Femtosecond Electron Diffraction Probing of Lattice Dynamics

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Structural Dynamics of Thin-Film Al

- Mono-atomic simple fcc structure, *a* = 4.05 Å
- Long MFP > 50 nm for 60 keV electrons
- Kinematical diffraction theory

Ideal system for quantitative study of structural dynamics with fs electron diffraction

Results of Previous Studies

Non-thermal melting (optical, ~ 500 fs)

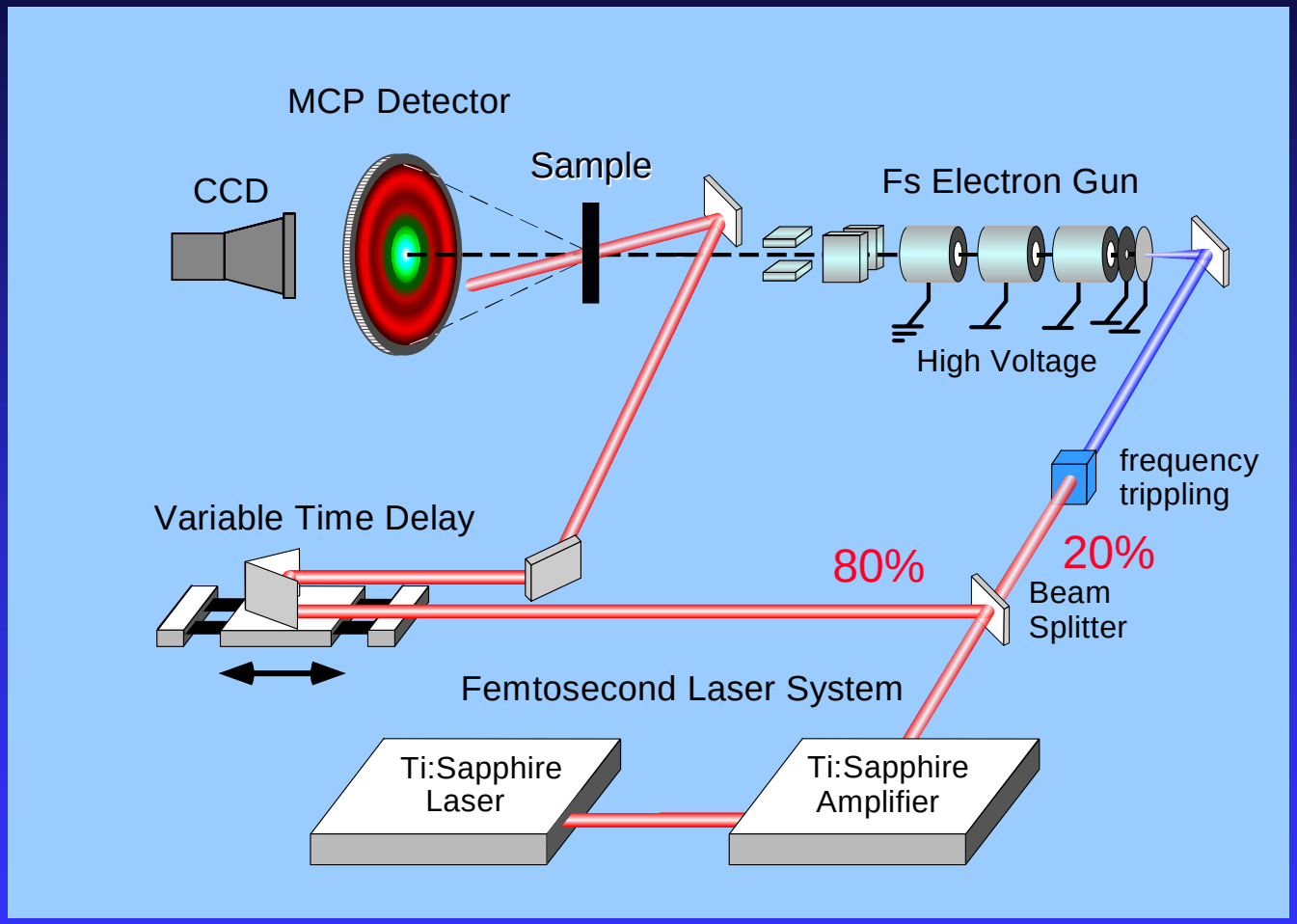
C. Guo, *et al., Phys. Rev. Lett.* **84,** 4493 (2000)

Strongly-driven thermal melting (diffraction, ~ 3.5 ps)

B. J. Siwick, *et al.*, *Science* **302,** 1382 (2003)

Neither study provide the detailed information on the atomic motions during melting

