DOI: 10.1017/S1431927605506081
Microsc Microanal 11(Suppl 2), 2005

Femtosecond Electron Diffraction: Probe and Control of Ultrafast Structural Dynamics

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Atomic motions on the time scale of single vibrational period determine the evolution of new phases in solids, the pathways of chemical reactions, and the efficiency of biological function. The techniques to study the structural dynamics on this fundamental level are being developed using both time-resolved x-ray and electron diffraction. Here we report a direct and real-time probe and control of lattice dynamics in thin-film aluminum using femtosecond electron diffraction (FED) [1-2] with sub milli-ångström spatial resolution on the femtosecond time scale. The experiment was conducted in our newly developed FED apparatus [3]. The structural dynamics were initiated in a free-standing polycrystalline thin-film Al of 20-nm thickness with 50-fs near infra-red (790 nm) laser pulses, and then probed by taking snapshots of transmission diffraction patterns at different time delays using 350-fs electron pulses (see Fig. 1).

The temporal evolution of (311) peak position and lattice temperature are shown in Fig. 2. It is clear that the coherent lattice motions, centered at a new and expanded equilibrium lattice position, were launched immediately after the optical excitation with a concurrent heating of the lattice. The new lattice position, established at an elevated lattice temperature after electron-phonon thermalization at ~5 ps delay time, corresponds to a lattice temperature jump of ~40 K. The displacements displayed in the figure represent an averaged value across the 20-nm film, corresponding to ~100 fm lattice plane space change at maximum displacement at ~6 ps delay time. The solid curve is a fit to the experimental data with a model incorporating both the finite stress build-up time and the subsequent damped oscillation. The oscillatory part of the fit gives a vibrational period of 8.1 ± 0.1 ps (~0.12 THz) and a damping time constant of 19.0 ± 2 ps. The same fitting to the data for all other Bragg peaks (data not shown) yielded essentially the same values for the period and damping time, which indicates a coherent acoustic wave was generated after the optical excitation.

In addition, the quantitative analysis using a simplified model of damped harmonic oscillator indicates that the driving force (stress) starts well before the lattice reaches its final equilibrium temperature, providing a direct and clear evidence of the non-thermal generation mechanism of coherent acoustic phonons.

To coherently control the lattice motions, we use two excitation laser pulses separated by a well defined time delay. As shown in Fig. 3, by setting the time delay equal to one vibrational period, a constructive interference was created between the two acoustic waves in the film. Compared with the single pulse data (upper curve), the vibrational amplitude was nearly doubled at a given time delay. In contrast, by setting a relative delay of half vibrational period and the second pulse energy to about 90% of the first pulse to match the amplitudes of the two waves at a half vibrational period, the acoustic wave was completely silenced due to the out-of-phase destructive interference.

References

[1] H. Park, Z. Hao, C. Tao, F. Popescu, and J. Cao, in press, *Ultrafast Phenomena XIV*. 2004, Springer: Berlin.

[2] H. Park, S. Nie, X. Wang, R. Clinite, and J. Cao, submitted for publication

[3] J. Cao, Z. Hao, H. Park, C. Tao, D. Kau, and L. Blaszczyk, "Femtosecond electron diffraction for direct measurement of ultrafast atomic motions," Applied Physics Letters 83, 1044-1046 (2003)

[4] This work was supported by the National Science Foundation under the Contract No. DMR-0305519. The help of Dr. Z. Hao, C. Tao, R. Clinite and Dr. F. Popescu is gratefully acknowledged.

