Ten-femtosecond (rms) single-electron diffraction

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Abstract: We report single-electron pulses of 12-fs/28-fs duration (rms/FWHM), characterized by laser streaking. A time-resolved diffraction study on fibrous graphite polycrystals reveals the practical feasibility of single-electron diffraction at potentially few-femtosecond resolution.
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1. Introduction

Ultrafast electron microscopy and diffraction [1-2] can provide a four-dimensional visualization of atomic motion in space and time. Currently, the temporal resolution is limited by space charge effects and dispersion to about 100 femtoseconds. This is often not enough to observe primary processes in molecules or condensed matter, i.e. the half periods of vibrational modes or optical phonons. Single-electron pulses are intrinsically immune to Coulomb repulsion and can in principle have few-fs duration and below [3-4]. Realization of that potential, however, requires a careful management of electron dispersion, similar to the handling of chirp in femtosecond optics. Furthermore, single-electron diffraction comes at a price: The high pump-probe repetition rate required for obtaining data in reasonable time restricts the investigations to mostly reversible processes. Also, thermal load on the sample must be handled. Here we report on our progress in overcoming these hurdles, reporting (i) a 15-fs (rms) laser-electron cross correlation (see Fig. 1) and (ii) a first time-resolved diffraction study with entirely single-electron methodology.

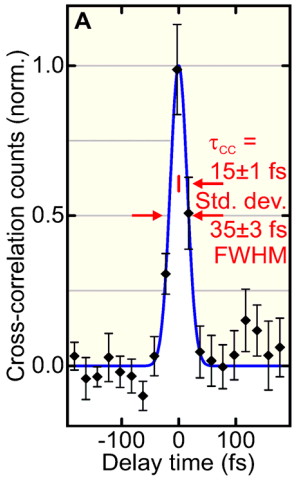
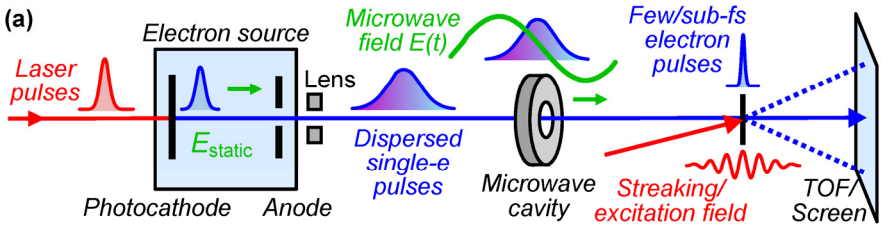


Fig. 1: Shortest electron pulses at 25 keV for atomic-scale diffraction.

2. Single-electron pulse compression

Our experimental approach is sketched in Figure 2(a). A femtosecond laser produces single-electron pulses, which are accelerated to 30-100 keV by a static electric field. The beam is collimated by a magnetic lens with ideally 'isochronic' properties [5]. The initial energy bandwidth translates to dispersion in time during propagation. A microwave cavity with optically enhanced jitter suppression [6] is applied to reshape the single-electron phase space from the temporal into the energetic domain, hence shortening the pulses at the cost of an increase in energy bandwidth [7]. The electron pulses can become shorter than the laser pulses used initially for photoemission. Pulse characterization is achieved by using an optical field for streaking the electron pulses in the energy domain [8], similar to attosecond XUV streaking. Figure 1 shows the shortest electron pulses achieved so far in our laboratory, having a cross-correlation width of 15 fs (rms) corresponding to a pulse duration of 12±2 fs (rms) or 28±5 fs (FWHM). These are the shortest diffraction-capable electron pulses that we know of. Figure 2(b) shows a diffraction pattern from the ground state of a molecular proton transfer switch compound, N-(triphenylmethyl)-salicylidenimine, demonstrating the ability to obtain atomic-scale resolution with compressed single-electron pulses [9]. Also, electron-energy-loss spectra (EELS) can be obtained.



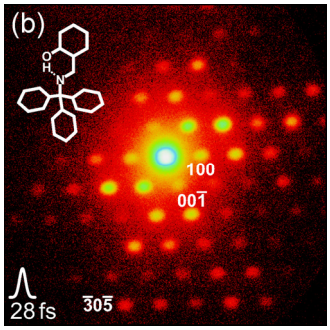


Fig. 2: Single-electron diffraction at ultimate temporal resolution. (a) Experimental concept. (b) Diffraction with compressed single-electron pulses from a molecular switch compound.