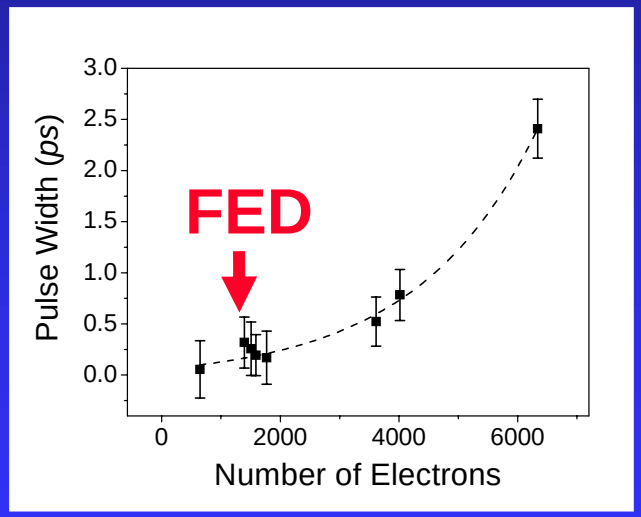
Femtosecond Electron Diffraction Setup

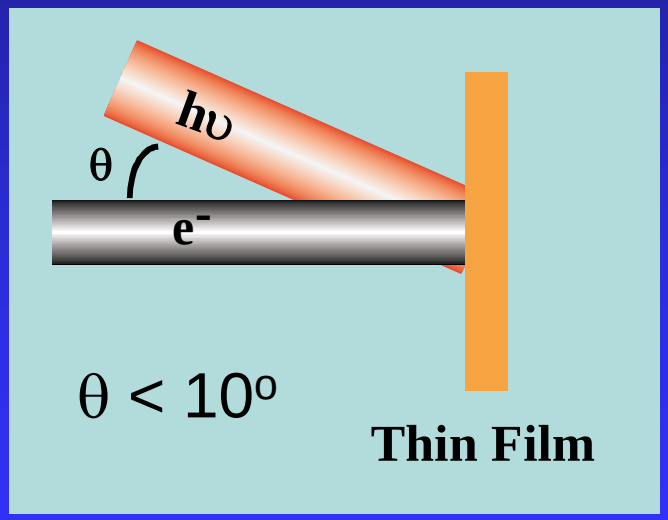


J. Cao, *et al.*, *Appl. Phys. Lett.* **83**, 1044 (2003)

Maintain Optimal Temporal and Spatial Resolution

- 300-fs electron pulse (<1600 e^-/pulse), small crossing angle
- Low excitation fluence ~ 2 mJ/cm^2 , no sample damage
- 1 KHz reprate and long data acquisition time for better SNR

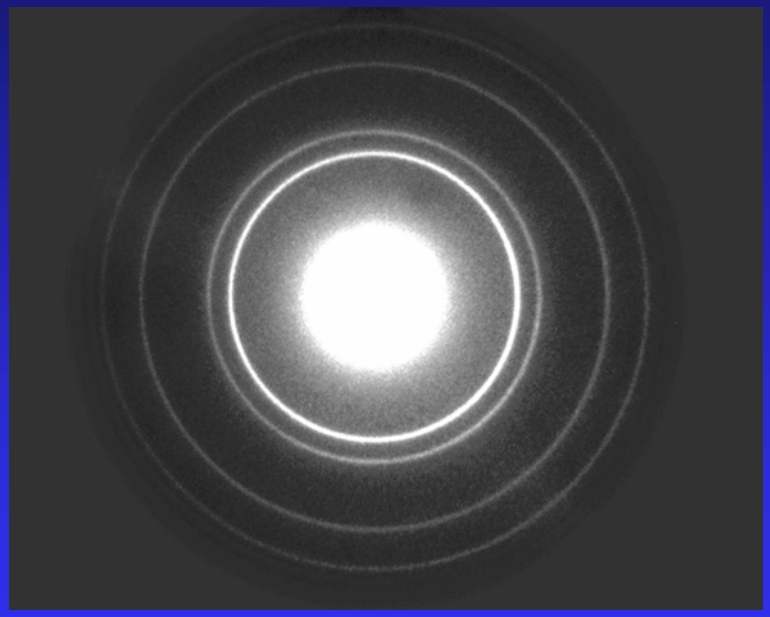




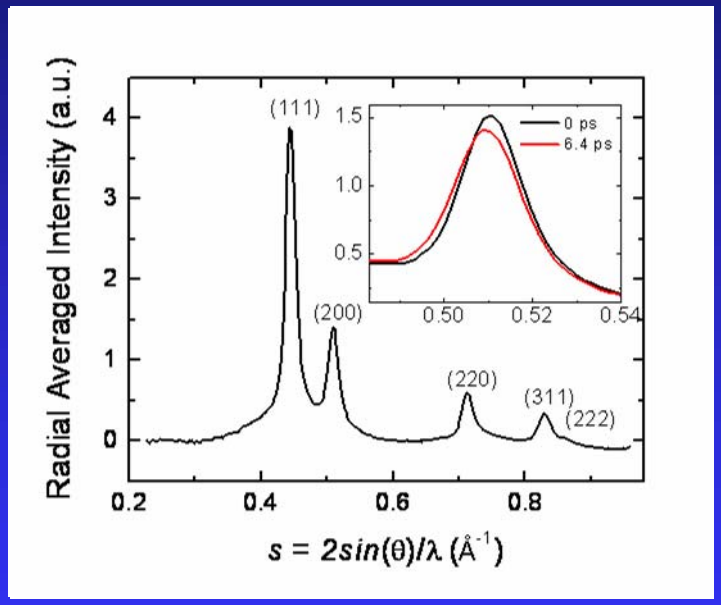
- Temporal resolution: ~400 fs
- Spatial resolution: sub-milli-angstrom