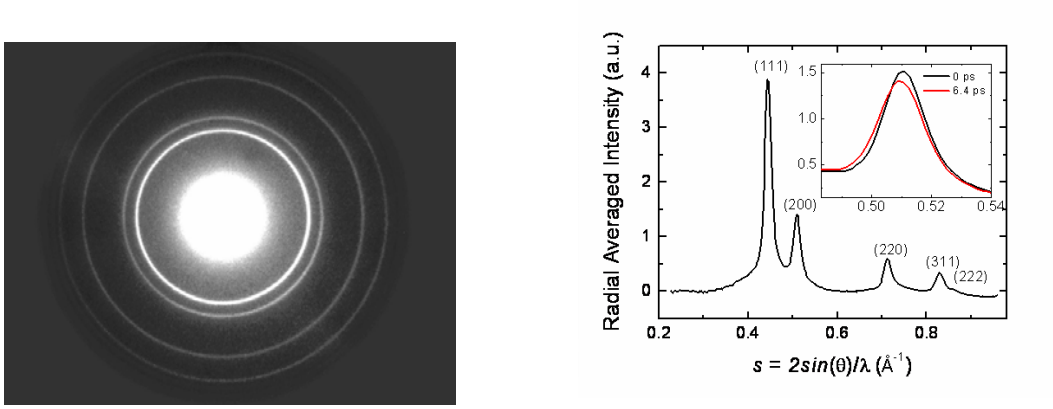

Fig. 1 Left Panel: Diffraction pattern of a 20-nm thick thin-film aluminum. It was recorded with fs electron pulses (~1500 e^-/pulse), at laser excitation fluence of 2.3 mJ/cm^2 and with 25-s integration time. Right Panel: The corresponding radial averaged intensity curve. Inset: Detailed view of intensity profile of (200) peak at two different time delays of 0 and 6.4 ps, respectively. The laser-induced Bragg peak position change and intensity drop are clearly seen.

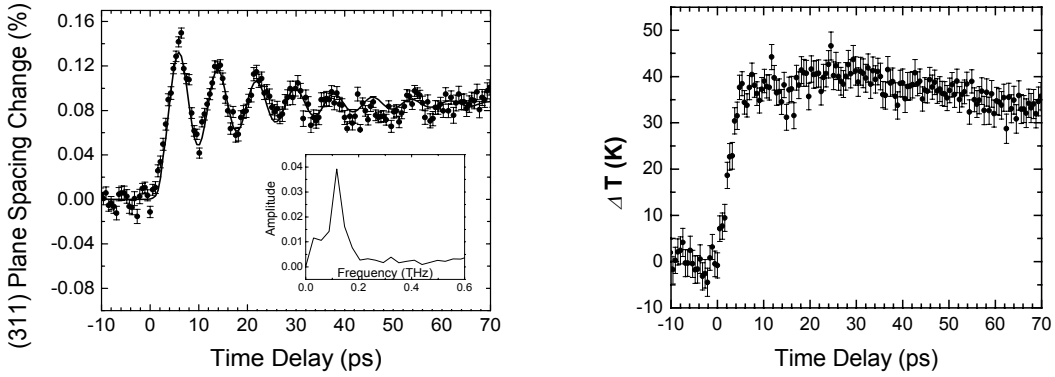

Fig. 2 Left Panel: The temporal evolution of (311) crystal plane spacing. The solid curve is a fit to the experimental data. Inset: The Fourier transform. The peak is centered at ~0.12 THz, corresponding to a 8-ps vibrational period. Right Panel: The temporal evolution of lattice temperature.

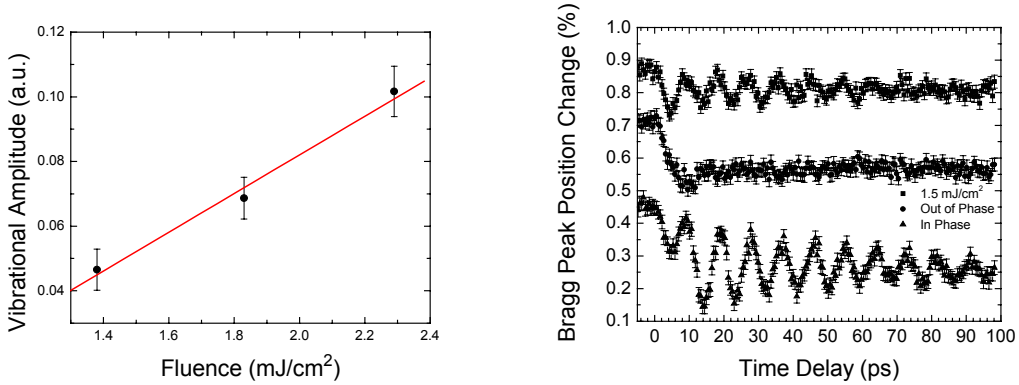

Fig. 3 Left Panel: The amplitude of coherent vibration as a function of excitation laser fluence. Right Panel: Coherent control of lattice vibrations using a pair of excitation laser pulses.

https://doi.org/10.1017/S1431927605506081 Published online by Cambridge University Press