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A Multislice Approach to Quantify Laser-Induced Lattice Temperature from Ultrafast Electron Diffraction Measurements of Single-Crystal Films

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Ultrafast electron diffraction (UED) is a powerful technique for recording atomic structure dynamics during pulsed laser or other excitations. Accurate quantification of the time-dependent lattice temperature is important for interpreting many kinds of UED experiments, such as understanding light-induced structural transformations or measuring electron-lattice coupling dynamics. However, single crystal films have posed some challenges to this, as channeling and multiple scattering effects complicate the observed diffraction signals. In many cases, this invalidates application of the Debye-Waller model and requires a full dynamical scattering treatment to recover accurate lattice temperatures. Yet, few such treatments have been demonstrated for UED, and none prior to this work included both temperature and sample topography which can dramatically alter the diffraction peak intensities.

Here, we introduce a multislice procedure for modeling UED patterns of rippled single-crystal films which enables accurate retrieval of photoinduced lattice temperature. We approximate the sample to have a Gaussian distribution of tilt angles relative to the probe. For each orientation, we compute the diffraction using the well-established fast Fourier transform (FFT) multislice approach[1]. Lattice temperature is incorporated via Debye-Waller damping of the projected atomic potentials[2]. The weighted incoherent sum of the patterns over the tilt distribution yields the total pattern from the film.

We demonstrated this approach for UED measurements of a quoted 11 nm single-crystal gold film (Ted Pella) performed at the High Repetition-rate Electron Scattering (HiRES) beamline at Lawrence Berkeley National Laboratory[3], using a beam energy of about 750 keV. We first find that the multislice approach dramatically improves matching of individual UED patterns in comparison to a kinematical model, as shown in Figure 1. Remarkably, without increasing the number of fit parameters, the multislice model reduces the error by a factor of 10, giving an R factor of 2% for the best fit. Furthermore, the retrieved film thickness of 13.5 nm is in good agreement with the quoted film thickness, and the rms tilt spread is consistent with the visible rippling of the freestanding film.

We then fit intensity changes in response to 1.2 eV photoexcitation for a series of pump laser fluences with both kinematical and multislice models to extract the lattice temperature rise, summarized in Figure 2. We observe clear, systematic deviations of the photoinduced signals from the Debye-Waller model, with 200, 400, and 600 peaks showing little change compared to 220, 420, and 620 peaks. The multislice model provides a better fit to the observed changes, improving the accuracy by a factor of 3. Critically, the multislice model retrieves a lattice temperature rise (12.3 K / (mJ cm^{-2})) that is more than 3 times larger than those obtained using the kinematical model (4.0 K / (mJ cm^{-2})). This better matches the temperature rise expected based on the known optical constants of gold (11.6 K / (mJ cm^{-2})).

Altogether, these results provide an example where dynamical scattering models are needed for accurate quantitative analysis of UED of single crystal films, even while using a relativistic electron beam. They also demonstrate an accurate multislice approach that can be applied to a wide range of materials, and can be further extended to incorporate more complex dynamics such as site-dependent atomic displacements and structural transformations. In the long term, this approach provides a step towards quantitative retrieval of the full time-dependent crystal structure from UED [4].