Diffraction Pattern of 20-nm Al Film

2D diffraction pattern



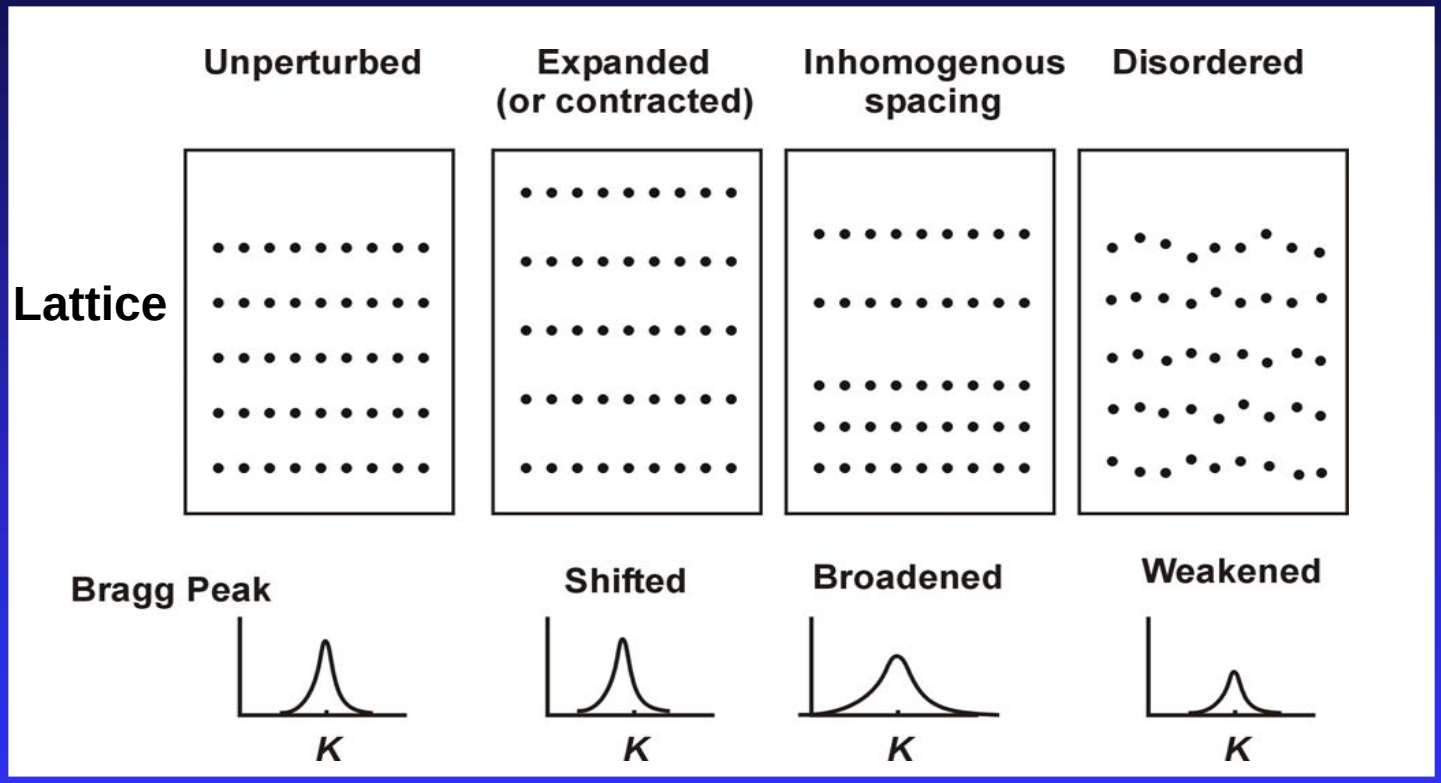
Intensity curve



Convert 2-D diffraction data to 1-D intensity curve

Laser fluence ~ 2.3 mJ/cm^2 , 25 s integration, ~1500 e/pulse and time step ~ 530 fs

