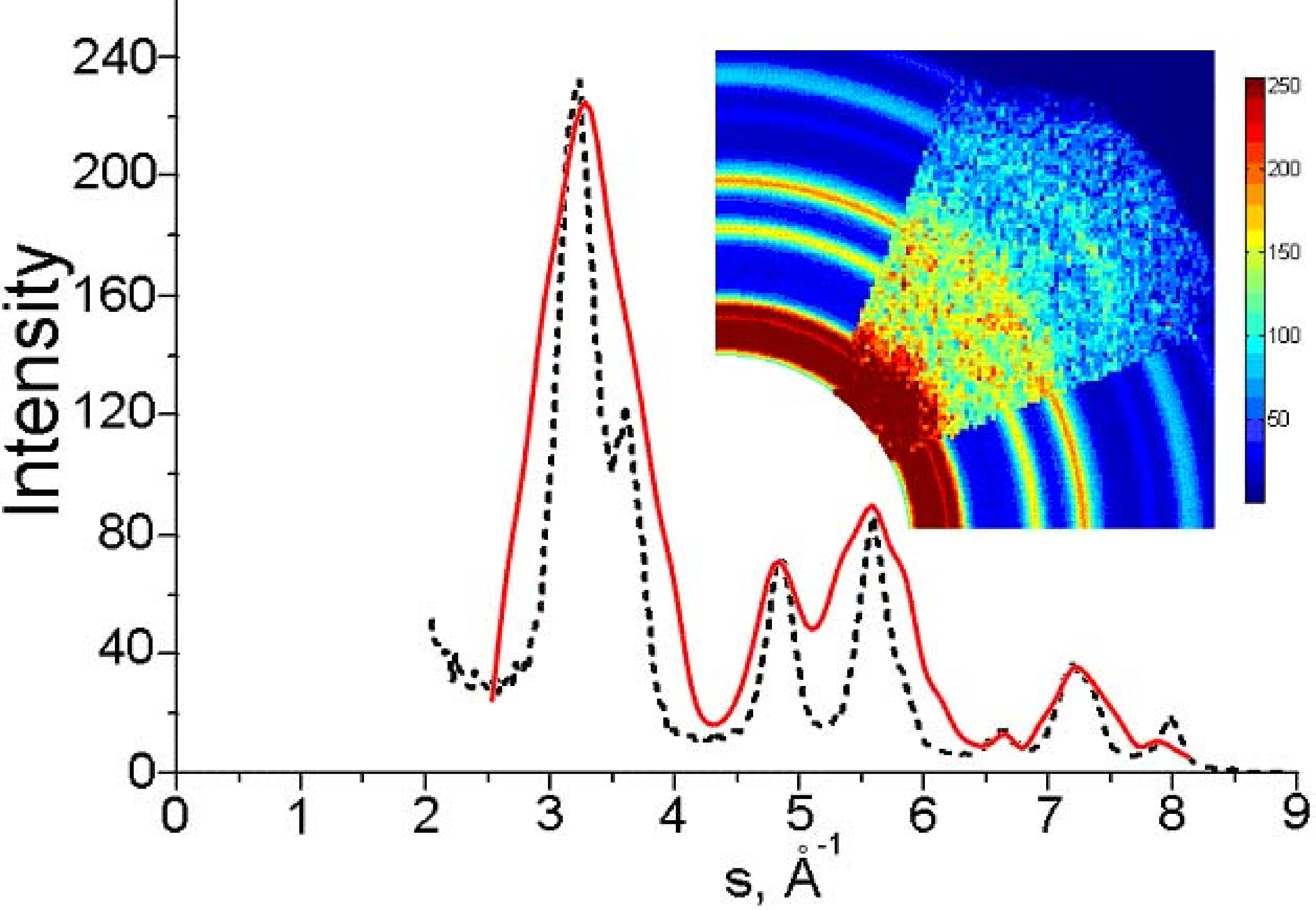
Parameter	**Value**	**Units**
Charge	2.9	pC
Energy	5.4	MeV
rms Energy Spread	6	keV
rms Pulse Length	560	fs
Longitudinal Emittance	2.5	keVps
Normalized Emittance	0.85	μm
Geometric Emittance	0.075	μm
Minimum rms divergence	45	μrad
Solenoid Field	1.7	kG
Gun Gradient	104	MV/m
Laser Gun Phase	40	degrees

Figure captions

Figure 1. Layout of the GTF beam line and the electron diffraction experiment.

Figure 2: Diffraction pattern of 160 nm aluminum foil observed with single electron pulse of ≈ 500 fs rms duration and beam energy of 5.4 MeV, solid line, and simulated pattern, dashed line. Inset: Pie slices of the observed and simulated patterns.


PNEUMATIC
ACTUATOR
QUADRUPOLE
LENSES
PNEUMATIC
ACTUATOR
rf-gun
Solenoid
Linac
160 nm
Al-foil
SCREEN
CCD
WINDOW



Intensity
240
200
160
120
80
40
0
0 1 2 3 4 5 6 7 8 9
s, Å^{-1}
250
200
150
100
50