Ultrafast structure dynamics in metal films

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Outline

- Dynamics of thermal expansion induced by ultrafast heating
- Measuring electron Grüneisen parameter with UED
- Ultrafast demagnetization in Ni
- Summary and acknowledgements

Ultrafast Heating of Metal Film

Ultrafast excitation break the thermal equilibrium: $T_e \neq T_l$

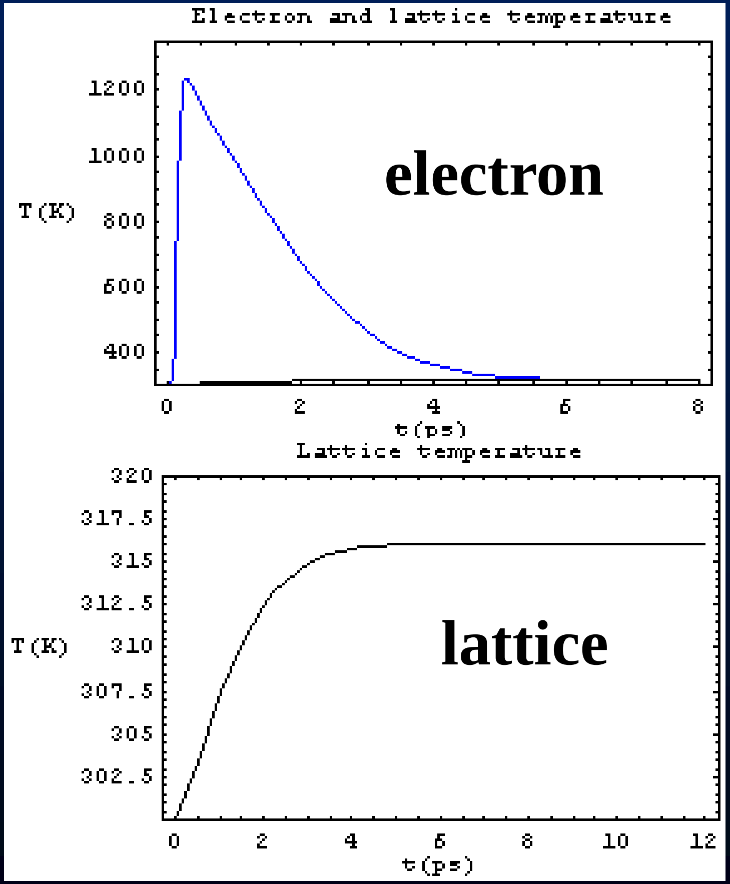
Two coupled subsystems

Electron: T_e

Phonon: T_l

Coupling constant: G

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

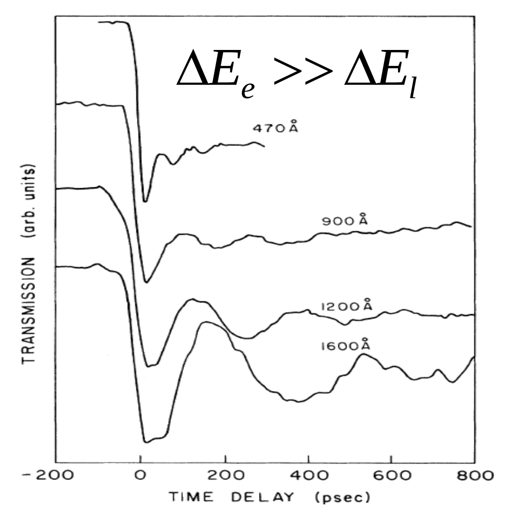


e-ph thermalization: ~1-3 ps

Study the lattice response to the fs laser ultrafast heating

Ultrafast Heating of thin film and nano-particles

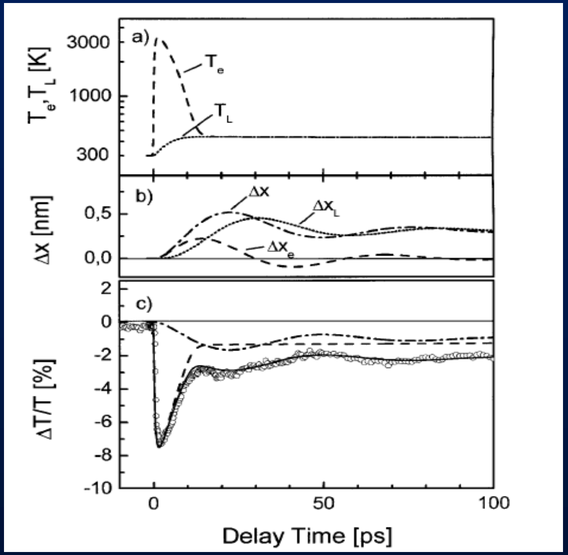
Thin Al films



C. Thomsen, et al, PRL 53, 989-992 (1984)

- the stress contains both electron contribution and lattice contribution.

Ag nano-particles



M. Perner, et al, PRL 85, 792-795 (2000)

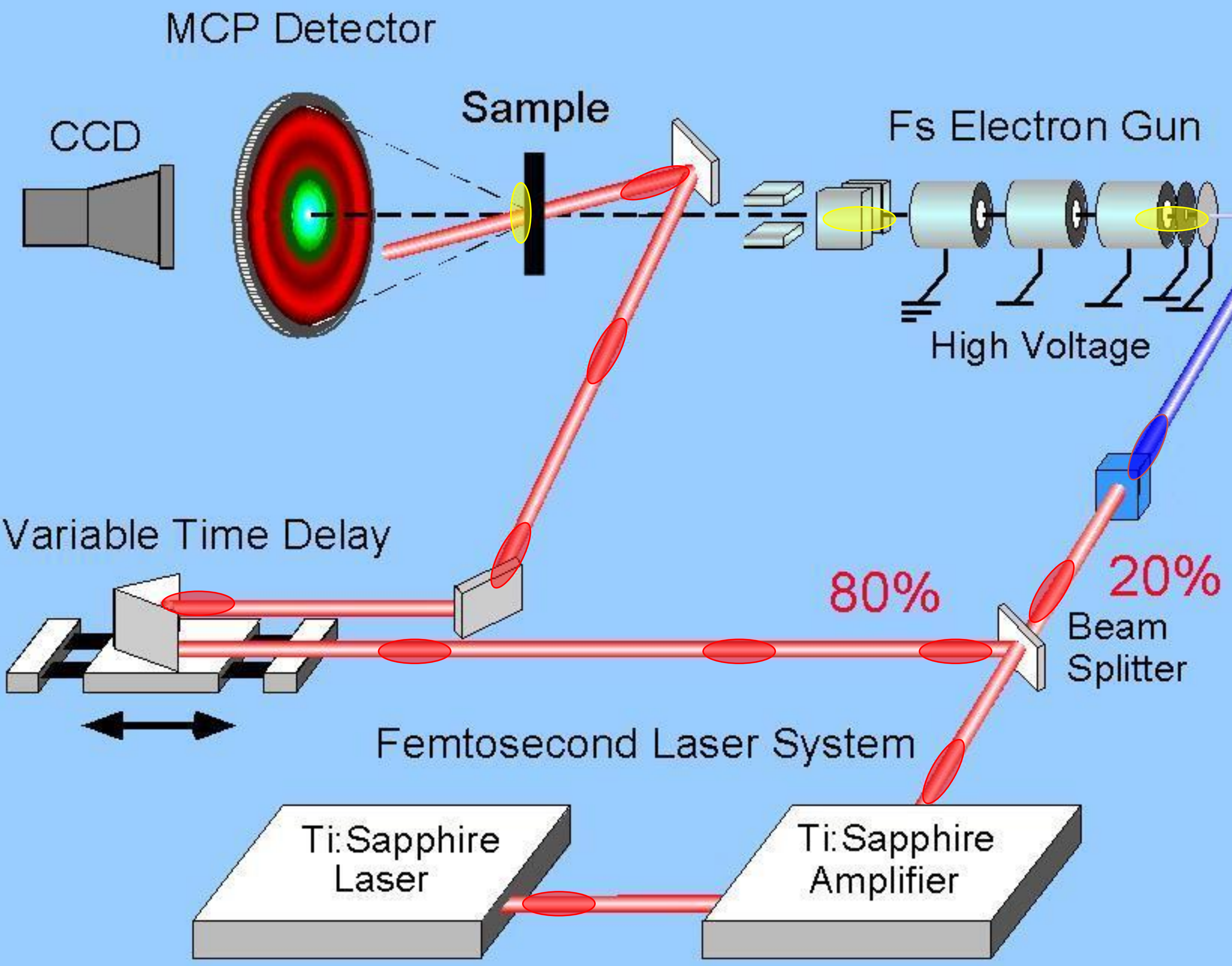
- The stresses are calculated from Two Temperature Model
- Hot electrons make significant contribution