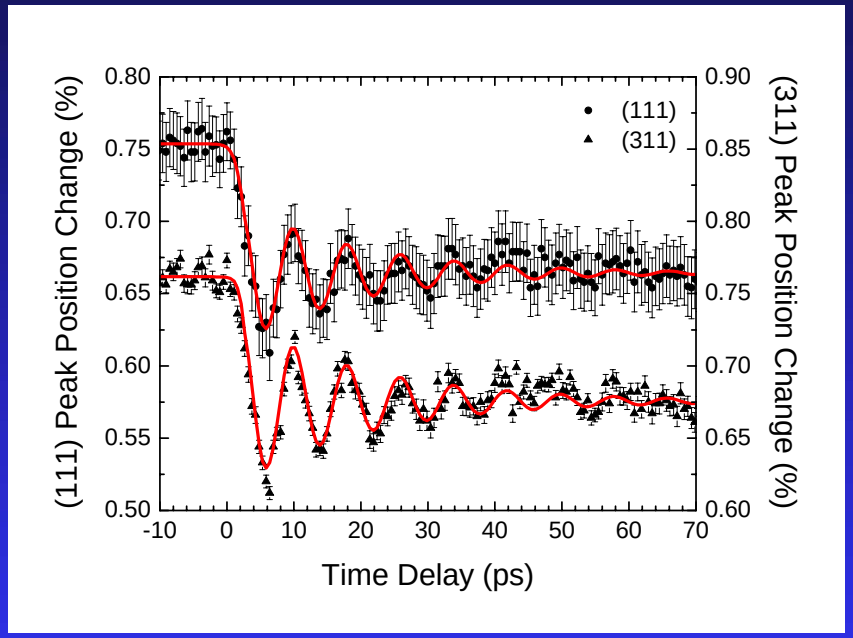
Structural Changes Probed with Diffraction

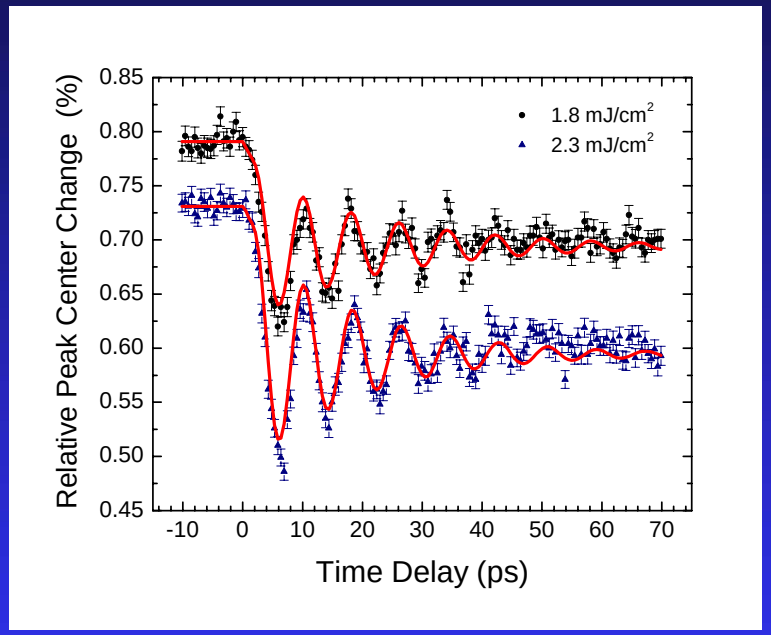


Three aspects of Bragg peak (position, intensity and width) give detailed knowledge of structure change.

Reproduced from: A. Rousse, *et al., Rev. Modern Phys.,* 73, 17(2001)

Temporal Evolution of Bragg Peaks





- Coherent and in-phase motions
- Amplitude is proportional the laser fluence
- 8 ps period and damping time constant~ 20 ps

Laser-Induced Breathing Motion of Lattice along Surface Normal

$$\Psi(x,t) = A(t)\cos(\pi x / L)\cdot\cos(\pi v t / L)$$

$$0 \le x \le L \text{ and } T = 2L/v.$$

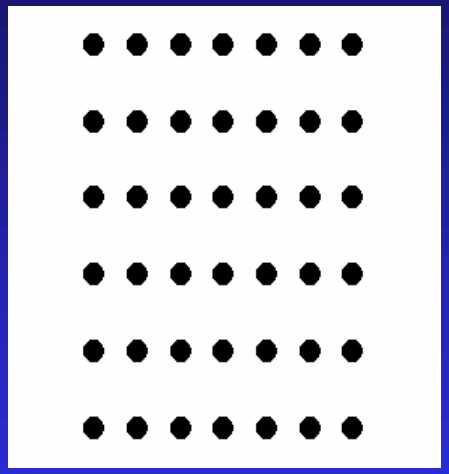
- impulsive and displacive excitation < vibrational period
- the open boundary 1-D standing wave
- breathing motions along the surface normal
- Vibrational period:

