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: \( G \)

\[
\begin{align*}
C_e(T_e) \frac{\partial T_e}{\partial t} &= K \nabla^2 T_e - G(T_e - T_l) + P(x,t) \\
C_l \frac{\partial T_l}{\partial t} &= G(T_e - T_l)
\end{align*}
\]

e-ph thermalization: \( \sim 1-3 \) ps

Study the lattice response to the fs laser ultrafast heating
Ultrafast Heating of thin film and nano-particles

Thin Al films

\[ \Delta E_e \gg \Delta E_l \]


- the stress contains both electron contribution and lattice contribution.

Ag nano-particles


- 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
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/cm², 2 mm beam size
e-beam: 60 keV, <1000 e/pulse, ~400 fs, ~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

(a)  (b)  (c)

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 $T \sim 6.3$ ps, standing wave
- Maximum displacement at time zero
- Detection sensitivity: $\Delta r/r \sim 0.02\%$, < milli-angstrom

Displacive excitation of coherent phonon

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

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

\(A\): surface area
\(\sigma\): thermal stress

Maximum displacement at \(t=0\)
Thermal lattice motions ($T_l$) $\leftrightarrow$ thermal stress

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

20-nm Al film

$\Delta T$ (K) vs Time Delay (ps)

$\tau_{e-ph} = 600 \pm 100$ fs

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

H. Park, X. Wang, S. Nie, R. Clinite and J. Cao, PRB Rapid Comm. 72, 100301 (2005)
Transient Stress of ultrafast heating

\[ \sigma = \sigma_e + \sigma_l = \gamma_e \Delta E_e + \gamma_l \Delta E_l = \gamma_e C_e \delta T_e + \gamma_l C_l \delta T_l \]

\( \gamma_e \) and \( \gamma_l \) are electronic and lattice Grüneisen constants
\( C_e \) and \( C_l \) are heat capacities;
\( \Delta E_l \) and \( \Delta E_e \) subsystem thermal energy

stress from lattice heating

\[ \sigma_l = \gamma_e C_l \delta T_l = \gamma_e E_{total} \left(1 - e^{-t/\tau_{e-ph}}\right) \]

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

\( \tau_{e-ph} = 600 \pm 100 \) fs

Graph showing \( \Delta T \) vs. time delay with a peak at 600 fs.
Harmonic Oscillator Approximation

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

\[
\sigma = \sigma_e + \sigma_l
\]

\[
= -\gamma_e E_{total} e^{-\frac{t}{\tau_{e-ph}}} - \gamma_l E_{total} \left(1 - e^{-\frac{t}{\tau_{e-ph}}} \right)
\]

\[
\gamma_l = 2.16, \quad \gamma_e = 1.6
\]

Damped harmonic oscillator gives a very good overall fitting

Fitting without \( \sigma_e \) creates a significant phase lag near time zero

Dynamics of thermal expansion

- Both e⁻ and lattice heating contribute, electronic contribution is significant at early times.

- If $3\tau_{e-ph} << T/4$, lattice dominant.

- If $3\tau_{e-ph} ~ or > T/4$, electrons contribute significantly.
Measurement of $\gamma_e$ in Ni in time domain

Overcome the LT restrictions, can measure $\gamma_e$ of magnetic materials

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


\(\gamma_e\) of Ni in paramagnetic state

\(\gamma_e\) 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

\(\gamma_e = 1.3\)

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

- Good agreement with FED measurement

\(LM\) does not play an important role in electronic thermal expansion


Laser-induced Demagnetization in Nickel

Demagnetization

\[ M \neq 0 \quad \text{FM} \]

\[ M = 0 \quad \text{PM} \]

Curie temperature (627K)

Mechanism: why so fast

Timescale: how fast to read and write

Laser-induced demagnetization in nickel involves the interaction between electron, lattice, and spin coupling. The Curie temperature (627K) indicates the transition from ferromagnetic (FM) to paramagnetic (PM) states. The interaction is ultrafast-heating with time-resolved effects.
Ultrafast demagnetization: spin-lattice interaction?

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

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


Traditional spin-orbit coupling: $\sim 100$ ps, too slow
Probe ultrafast demagnetization with UED

Ultrafast demagnetization?

- Yes. $\Delta E_e \rightarrow \Delta E_s$ then, $\Delta E_{es} \rightarrow \Delta E_L$ (two step)
- No. $\Delta E_e \rightarrow \Delta E_s \quad \Delta E_e \rightarrow \Delta E_L$ (one step)

Probe demagnetization dynamics by monitoring energy flow rate among sub-systems in real time
Energy Flow among Three Systems

- $T_e \neq T_l$
- $T_e \neq T_s$
- $T_e = T_s \neq T_l$

Electron.

Ultrafast demagnetization

Slow down

The lattice heating
\( \tau_{e-ph} \) 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)

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

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


The demagnetization is completed in less than 2 ps
Ultrafast Electron diffuse scattering (UEDS)

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 $\leftrightarrow$ pair correlation function

order-disorder transition, melting

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

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 $\gamma_e$ at or above room temperature.

- Measure $\gamma_e$ 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.
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