A direct measurement of lattice temperature is not achieved

Some Outstanding Issues

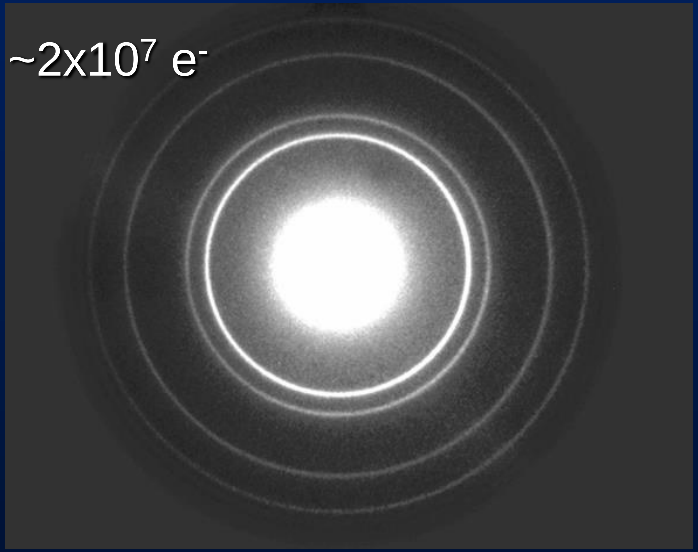
- How does lattice response to ultrafast heating
- Correlation between thermal and coherent motions
- The role of hot electrons in thermal expansion

UED: a direct probe of structural dynamics, record both coherent and thermal lattice motions simultaneously in real time

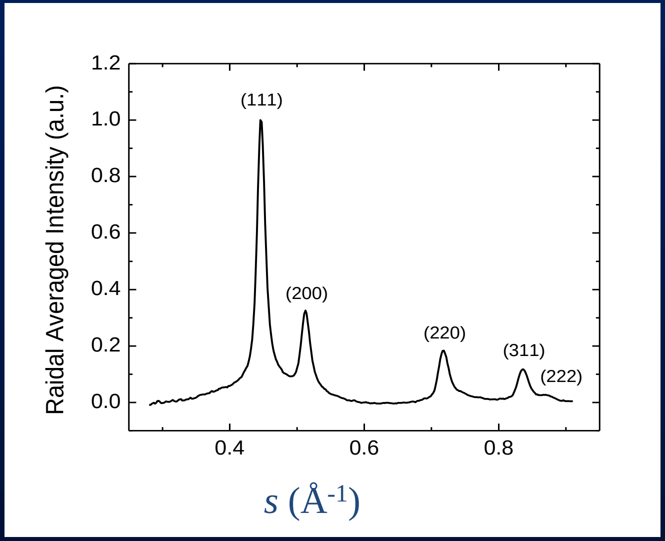

MCP Detector
CCD
Sample
Fs Electron Gun
High Voltage
Variable Time Delay
80%
20%
Beam
Splitter
Femtosecond Laser System
Ti:Sapphire
Laser
Ti:Sapphire
Amplifier

Diffraction pattern of 20-nm Al film

2D diffraction pattern



Intensity curve



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

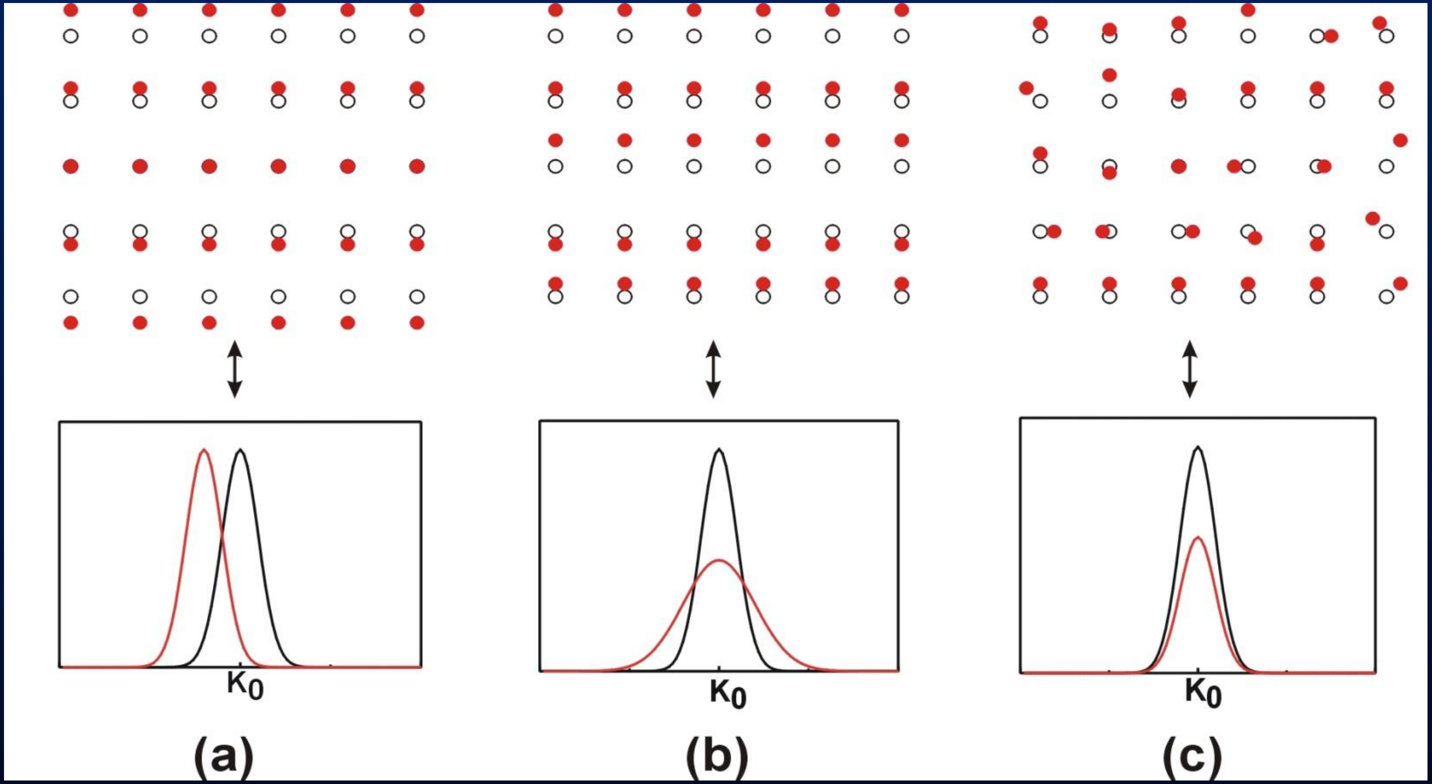
Pump: fluence ~ 2.3 mJ/cm^2 , 2 mm beam size