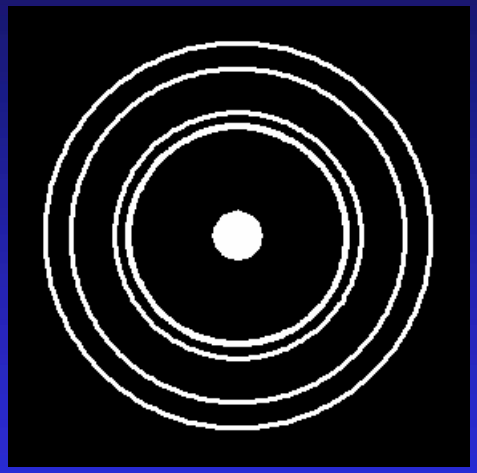
given $v = 5000$ m/s and $L = 20$ nm, $T = 2L/v = 8$ ps

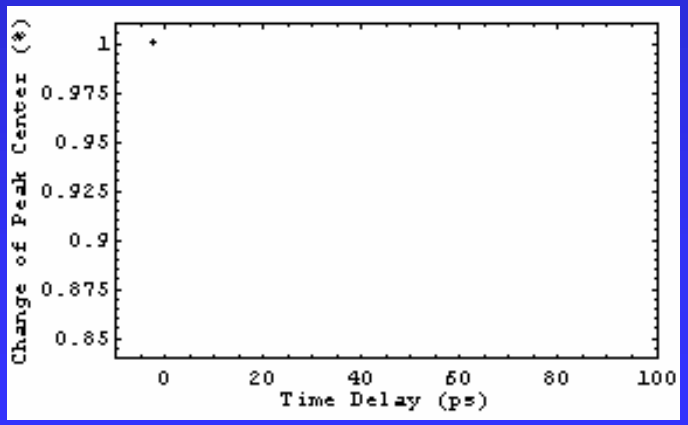
in excellent agreement with FED data

Animation of Breathing Motion

Laser



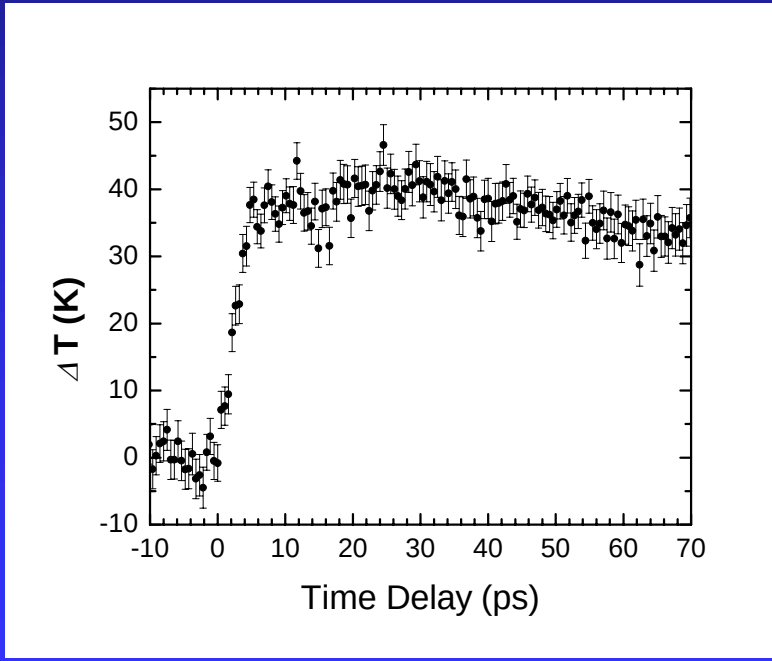




Temporal Evolution of Lattice Temperature

Debye-Waller factor: $I(T)=I_o \exp[-a(h^2+k^2+l^2)T]$

- Lattice reaches equilibrium T ~ 5 ps
- Vibration nearly reach the first maximum displacement
- Two modes of atomic motion launched concurrently



Atomic-Level View of Laser-Induced Lattice Dynamics

Both coherent and thermal lattice motions start simultaneously. A coherent lattice vibration with a period of 8 ps starting immediately after the optical excitation with a concurrent heating of the lattice, reaching its final equilibrium temperature ~ 5 ps later.

