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Research Article

Relativistic Ultrafast Electron Microscopy: Single-Shot Diffraction Imaging with Femtosecond Electron Pulses

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We report on a single-shot diffraction imaging methodology using relativistic femtosecond electron pulses generated by a radio-frequency acceleration-based photoemission gun. The electron pulses exhibit excellent characteristics, including a root-mean-square (rms) illumination convergence of $31 \pm 2\ \mu$rad, a spatial coherence length of 5.6 ± 0.4 nm, and a pulse duration of approximately 100 fs with $(6.3 \pm 0.6) \times 10^6$ electrons per pulse at 3.1 MeV energy. These pulses facilitate high-quality diffraction images of gold single crystals with a single shot. The rms spot width of the diffracted beams was obtained as 0.018 ± 0.001 Å^{-1}, indicating excellent spatial resolution.

1. Introduction

Recently, single-shot diffraction imaging with ultrashort X-ray pulses generated from free-electron lasers has facilitated the study of structural dynamics of irreversible processes in material samples [2] and the acquisition of direct structural information in chemistry and biology before sample damage [3]. However, ultrafast electron diffraction and microscopy (UED and UEM) with electron pulses are also very promising techniques for the study of ultrafast structural dynamics in materials because electrons are complementary to X-rays in a number of ways [4]:

(1) Electrons have a larger elastic scattering cross section and can easily be focused. Measurement using electrons is used to observe structural information of small or thin crystals, light-element materials, and gas phase samples [5].

(2) Electron imaging technology with high spatial resolution is well developed.

(3) Femtosecond electron pulses are achievable using photoemission guns. The instrument is compact.

The most widely used UED [6–8] and UEM [9–18] instruments employ a static dc acceleration-based photoemission gun for generating short electron pulses with energies ≤ 200 keV. The main obstacle to using the dc guns is the significant space charge effect [7, 8]. The space charge force of electrons in the nonrelativistic energy region not only broadens the pulse width but also acts to increase energy spread and beam divergence. This leads to a loss in spatial resolution [19]. Current state-of-the-art dc guns generate ~ 300 fs electron pulses that contain several thousand electrons per pulse at ~100 keV energies and have a beam convergence in the milliradian range [20, 21]. However, because of the relatively low number of electrons per pulse, such dc gun-based UED and UEM instruments are difficult to operate in single-shot mode. To improve temporal and spatial resolution, a stroboscopic methodology using single-electron pulses in the UEM system has been proposed. However, this approach limits the potential applications to reversible processes [11, 18].

To overcome the space charge problem, we have developed a prototype relativistic UEM with a radio-frequency (rf) acceleration-based photoemission electron gun [1]. The rf gun is an advanced electron source for generating high-brightness relativistic-energy electron beams in a particle accelerator field [22–24] and has been applied widely in free-electron lasers [25]. The relativistic UEM using the rf gun