e-beam: 60 keV, <1000 e/pulse, ~400fs , ~300 μm

Following the structural dynamics by taking snapshots of diffraction patterns at different delay times

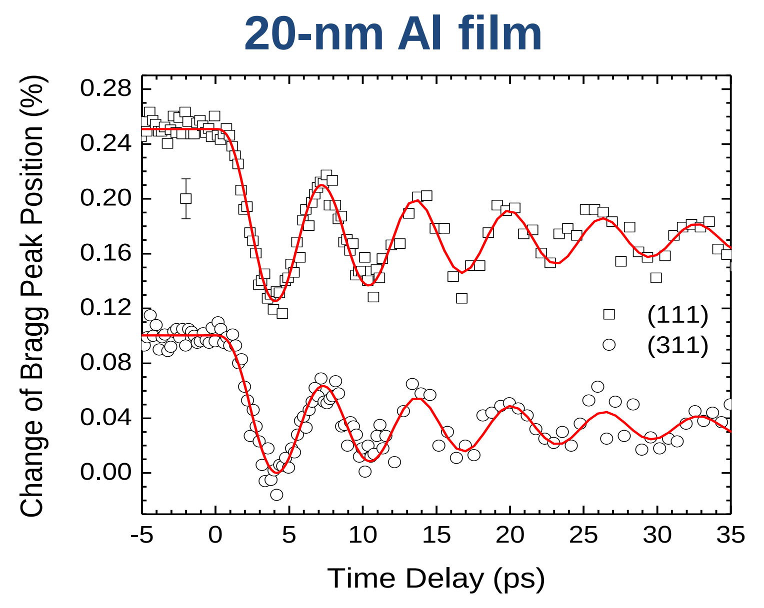
Structural changes probed with diffraction

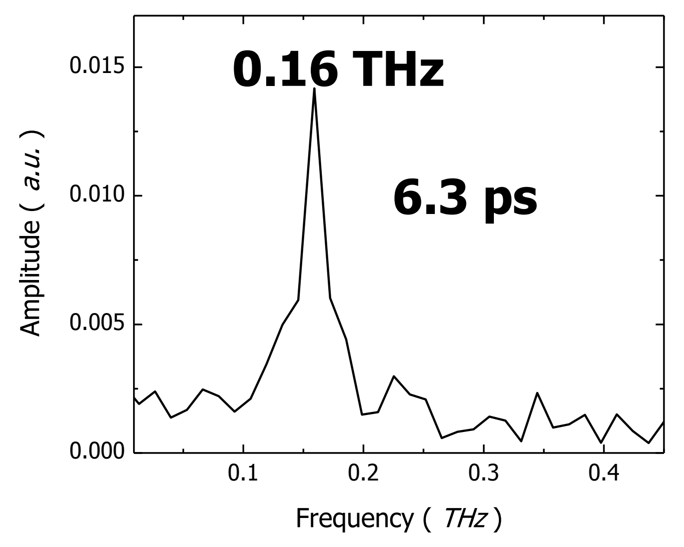
Expansion **Inhomogenous spacing** **Disorder**



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

Coherent lattice motions: breathing motion along surface normal



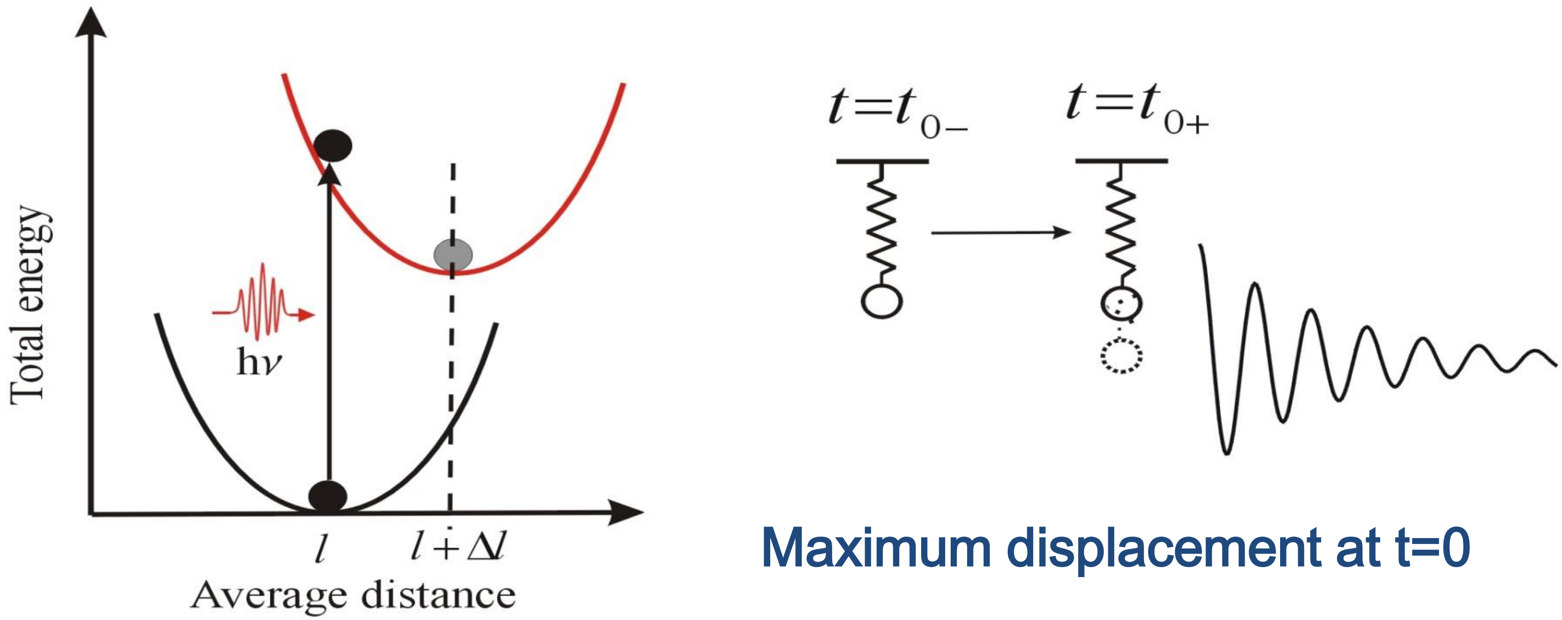


- Coherent and in-phase motions of all Bragg peaks
- Single mode with vibrational period *T* ~ 6.3 ps, standing wave
- maximum displacement at time zero
- Detection sensitivity: $\Delta r/r$ ~ 0.02 %, < milli-angstrom

H. Park, X. Wang, S. Nie, R. Clinite and J. Cao, *Solid Sate Commun.* **136**, 559-563 (2005)

Displacive excitation of coherent phonon

DECP : Laser energy is deposited into the system quasi-instantaneously and alters the system equilibrium position.



