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Reducing Transient Electric Fields Effect in Ultrafast Electron Diffraction Using Multiple Laser Pulse Train

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The technique of Ultrafast Electron Diffraction (UED) has attracted significant interest recently due to its promise for time-resolved and ultrafast laser-induced structural dynamics study. In a UED experiment, a laser pulse is split into two, one for pump and the other for probe. The pump laser pulse hits the sample directly, and the probe laser pulse is used to generate picoseconds (ps), or sub-ps, electron pulses for transmission or reflection high energy electron diffraction (RHEED). In RHEED, the electron pulse strikes the sample at a glancing angle. By controlling the optical path between pump and probe pulses, diffraction patterns at different time delay can be recorded. After the laser pulse excites the sample, a change in diffraction pattern is expected coming from laser-induced structural change. However, the laser pulse also emits hot electrons through thermionic emission or multiphoton photoemission. The hot electron cloud, interacting with the probing electron beam, also induces the diffraction pattern change. This is the so-called transient electron field (TEF) effect[1-4]. How to remove or reduce TEF becomes a major challenge in UED experiment. Here, we report a method to attenuate the TEF by splitting single pump pulse into multiple pulse train. UED experiment, as shown in Fig. 1, is performed on epitaxial silver nanocrystals on TiO_2 substrate, and a simulation of TEF based on model in [1] is applied to compare the electron emission behavior upon the excitation of different pulse train.

In order to reduce the TEF effect, we split the single laser pulse (Ti:sapphire, 130 fs, 800nm wavelength) into multiple pulses using beam splitters (50% transmission, 50% reflection). The pulses are collimated afterwards. By adjusting the optical path, a 2 ps interval between 2 pulses can be precisely controlled. We can obtain up to 8 pulses in each pulse train using this method. By dividing single pulse into multiple pulses, the laser pulse with high peak energy is spread to multiple laser pulses with low peak energy in a longer duration. Thus, the total energy transferred to the sample is almost the same. Fig. 2 shows Ag(222) diffraction beam deflection under 2 pulses, 4 pulses, and 8 pulses irradiation as a function of delay time. The laser fluence used for pump is 2.9 mJ/cm^2. A clear reduction in TEF-induced deflection can be observed as the pulse number in the pulse train increases. The reduction can be attributed to a reduction in the electron temperature after the laser pulse excitation, and the resulted electron emission by pulse splitting. This explanation is confirmed by the two-temperature model [5], which is used to simulate the temperature change in Ag after laser excitation. In the simulation the lattice temperature all rises up to 15K in 2, 4, and 8 pulses cases. However, the electron peak temperature decreases as the pulse number increases. The results demonstrate the effectiveness of reducing TEF in UED experiment through splitting single pulse into multiple pulse train.

If there is no TEF effect in the UED experiment, the diffraction pattern change would only originate from the sample, e.g. thermal expansion. The lattice temperature rise is expected to be 15K approximately, so the percentage change in diffraction angle due to thermal expansion is on the order of

10^{-4}, which is much smaller than the experiment result ($\sim 10^{-2}$).

Using a moving electron cloud model [1], we are able to fit the measured electron beam deflection. The average initial speed of electrons v_0, the speed of electron width distribution v_{wz}, the percentage of electrons that falls back to the surface Θ_1/Θ, and the charge density of ionized surface σ_0 listed in Table 1 were obtained through fitting. From Table 1, it can be deduced that the average initial speed and the surface charge density decreases as the pulse number increase. Thus, the use of pulse train provide an effective method for reducing the TEF effects in UED [6].

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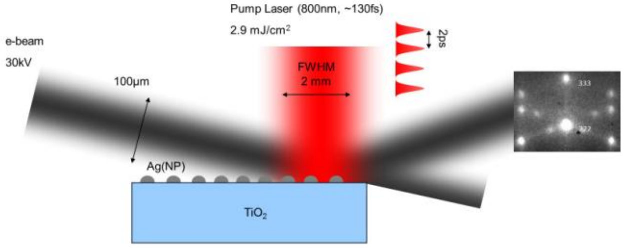

Figure 1. A schematic diagram of ultrafast electron diffraction setup. The electron pulse hits the sample at a glancing angle and the pump pulse is along the normal direction of sample surface. The pump laser pulse is divided into different multiple pulse train. The diffraction pattern is recorded by the CCD camera.

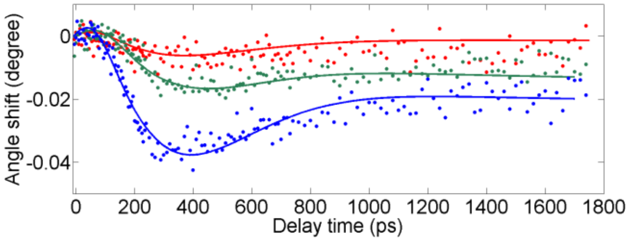

Figure 2. Comparison of electron beam position change of Ag(222) along surface normal direction and fitting result (black curve) upon 2-pulse (red curve), 4-pulse (green curve), and 8-pulse (blue curve) train pump. The peak position was obtained from fitting the intensity profile by Lorentz function.

Table 1. Fitting Result for TEF

Pulse Number	v_0 (μm/ps)	v_{wz} (μm/ps)	Θ_1/Θ (%)	$\sigma_0 \times 10^{-6}$ (e/mm^2)
2	0.24	0.5	96.8	0.16
4	0.20	0.4	96.6	0.11
8	0.15	0.4	96.6	0.019