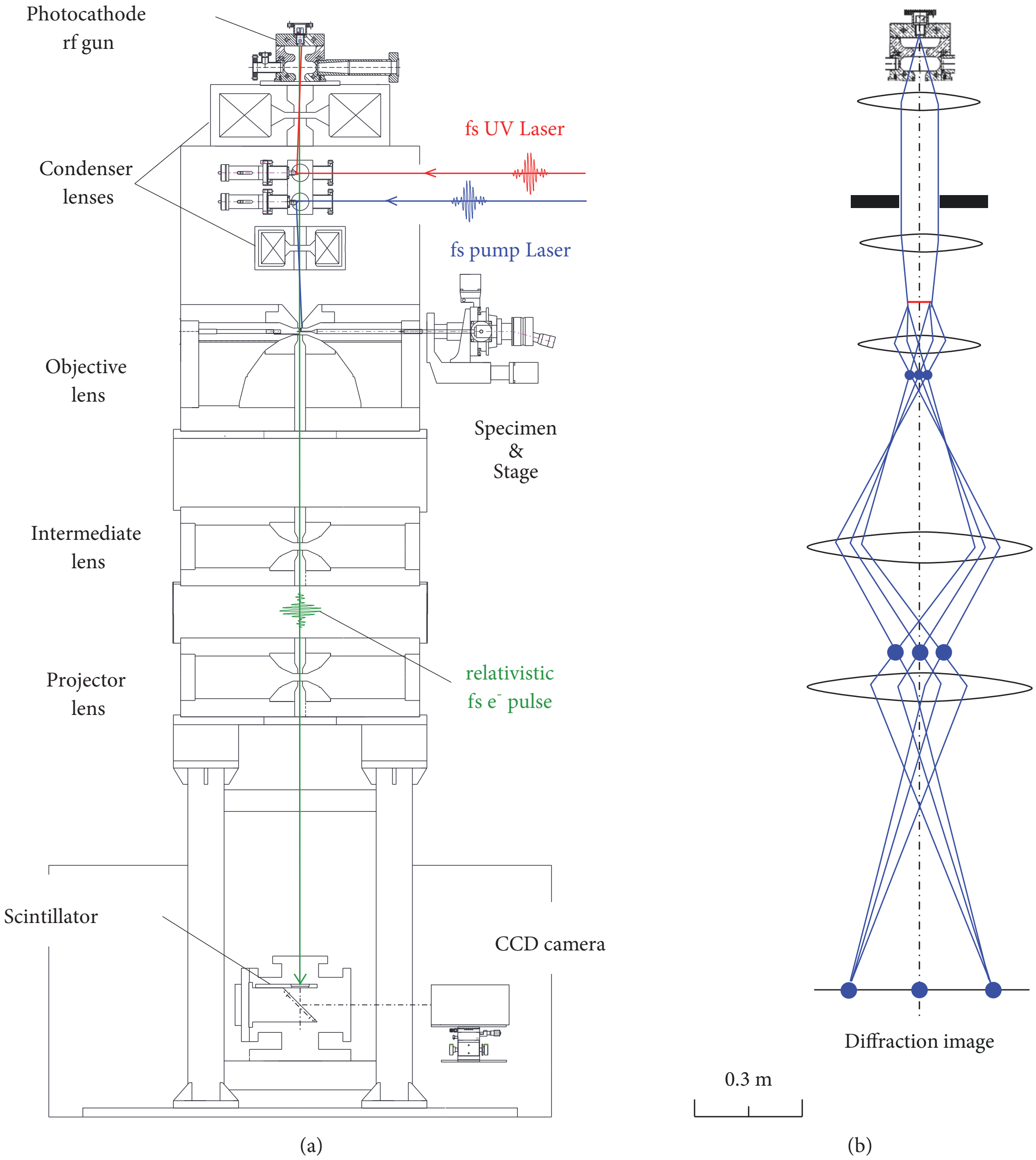
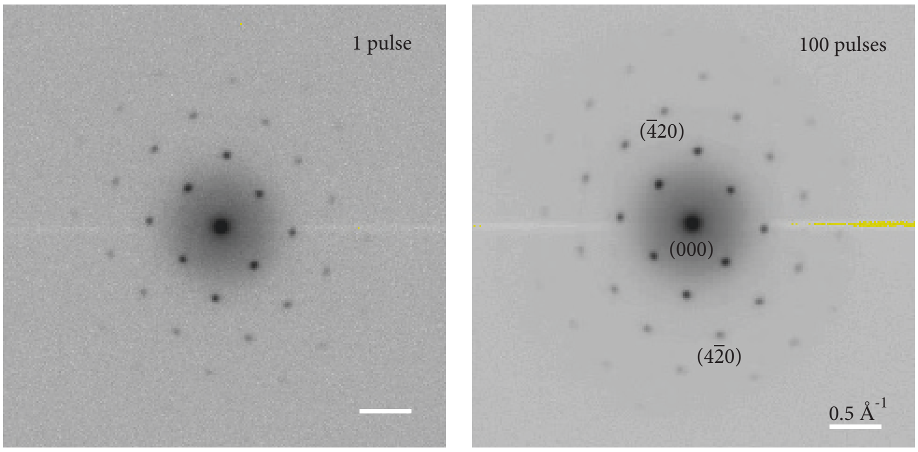


Figure 1: Prototype relativistic UEM with MeV femtosecond electron pulses generated by a photoemission rf gun: (a) cross-sectional schematic of prototypical UEM [1] and (b) ray diagram for electron diffraction imaging.