Maximum displacement at t=0

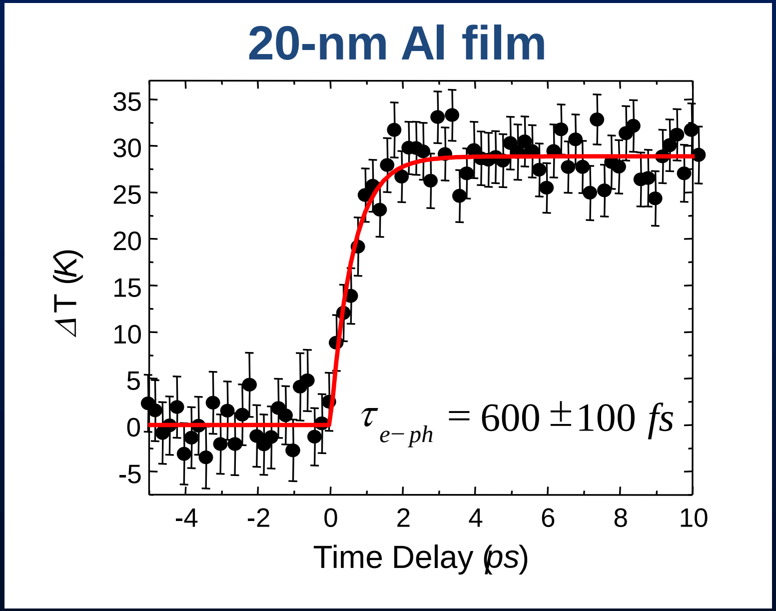
$$\frac{d^2Q}{dt^2}+2\beta\frac{dQ}{dt}+\omega^2 x = F/m = \sigma A/m$$

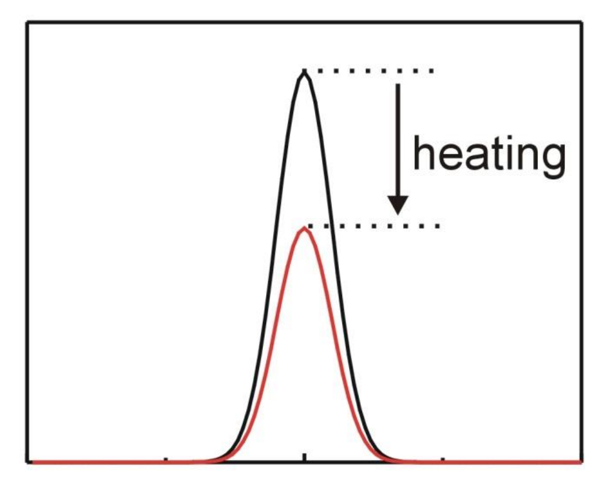
A: surface area

σ: thermal stress

Thermal lattice motions (T_l)←→ thermal stress

Debye-Waller effect: $I_{hkl}(T)=I_o \exp[-a(h^2+k^2+l^2)T]$



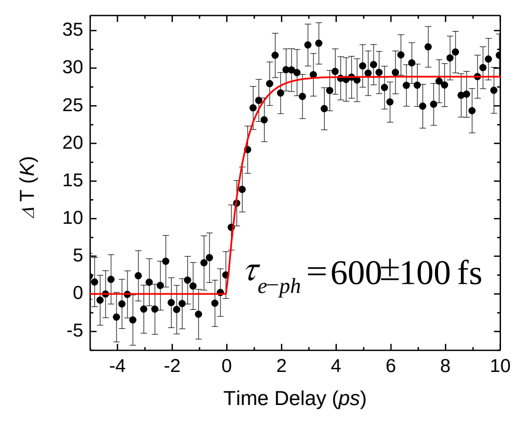


$$\delta T_l = \Delta T(1-e^{-t/\tau_{e-ph}})$$

Debye-Waller effect

H. Park, X. Wang, S. Nie, R. Clinite and J. Cao, *PRB Rapid Comm.* 72, 100301(2005)

Transient Stress of ultrafast heating



$$\sigma = \sigma_e + \sigma_l = \gamma_e \Delta E_e + \gamma_l \Delta E_l$$

$$= \gamma_e C_e \delta T_e + \gamma_l C_l \delta T_l$$

γ_e and γ_l are electronic and lattice Grüneisen constants

C_e and C_l are heat capacities;

ΔE_l and ΔE_e subsystem thermal energy

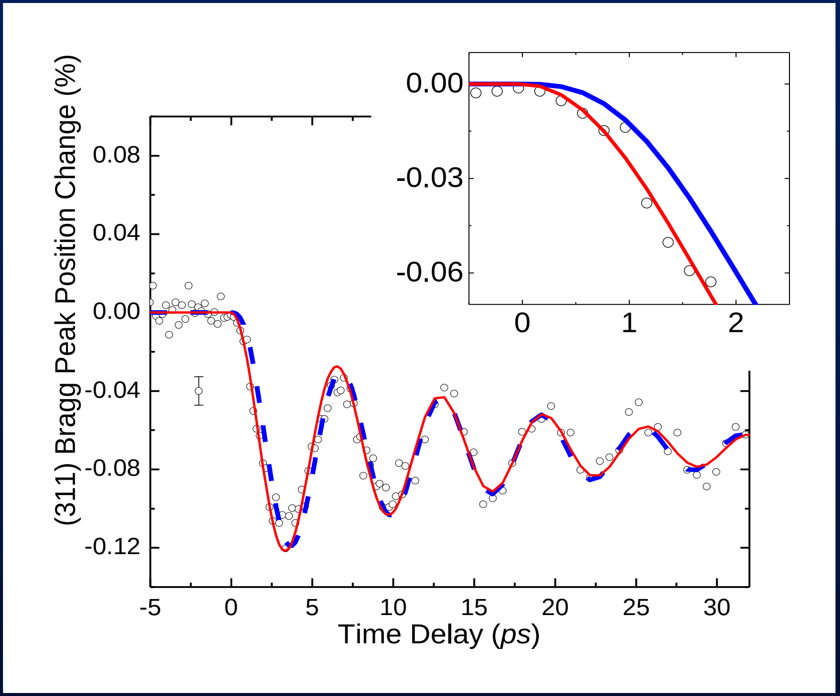
stress from lattice heating $$\sigma_l = \gamma_e C_l \delta T_l = \gamma_e E_{total}(1 - e^{-t/\tau_{e-ph}})$$

Energy conservation $$C_e \delta T_e + C_l \delta T_l = E_{total}$$

Total stress $$\sigma = \sigma_e + \sigma_l = -\gamma_e E_{total} e^{-t/\tau_{e-ph}} - \gamma_l E_{total}(1 - e^{-t/\tau_{e-ph}})$$