TTM Overestimate the e-ph Coupling

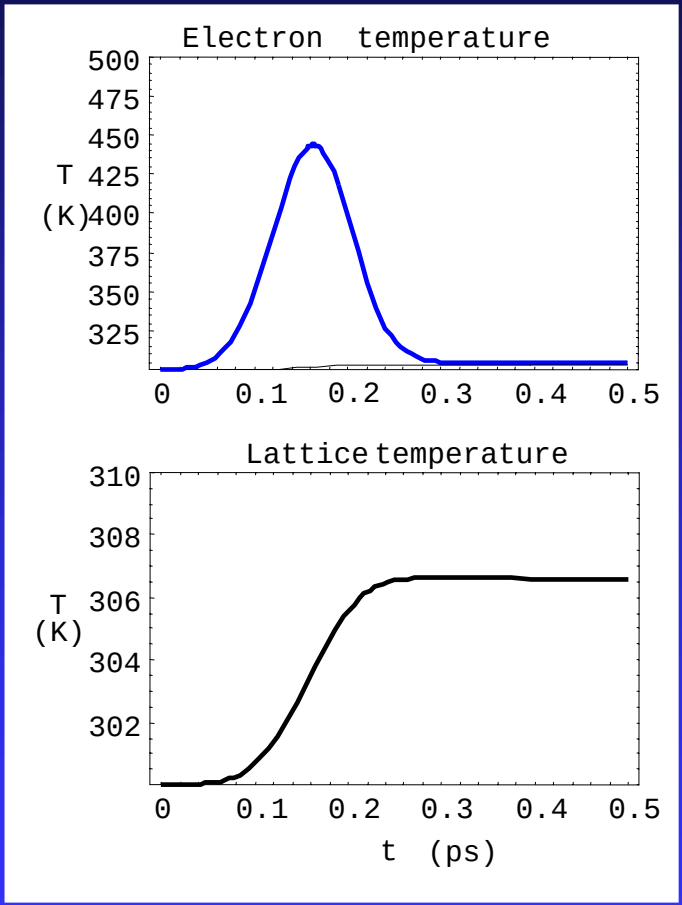
Two temperature model (TTM)

Electron: T_e

Phonon: T_l

Coupling constant: G

$$\begin{cases} C_e(T_e)\dfrac{\partial T_e}{\partial t} = K\nabla^2 T_e - G(T_e - T_l) + P(x,t) \\ C_l \dfrac{\partial T_l}{\partial t} = G(T_e - T_l) \end{cases}$$



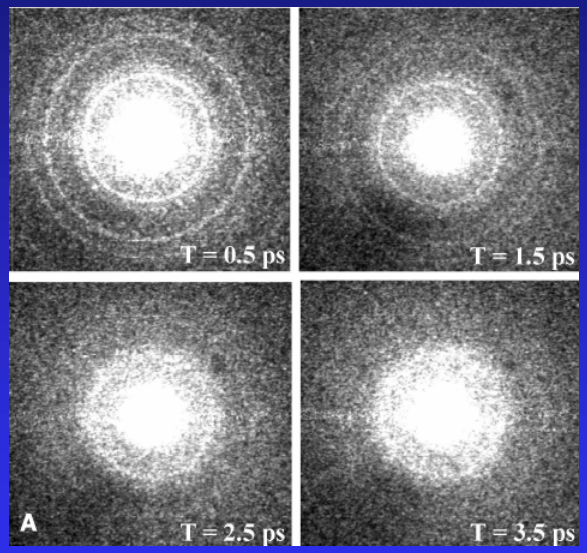
Lattice thermalization: ~300 fs

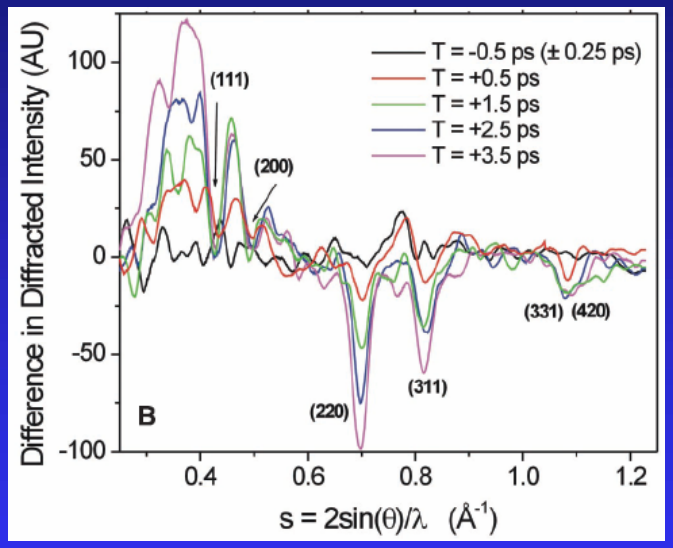
For ~ 5 ps lattice thermalization:
a G ~10 times smaller is needed

G. Tas & H. J. Maris, *Phys. Rev. B* **49,** 15046 (1994)

Previous Time-Resolved Electron Diffraction Study

Strongly-driven thermal melting (superheating)