has three crucial advantages over nonrelativistic UED and UEM systems. Firstly, it can perform single-shot diffraction imaging with femtosecond temporal resolution. Relativistic femtosecond electron pulses containing 10^6 electrons per pulse have recently been generated using rf guns with femtosecond laser pulses [26], and they have been employed in UED experiments [27–35]. Secondly, relativistic-energy electron beams greatly enhance the extinction distance for elastic scattering and provide structural information that is essentially free from multiple scattering and inelastic effects [36, 37]. Our previous UED study of the structural dynamics of laser-irradiated gold nanofilms indicate that the kinematic theory can be applied in the case of 3 MeV probe electrons with the assumption of single scattering events [38, 39]. This allows us to easily understand and explain structural dynamics. Thirdly, a thick sample can be used for measurement, thereby obviating the requirement to prepare suitable thin samples. In this letter, we report on a single-shot diffraction imaging methodology using our relativistic UEM with femtosecond electron pulses.

2. Single-Shot Diffraction Imaging Methodology Using Relativistic Femtosecond Electron Pulses

Figure 1 shows the schematic of a prototype relativistic UEM constructed with a photocathode rf gun, an electron illumination system, an objective lens, an intermediate lens, a projector lens, and an image measurement system. The design



(a)

(b)

Figure 2: Relativistic diffraction images of 10 nm thick (100)-orientated single-crystalline gold film measured with (a) single-pulse (single-shot) and (b) 100-pulse integration. The energy of the femtosecond electron pulses was 3.1 MeV, and the number of electrons in each pulse was $(6.3 \pm 0.6) \times 10^6$.

and characteristics of each component have been reported in [1]. The photocathode rf gun was driven by a Ti:sapphire femtosecond laser to generate femtosecond electron pulses. The electron energy was 3.1 MeV. The repetition rate of the electron pulses, which was limited by our klystron modulator, was 10 Hz. The electron pulses were paralleled by a condenser lens in the electron illumination system, collimated with a 1.0 mm diameter condenser pinhole, and then injected onto the specimen.

The objective, intermediate, and projector lenses are utilized for diffraction imaging. The pole pieces in the objective lens were made of a soft magnetic alloy (Permendur) [1] and generated a magnetic field strength of 2.3 T at the center of the pole pieces. The focal length of the objective lens was 5.8 mm for a 3 MeV electron beam. For diffraction measurements, we precisely adjusted the intermediate lens, so that the back-focal plane of the objective lens acted as the object plane of the intermediate lens. The diffraction pattern (DP) was then projected onto a viewing screen (scintillator) using the projection lens. To achieve high sensitivity to MeV electron detection with a high damage threshold, we chose a Tl-doped CsI columnar crystal scintillator equipped with a fiber optic plate (Hamamatsu Photonics) to convert the relativistic-energy DPs into optical images [1]. The optical images were detected with an electron-multiplying charge-coupled device (CCD) of 512 × 512 pixels.

3. Experimental Results

In the demonstration for electron diffraction imaging, we used a single-crystalline gold film with a thickness of 10 nm, which was placed on a gold mesh (Cat. No. P066, TAAB Laboratories Equipment Ltd., Reading, UK) as the specimen. We removed the objective aperture and readjusted the position of the specimen along the optical axis to optimize image contrast. Figure 2 shows the DPs of a (100)-orientated single-crystalline gold sample observed both via a single-pulse (single-shot) and via 100-pulse integration. The energy of the electron pulses was 3.1 MeV, and the number of electrons per pulse was $(6.3 \pm 0.6) \times 10^6$. The fluctuation in the number of electrons per pulse was mainly caused by the instability of the incident UV laser pulse energy. The pulse duration was not measured in the experiments; however, we estimated it to be 99 ± 5 fs rms by the theoretical simulation with the aid of General Particle Tracer (GPT) code [40] using the incident UV laser pulse at the rf gun launch phase of 30°, and the electron number per pulse of $(6.3 \pm 0.6) \times 10^6$. The error of the pulse duration is due to the space charge effect in the region of the measured fluctuation of the electron number, and the change in the launch phase of $30^\circ \pm 10^\circ$ in the rf gun. Figure 3 represents the intensity profiles along the (-420) and (4-20) spots in the images acquired by single-shot and 100-pulse integration. As illustrated in Figures 2 and 3, sharp DPs and good contrast were observed. Higher-order spots of (-420) and (4-20) from the gold single crystals with scattering vectors up to 1.1 Å^{-1} were captured clearly with a single shot. The rms width of the zeroth-order spot (000) in the single shot was measured as 0.018 ± 0.001 Å^{-1}, indicating an excellent spatial resolution for the MeV diffracted beam.