Measure the transient stress directly using UED

Harmonic Oscillator Approximation



$$\frac{d^2Q}{dt^2}+\beta\frac{dQ}{dt}+\omega^2x=\sigma$$

$$\sigma=\sigma_e+\sigma_l$$

$$=-\gamma_e E_{total}e^{-t/\tau_{e-ph}}-\gamma_l E_{total}(1-e^{-t/\tau_{e-ph}})$$

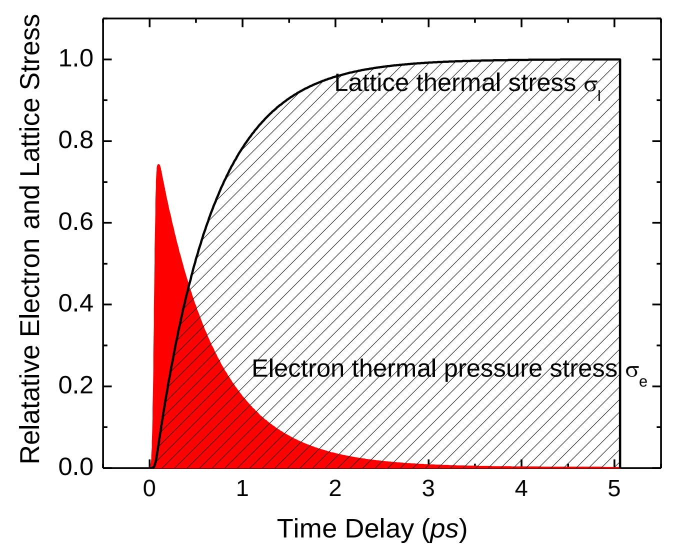
$\gamma_l = 2.16, \gamma_e = 1.6$

Damped harmonic oscillator gives a very good overall fitting

Fitting without σ_e creates a significant phase lag near timezero

H. Park, et al, PRB Rapid Comm. 72, 100301(2005)

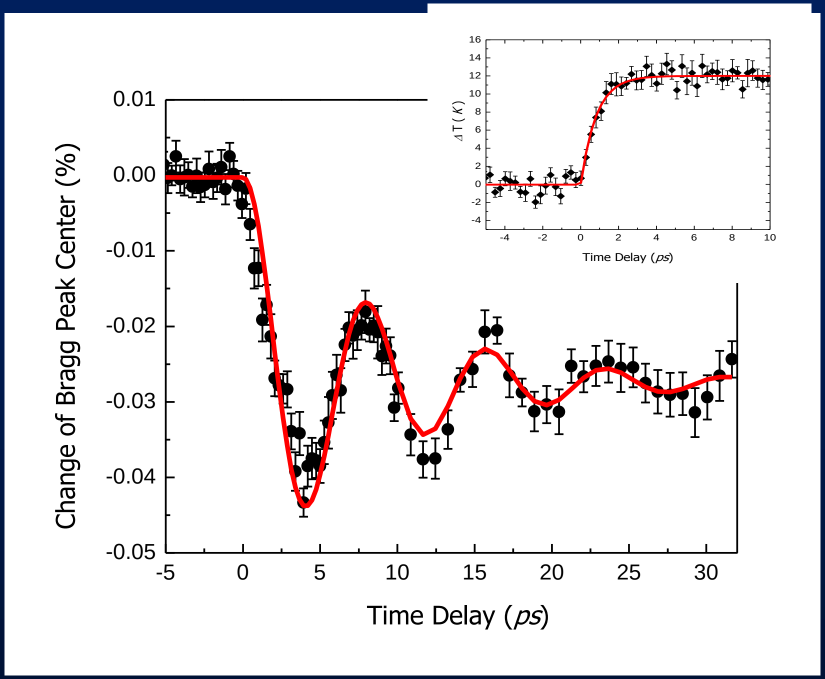
Dynamics of thermal expansion



- Both e$^-$ and lattice heating contribute, electronic contribution is significant at early times
- If $3\tau_{e\text{-}ph} \ll T/4$, lattice dominant

  $3\tau_{e\text{-}ph}$ ~ or > $T/4$, electrons contribute significantly

Measurement of γ_e in Ni in time domain



Coherent lattice motions

$$\frac{d^2Q}{dt^2}+2\beta\frac{dQ}{dt}+\omega_0^2Q=\sigma(t)$$

$$\sigma=A-Be^{-t/\tau_{e-ph}}$$

$$\frac{A}{B}=\frac{\gamma_l}{\gamma_l-\gamma_e}$$

Overcome the LT restrictions, can measure γ_e of magnetic materials

$\gamma_e = 1.4 \pm 0.3$ at T = 680 K > Curie point 630 K

X Wang, et.al., *Appl. Phys. Lett.* **92**, 121918 (2008)

S. Nie, X. Wang, H. Park, R. Clinite, J. Cao *Phys. Rev. Lett.* **96,** 025901 (2006)

γ_e of Ni in paramagnetic state

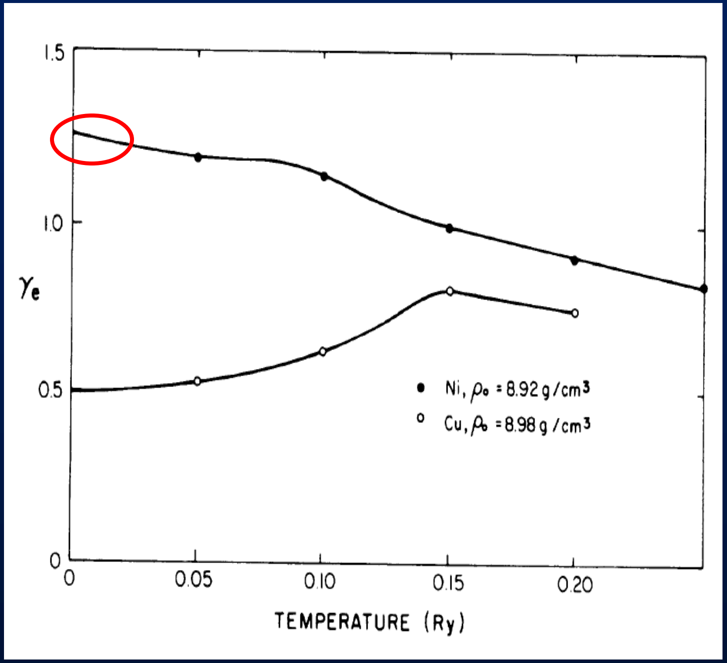
γ_e of paramagnetic state Ni has been calculated with DFT

- Finite temperature LMTO band-structure method and LSDA

- Local magnetic moment (*LM*) that persists in paramagnetic state Ni was neglected

- γ_e = 1.3

$$\alpha_e = \frac{\gamma_e}{3BV}(C_e + \frac{1}{4} I \frac{\partial < m^2 >}{\partial T})$$



