Coherent dynamics of structural symmetry during the ultrafast melting of a charge density wave

T. Huber[1], S.O. Mariager[2], A. Ferrer [1,2], H. Schaefer [3], J.A. Johnson [2], S. Gruebel [2], A. Luebcke [2,5], A. Caviezel [2], L. Huber [1], T. Kubacka [1], C. Dornes [1], C. Laulhe [6], S. Ravy [6], G. Ingold [2], P. Beaud [2], J. Demsar [3,4], and S.L. Johnson [1]

[1]Institute for Quantum Electronics, Physics Department, ETH Zurich, CH-8093, Switzerland
[2]Swiss Light Source, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland
[3]Physics Department, Universitaet Konstanz, D-78457, Germany
[4]Institute of Physics, Ilmenau University of Technology, D-98693 Ilmenau, Germany
[5]Laboratoire de Spectroscopie Ultrarapide, EPF Lausanne, CH-1015 Lausanne, Switzerland and
[6]Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin, BP 48, FR-91192 Gif-sur-Yvette Cedex, France
E-mail: tihuber@phys.ethz.ch

Abstract: We use time-resolved hard x-ray diffraction to directly follow the dynamics of structural symmetry change during the ultrafast melting of a charge density wave. We observe a transient recovery of the periodic lattice distortion on a sub-picosecond timescale.
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1. Introduction

In strongly correlated electron systems, the coupling of the lattice to long-range electronic order can lead to a break of structural symmetry upon cooling below the critical temperature. In prototypical charge density wave (CDW) compounds, this leads to a low temperature periodic lattice distortion with a modulation vector reproducing the Fermi-nesting vector of the system [1]. The ultrafast melting of electronic order driven by femtosecond laser pulses has been studied in various systems [2-5], but we still lack crucial direct information about the dynamics of structural symmetry associated with CDW melting. Here we show that with sufficient space and time resolution in a time-resolved hard x-ray experiment, we can observe the coherent structural dynamics associated with CDW formation. As a model system, we choose the prototypical one-dimensional CDW compound $K_{0.3}MoO_3$ (usually termed Blue Bronze), widely regarded as a textbook system for Fermi-nesting driven CDW formation. In $K_{0.3}MoO_3$, an incommensurate CDW and an accompanying periodic lattice distortion occurs below $T_c = 183$ K [6,7].

2. Experimental

The measurements were carried out at the hard x-ray slicing source FEMTO at the Swiss Light Source (SLS) of the Paul-Scherrer-Institut in Villigen, Switzerland [7]. We employed a pump-probe scheme, using 1.5 eV pump pulses to excite a single crystal $K_{0.3}MoO_3$ cooled well below the metal-to-CDW transition temperature to T = 95 K. In the low temperature phase, the periodic lattice modulation leads to superlattice (SL) reflections that can be directly measured with the 7 keV x-ray pulses provided by the synchrotron slicing source. The x-ray pulse duration (FWHM) is around 100 fs, assuring sufficient time-resolution to resolve all relevant collective excitations of the CDW ground state. The measurements were carried out in a grazing incidence geometry, allowing for a close match of pump and probe penetration depth. The time evolution of the measured SL reflection gives a direct view of the nonequilibrium structural dynamics and is not distorted by electronic relaxation processes right after intense photoexcitation.

3. Results and discussion

In Fig. 1, we present the measured coherent structural dynamics after photoexcitation for all relevant excitation fluences. For fluences below the melting threshold of the CDW condensate (F = 0.3 mJ/cm^2), we can directly follow the structural dynamics associated with the collective amplitude mode (AM) of the system. The relaxation timescale of the non-oscillatory signal is $t_{disp} = 3$ ps. Upon increasing the fluence to F = 1 mJ/cm^2, t_{disp} increases to around 10 ps, and we do not observe a clear signature of the AM. The timescale of the initial drop of SL diffraction intensity is around 100 fs, limited by the experimental time resolution. For higher fluences (F = 2.7 mJ/cm^2) well above the melting threshold [3], we observe a transient recovery of the SL diffraction intensity, peaking at a delay of around t = 350 fs. After the transient recovery, we do not observe a clear relaxation of the diffraction signal on the measured timescale. The background level for time delays t > 0.5 ps with increasing fluence saturates at a background level of $I(t)/I_0 \sim 0.4$ that we attribute to diffraction of non-excited surface regions of the cleaved single crystal. A striking

property of the transient recovery is its timescale, which is faster than dynamics that could be associated with the AM.

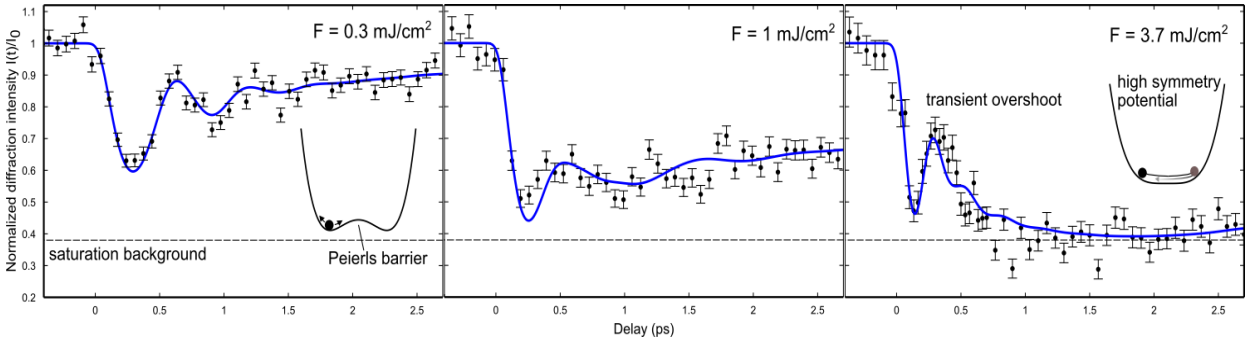

Fig. 1. Time evolution of (1 3.25 -0.5) SL peak diffraction intensity, normalized to the equilibrium diffraction intensity I_0. The dashed line depicts the saturation background. Errorbars correspond to photon counting statistics. Lines are fits to the model as described in the text. The low fluence data shows a clear signature of the soft mode of the system at 1.62 THz. For the highest fluence, we observe a transient overshoot.

We can model the results as a coherent motion along the structural coordinate of the Peierls distortion in a fluence dependent double-well potential: For low fluences, the system oscillates around low-temperature equilibrium positions. For fluences above the melting threshold, the Peierls barrier collapses immediately, leading to a coherent motion in a high symmetry potential landscape. The measured diffraction signal in our experimental geometry then results from an incoherent superposition of the signal with different excitation levels of the inhomogeneously excited crystal. In Fig 1., we show fits to the data according to the model, where the only fluence dependent parameters are the height of the Peierls barrier and the relaxation timescale t_{disp}. To account for the evolution of the transient overshoot after the first peak, we introduce a phenomenological time-dependent damping parameter describing the damping of the motion along the distortion coordinate in the transient state after intense photoexcitation.

The agreement with the model indicates that during the ultrafast melting of a CDW, the structural dynamics are determined by the properties of the high-symmetry phase and not by the lattice modes of the initial state. This could have wide implications for the understanding of materials with similar periodic lattice modulations that are the result of a coupling of the lattice to electronic order. Ultimately, the results help explain how the structural symmetry dynamics associated with so-called nonthermal phase transitions can be fast, as opposed to the much slower behavior observed in adiabatic soft-mode phase transitions.

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