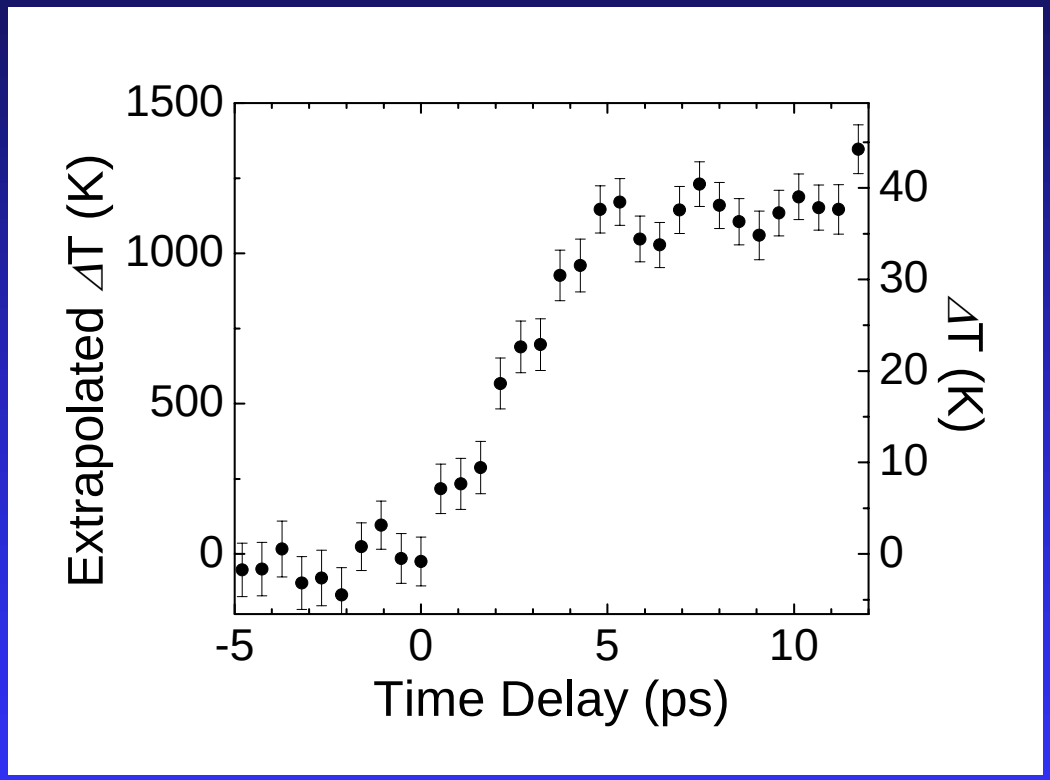




3.5 ps melting time at laser fluence 70 mJ/cm^2
3.5 ps > 1.5 ps e-ph thermalization (~ 1500K)
predicted by TTM simulation

B. J. Siwick, *et al*., *Science* **302,** 1382 (2003)

Extrapolated Lattice Temperature at Fluence of 70 mJ/cm^2



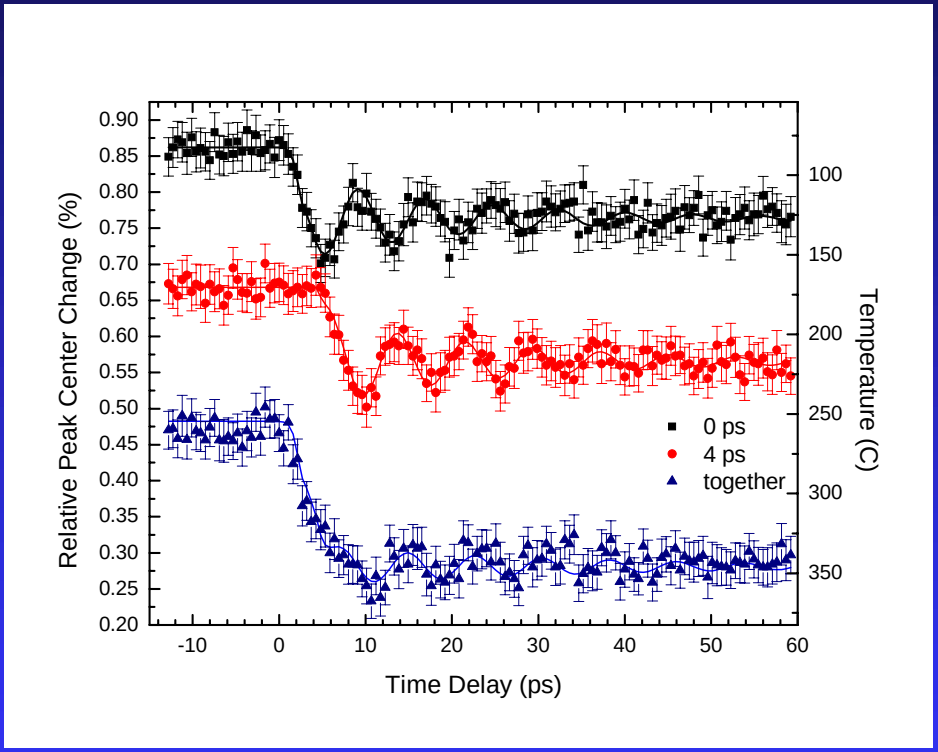
At 1.5 ps, T_l ~700 K < melting temp. 933K
Superheating highly unlikely

Proposed Melting Mechanism

The large coherent atomic motions weaken the lattice structure significantly. Under this condition, less atomic random motions (lower temperature), less than that are required in a normal thermal melting, led to the loss of the lattice long-range order. This interplay of coherent and thermal atomic motions melts the lattice within the first half cycle of coherent vibration.