Levy *et al. Phys. Rev. B* ***35****, 9474 (1987)*

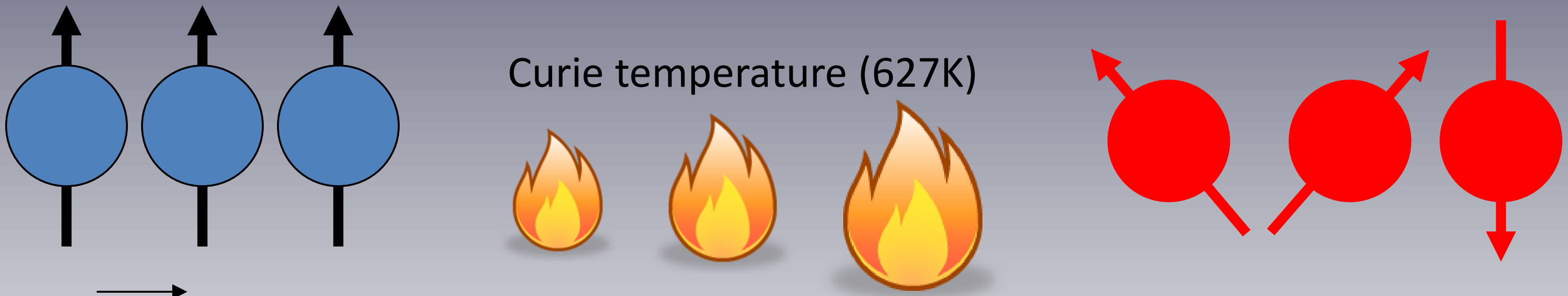
- Good agreement with FED measurement

LM does not play an important role in electronic thermal expansion

(Y. Kakehashi and J. H. Samson, *PRB* **34**, 1734 (1986))

Laser-induced Demagnetization in Nickel

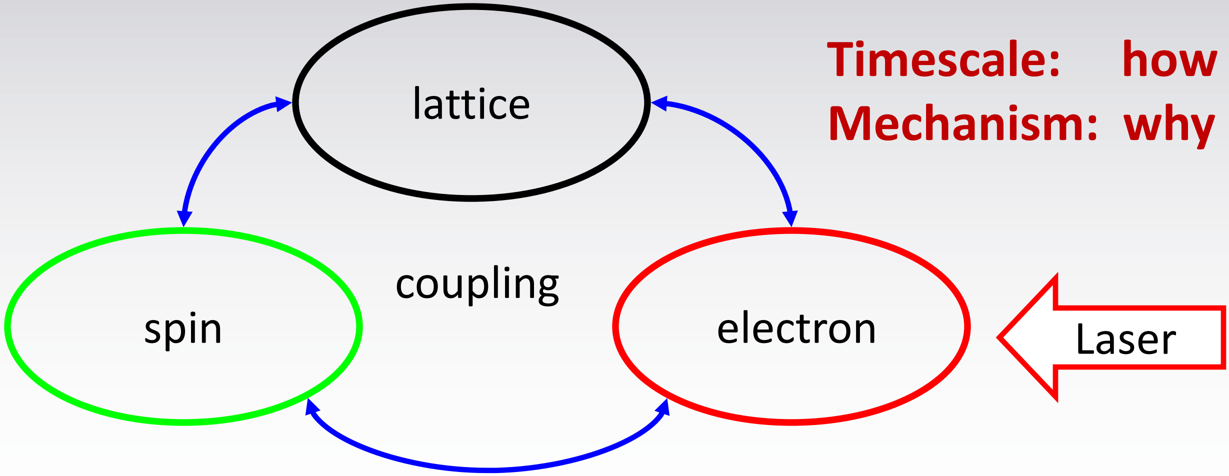
Demagnetization



Curie temperature (627K)

$\vec{M} \neq 0$ FM

$\vec{M} = 0$ PM



Timescale: how fast to read and write

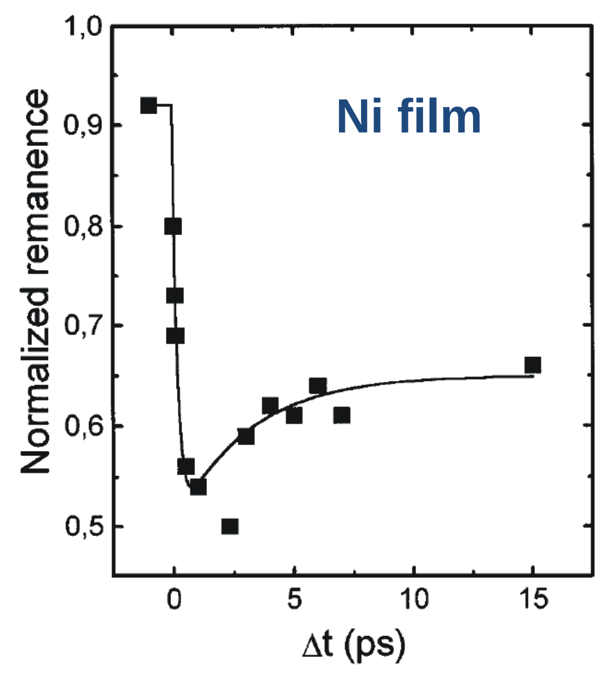
Mechanism: why so fast

ultrafast-heating

time-resolved

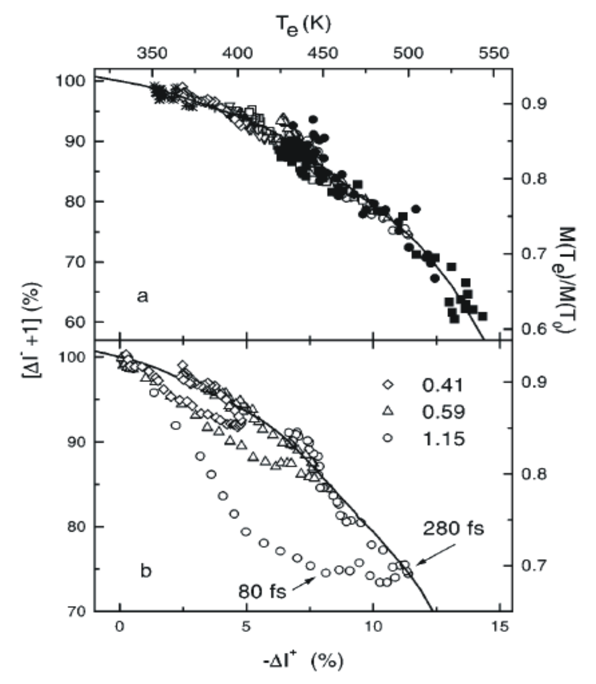
Ultrafast demagnetization: spin-lattice interaction?