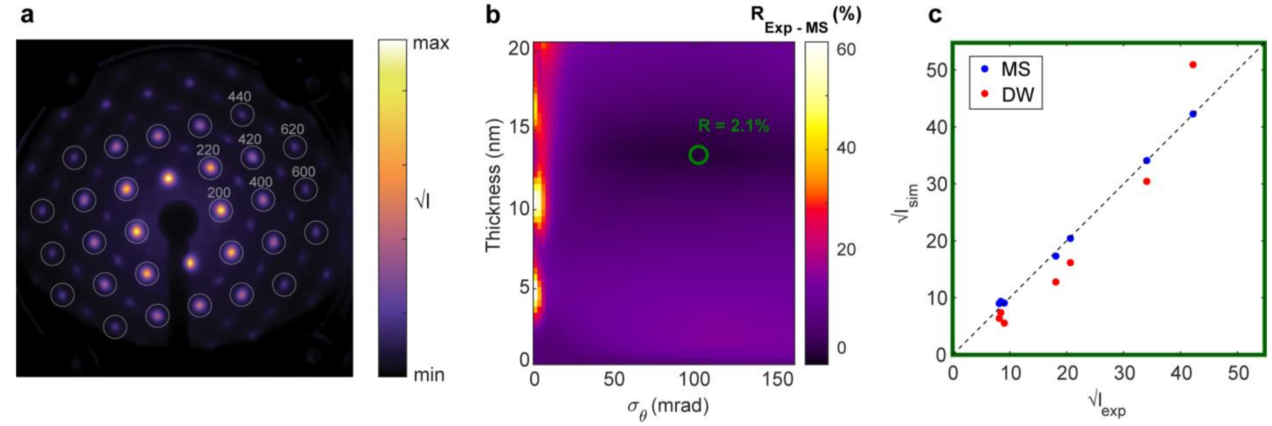

Figure 1. Quantitative matching of a relativistic UED pattern from an 11 nm single-crystal gold film. a) Measured UED pattern at HiRES using a 750 keV electron beam. The circled peaks from the first seven diffraction orders are included in the quantitative analysis. b) Map of the crystallographic R factor between the experimental and multislice peak intensities over the film thickness and rms tilt spread (σ_θ). c) Scatter plot of the simulated diffraction intensities vs the measured intensities for the best-fit multislice film parameters: thickness = 13.5 nm and σ_θ = 105 mrad.

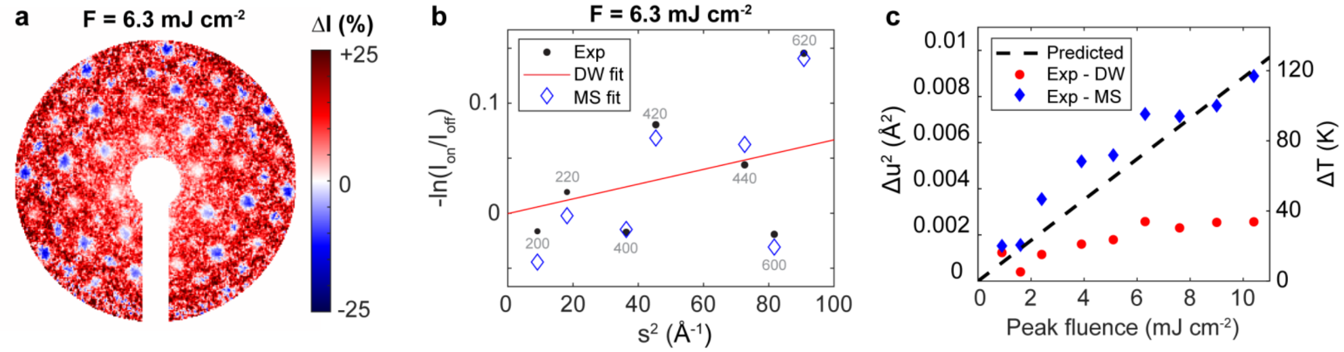

Figure 2. Quantifying light-induced lattice heating from UED of single-crystal gold. a) Photoinduced difference pattern recorded at HiRES using a peak laser fluence of 6.3 mJ cm^{-2}. b) Photoinduced diffraction changes at the same fluence (dots), Debye-Waller fit (line) and multislice fit (diamonds). c) Extracted fluence-dependent increase in total rms atomic displacements (Δu^2) and in lattice temperature (ΔT) using Debye-Waller (DW) and multislice (MS) models. Predicted changes based on known optical constants of gold are superimposed as a dashed line for comparison.

References:

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