3. Time-resolved single-electron diffraction

Roughly 10^7 incoming electrons are required for a suitable diffraction image and about 10-100 different pump-probe delays must be scanned for a complete, dynamical picture. In the single-electron regime, an acceptable overall measurement time can only be achieved at rather high pump-probe repetition rates, potentially causing problems with average power and reversibility.

Here we report on our first success with using single-electron pulses, intrinsically space-charge-free over the entire trajectory right from the source, for recording time-resolved diffraction of a realistic sample, here a fibrous polycrystalline thin-film made from highly-ordered pyrolytic graphite (HOPG). An extremely fine TEM mesh is used as a support structure for heat removal. The graphite layers are tilted by about 20° in order to split the diffraction rings into arcs with in-plane and out-of-plane contributions. Thanks to the good spatial coherence of single-electron pulses [9], Bragg spots are discernible, corresponding to selected-area diffraction from individual grains.

Figure 3 shows the time-resolved intensity of 27 selected spots, partially obtained with ten-electron pulses at 128 kHz or alternatively with genuine single-electron pulses at 256 kHz, at the natural few-hundred femtosecond resolution of single-electrons without the microwave compressor [8].

Laser-induced excited carriers first relax into strongly coupled optical phonons (SCOP) and subsequently thermalize with all phonons, causing significant thermal expansion along the *c*-axis. Shifts of the grain-specific rocking curves cause Bragg spot intensities to increase or decrease. The data hence reveals the time-resolved *c*-axis dynamics of each grain individually. The blue and red lines are fits to an oscillator model with a driving term related to a two-temperature model. The results reveal a rather homogeneous oscillation period and film thickness (13-18 nm), but also a surprisingly wide distribution of electron-phonon thermalization times (2-12 ps), indicating the major influence of inhomogeneity for the structural dynamics of fibrous polycrystalline graphite [10].

4. Conclusion

These results show that time-resolved single-electron diffraction meets the requirements of realistic samples. Combined with microwave compression, it is a promising approach for visualizing structural dynamics in the few-fs regime or eventually even below.

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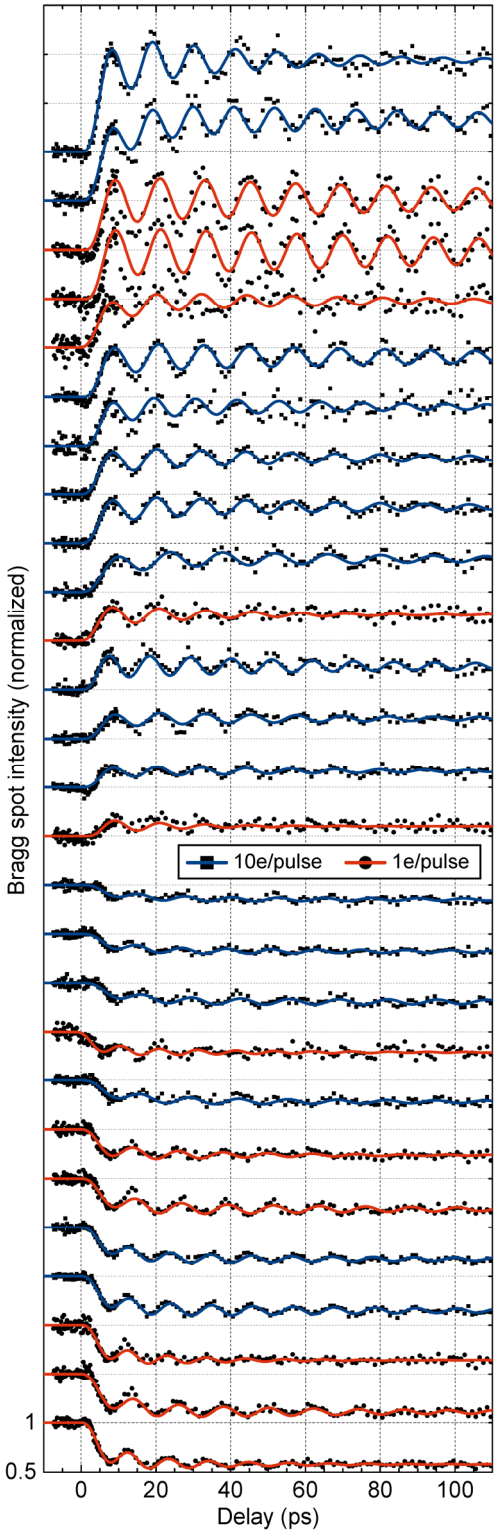

Fig. 3: Time-resolved single-electron diffraction on fibrous graphite polycrystals reveals coherent acoustic phonons of individual grains.