- Time-resolved *MOKE*
- Demagnetization time $\tau_M < 1$ ps
- Spin temperature $T_s < T_e$



Beaurepaire *et al*.,
Phys. Rev. Lett. **76**, 4250 (1996)

- Time-resolved *MSHG*
- Demagnetization time $\tau_M < 280$ fs
- Spin temperature $T_s(t) \sim T_e(t)$



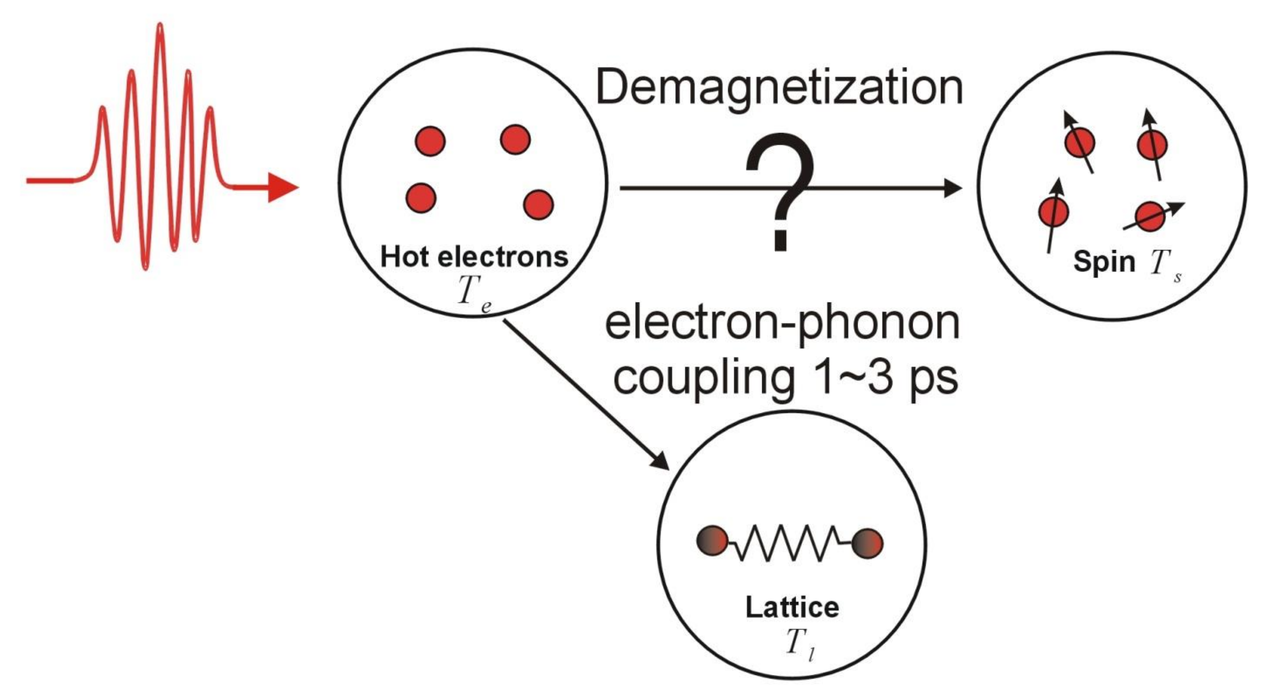
Hohlfeld *et al*.,
Phys. Rev. Lett. **78**, 4861 (1997)

Traditional spin-orbit coupling: ~ 100 ps, too slow

Probe ultrafast demagnetization with UED

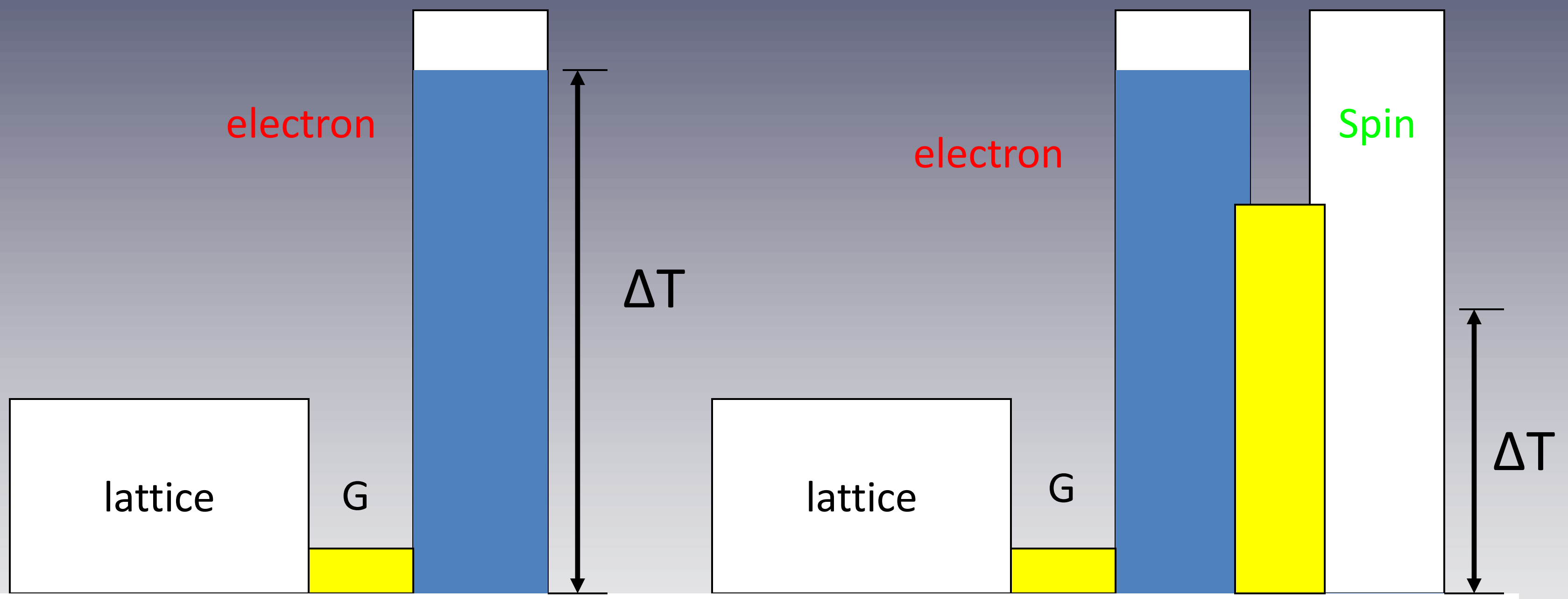
Ultrafast demagnetization ?

- Yes. $\Delta E_{\mathrm{e}} \rightarrow \Delta E_{\mathrm{s}}$ then, $\Delta E_{\mathrm{es}} \rightarrow \Delta E_L$ (two step)
- No. $\Delta E_{\mathrm{e}} \nrightarrow \Delta E_{\mathrm{s}}$ $\Delta E_{\mathrm{e}} \rightarrow \Delta E_L$ (one step)



Probe demagnetization dynamics by monitoring energy flow rate among sub-systems in real time