Based on the width of the (000) spot and the measured distance of the diffraction spots from the (000) position, we estimated the rms illumination convergence angle (α) of the electron beam at the specimen to be $\alpha = 31 \pm 2$ μrad. This convergence angle is two orders smaller than that of nonrelativistic UEDs. Additionally, the coherence of the electron source is an important parameter in diffraction imaging, especially in terms of the spatial coherence (transverse coherence), which determines the sharpness of the DPs and the diffraction contrast in the acquired images. The spatial coherence length is defined as [41]

$$d_c = \frac{\lambda}{2\alpha}, \tag{1}$$

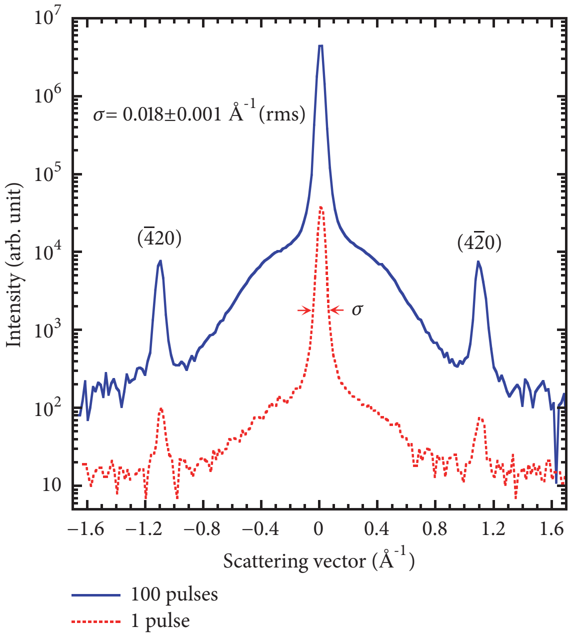


Figure 3: Intensity profiles along the (-420) and (4-20) spots of the images acquired by single-shot (broken curve) and 100-pulse integration (solid curve). The rms width of the (000) spot was obtained as 0.018 ± 0.001 Å^{-1} from the intensity profile of the single-shot image with a Gaussian fit.

where λ is electron wavelength and α is the rms illumination convergence angle. From the obtained illumination convergence angle, we evaluated the spatial coherence length of the electron pulses generated with the rf gun to be d_c = 5.6 ± 0.4 nm, which is twice as large as that of current UED systems [8, 18, 42]. This allows us to detect sharp DPs and acquire good contrast diffraction images with a single-shot and integration measurements, as shown in Figure 2.