A. M. Lindenberg, *et al. Phys. Rev. Lett.* **84,** 111 (2000)
K. Sokolowski-Tinten, *et al*. *Nature* **422**, 287 (2003)

Coherent Control of Lattice Motions



Using a sequence of pulses to control both phase and amplitude of lattice motion

Summary

- FED capable of measuring ultrafast structural dynamics on the fundamental timescale of single atomic vibrational period
  - Time resolution ~ 400 fs
  - Sub-milli-angstrom spatial resolution
- The structure dynamics of Al film
  - Coherent and thermal motions
  - Coherent control of atomic motions

Acknowledgments

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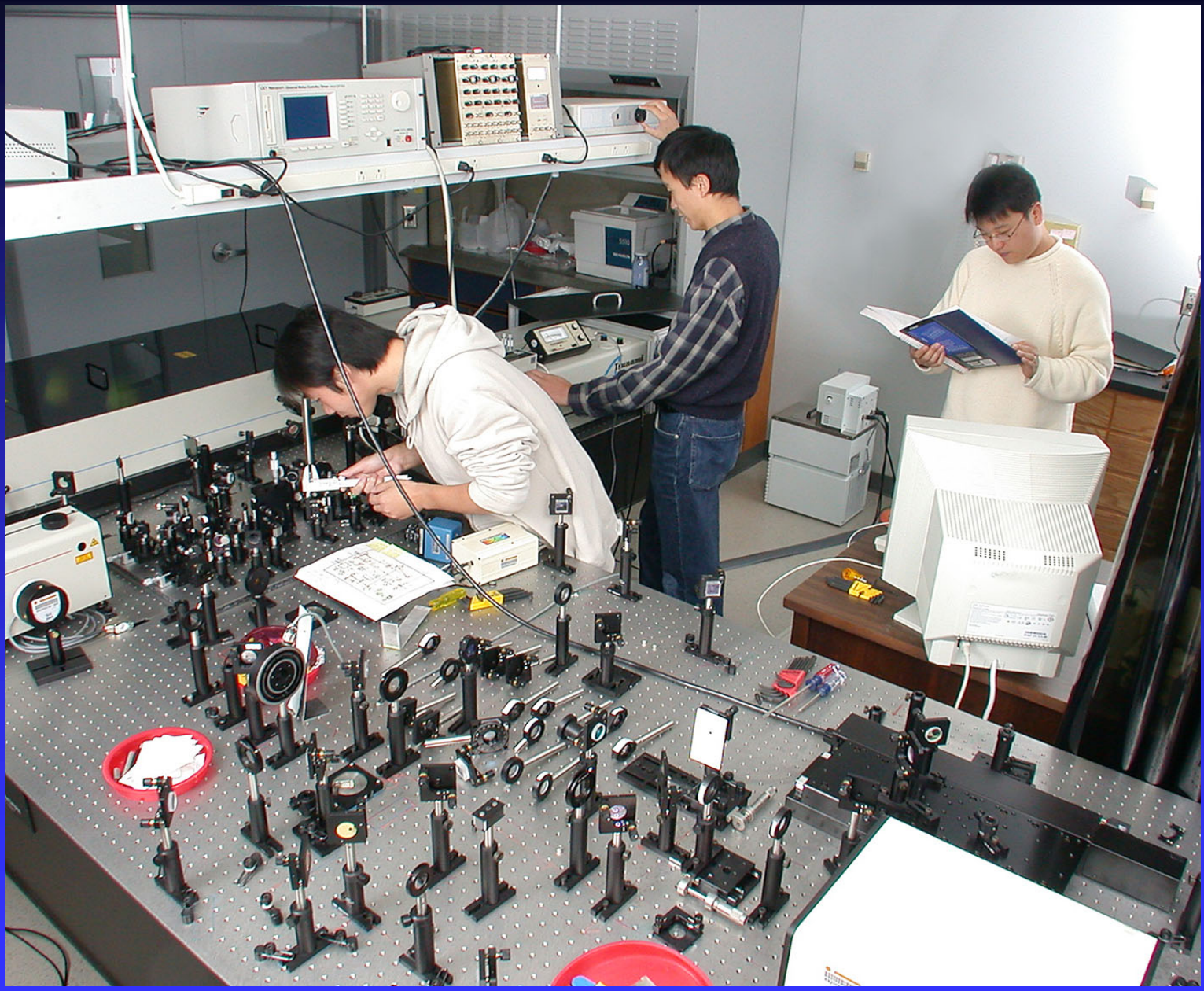
Physics & NHMFL Machine Shops

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