ELECTRONIC THERMAL EXPANSION AND ITS ROLE IN THE GENERATION OF COHERENT ACOUSTIC PHONONS

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Introduction

Under thermal equilibrium conditions and at or above room temperature, thermal expansion of metals without magnetic ordering is dominated by lattice heating. This is largely due to the fact that the lattice heat capacity is about two orders of magnitude larger than that of conduction electrons. However, under highly non-equilibrium conditions created by the fs optical excitation, the conduction electrons are transiently heated well above the lattice temperature and thus contribute significantly to thermal expansion. Such an example reported here is the generation of coherent acoustic phonons by fs optical pulses. By probing the associated thermal expansion dynamics in real time with femtosecond electron diffraction (FED), we are able to differentiate the role of hot electrons from that of lattice heating and directly measure their contributions to driving coherent acoustic phonons. The results provide unambiguous evidence that electronic thermal expansion contributes significantly to the generation of coherent acoustic phonons.

Results and Discussion

Ultrafast thermal expansion in free-standing polycrystalline Al films of 20-nm thickness was initiated by nearly impulsive excitation with 50-fs near infra-red (790 nm) laser pulses. The associated structural dynamics were monitored by taking snapshots of diffraction patterns at different delay times using transmission femtosecond electron diffraction.

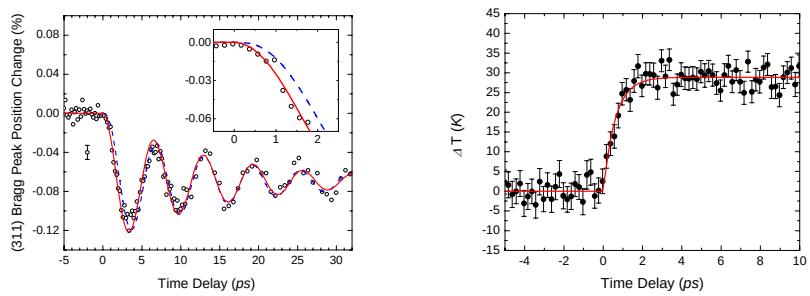

Fig. 1. Lattice dynamics probed with FED. Left panel: Temporal evolution of (311) Bragg peak positions. Positive time delays correspond to probe electron pulses arriving after excitation laser pulses. The solid line is the fit to the experimental data using harmonic oscillator model. The dashed curve is a fit excluding σ_e, which lags behind the data with a phase shift of approximately 18˚. Inset: Detailed view of the above two fitting results in the range of -0.5 to 2.5 ps. Right panel: Temporal evolution of the lattice temperature with a time constant of 600 ± 100 fs for lattice thermalization.

Temporal evolutions of both coherent and thermal lattice motions are shown in Fig. 1. The coherent lattice vibration (coherent phonons), with a period of ~6.4 ps, was centered at a new and expanded equilibrium lattice position, corresponding to an elevated lattice temperature of ~30 K. The lattice temperature displays an exponential temporal behavior with a time constant of ~600 fs.

We show that the above phonon dynamics can be well-fitted with a damped classical harmonic oscillator model using a driving force that includes both electronic (σ_e) and lattice (σ) thermal stresses. Notably, fitting without σ_e generates a significant phase lag of 18 degrees, indicating that the pressure of hot free electrons contributes significantly in driving the coherent acoustic phonons under these conditions when electrons and phonons are not thermalized [1].

Using this transient heating method, we are also able to accurately measure the electronic Grüneisen constant γ_e of aluminum at room temperature using FED [2]. This new approach opens the way to determining the γ_e values in many magnetic materials, such as rare earth elements with low Curie temperatures, where the accurate measurement of γ_e is not amenable with other traditional techniques.

Acknowledgements

The project is supported by the National Science Foundation under the grant number DMR-0305519.

References

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[2] S. Nie, *et al.*, Phys. Rev. Lett., in press (2005).