4. Summary

In summary, we have proposed a single-shot diffraction imaging methodology with a relativistic UEM based on an rf gun. The rf gun generated femtosecond electron pulses with pulse durations of approximately 100 fs that contained $(6.3 \pm 0.6) \times 10^6$ electrons per pulse at an energy of 3.1 MeV. The number of electrons per pulse was two or three orders higher than that of nonrelativistic UEDs. In our experiments, the electron pulses exhibited excellent characteristics, including an rms illumination convergence angle of the electron beam at the specimen of $\alpha = 31 \pm 2$ μrad, and a spatial coherence length of $d_c = 5.6 \pm 0.4$ nm. The convergence angle was two orders smaller than of nonrelativistic UEDs, while the spatial coherence length is twice as large as that of current UED systems. Using these pulses, we obtained a high-quality diffraction image from single-crystal gold with a single shot. The measurements were successful in facilitating the detection of higher-order DPs with a scattering vector up to 1.1 Å^{-1} and a spatial resolution of 0.018 ± 0.001 Å^{-1}. Single-shot diffraction imaging methodology with relativistic femtosecond electron pulses is promising for studying ultrafast phenomena in materials, i.e., phase transformations of crystalline materials, chemical reactions, and structural dynamics of biomolecules at the femtosecond time scale.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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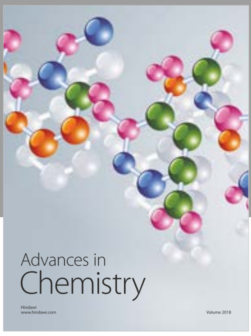
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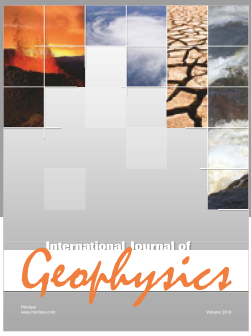
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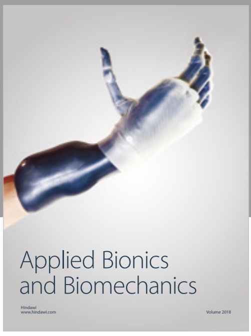
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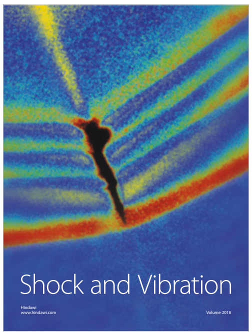
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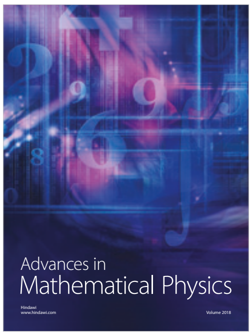
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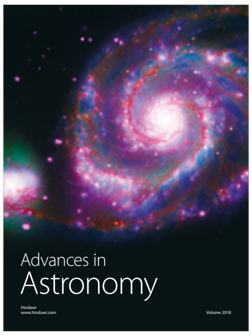
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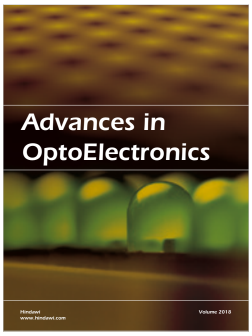
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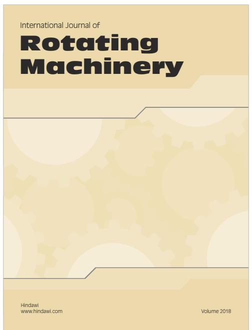
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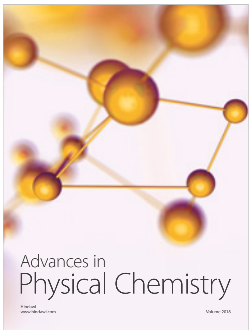
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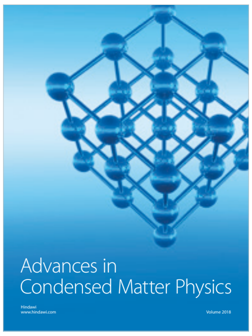
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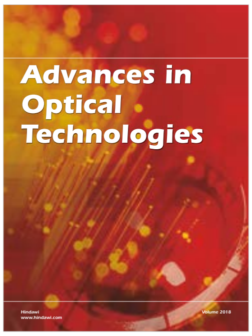
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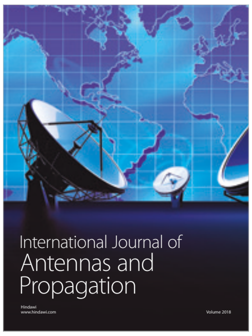
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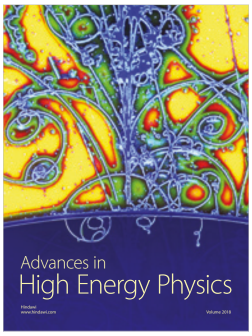
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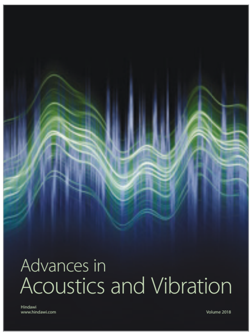
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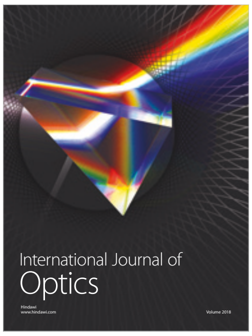
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