Energy Flow among Three Systems



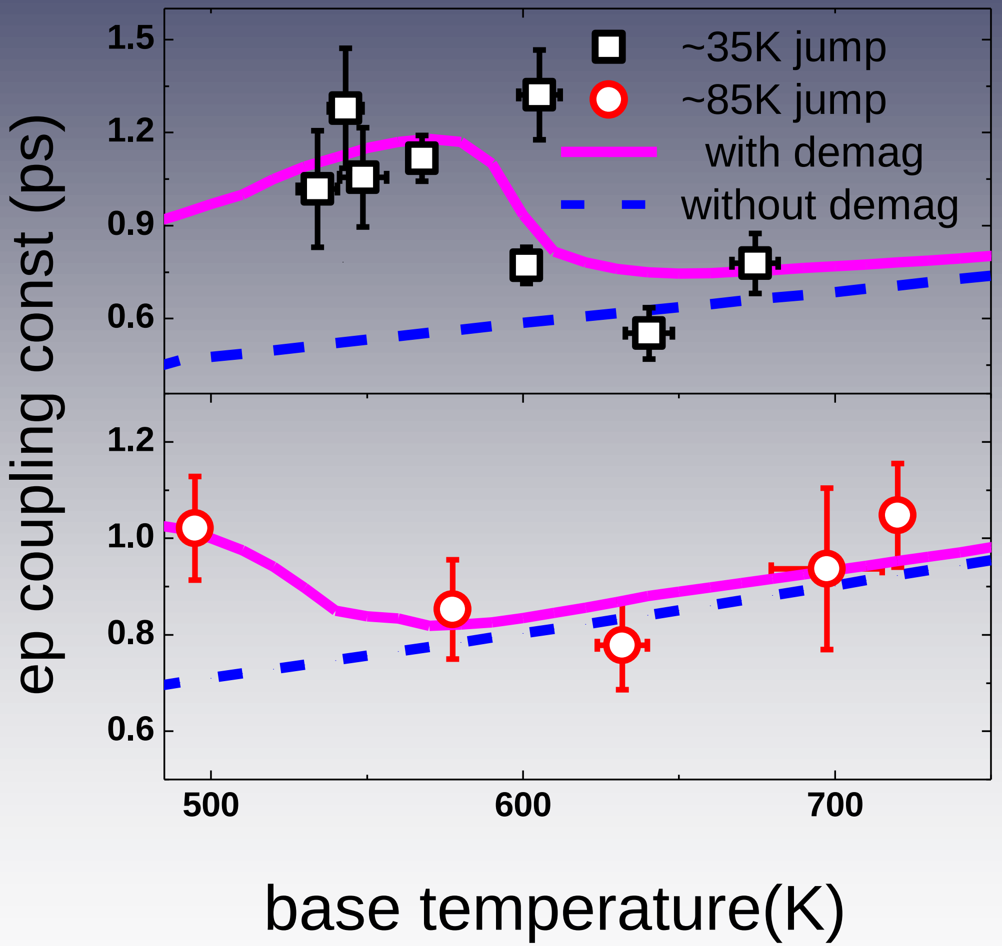
$T_e \neq T_l$ $T_e \neq T_s$

$T_e = T_s \neq T_l$

ultrafast demagnetization ↔ the lattice heating

slow down

$\tau_{e\text{-}ph}$ vs Sample Temperature



- e-ph coupling time curve resembles the heat capacity curve
- smear-out effect due to high excitation energy
- TTM simulation (assume electron kinetic energy and magnetic order can be characterized by one temperature)

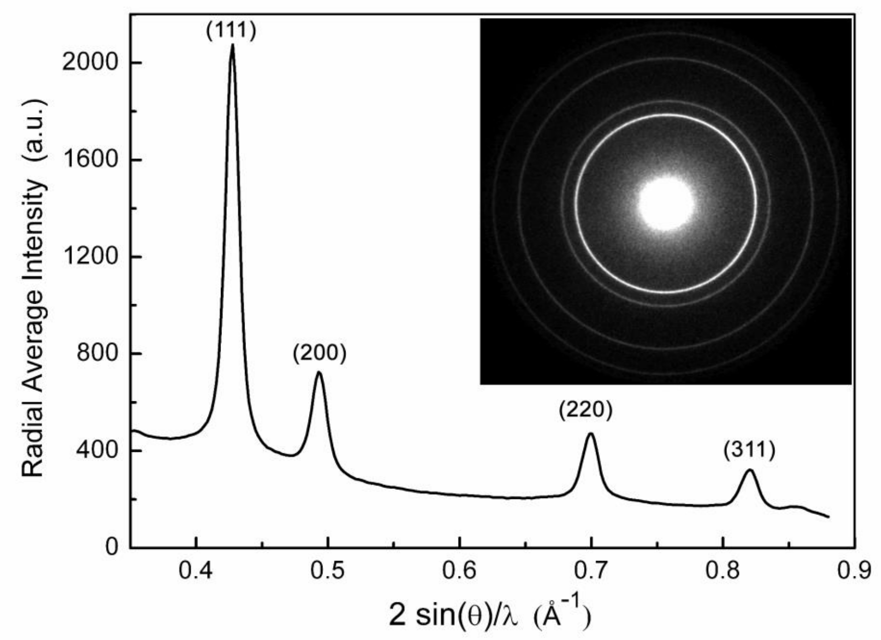
$$(C_e + C_m)\frac{\partial T_{em}}{\partial t} = -(G_{el} + G_{ml})(T_{em} - T_l)$$

$$C_l \frac{\partial T_l}{\partial t} = +(G_{el} + G_{ml})(T_{em} - T_l)$$

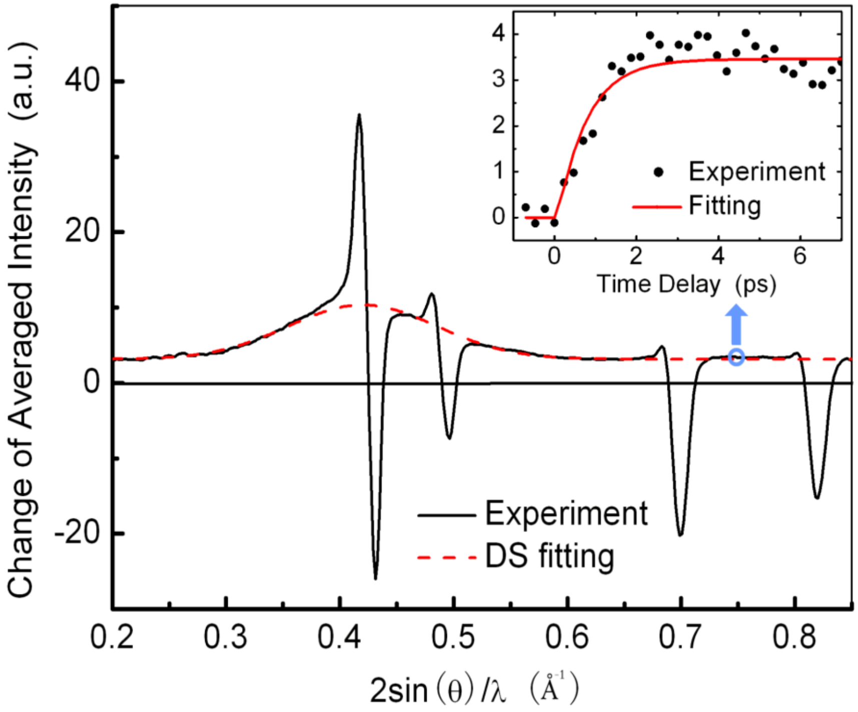
X. Wang, et.al., Phys. Rev. B 81, 220301(R) (2010)

The demagnetization is completed in less than 2 ps

Ultrafast Electron diffuse scattering (UEDS)



PF Zhu et al, APL, 103, 231914 (2013)



Bragg Peaks + diffuse scattering

$$I(S,t=20)-I(S,t<0)$$

DS: due to divergence from an ideal crystal, local (short ranged) correlations (defects and disorder)

diffuse ring ←→ pair correlation function

order-disorder transition, melting

Zhu, UED SJTU

Summary

- Under ultrafast heating condition, electronic thermal stress contributes significantly to the lattice thermal expansion
- A time domain method of measuring electronic Grüneisen parameter γ_e at or above room temperature
- Measure γ_e of Ni in paramagnetic state and local magnetic moment does not contribute significantly to electronic thermal expansion
- Optical-induced ultrafast demagnetization in Ni was confirmed from the UED

Acknowledgments

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Xuan Wang, Rick Clinite, Junjie Li

Martech group
Physics department machine shop

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