Lattice dynamics and spin-phonon interaction in thin films and nanostructures

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Nuclear resonance scattering
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Inelastic X-ray scattering
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UHV-Analysis Laboratory at ANKA
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Outline

- Motivation to study phonons in nanoscale materials
- Methods for probing lattice dynamics at the nanoscale
- Lattice dynamics in thin films and nanostructures:
  - phonon dispersions and phonon density of states
  - spin-orbit coupling, $4f$ electron correlations
  - electron-phonon and spin-phonon coupling
- Conclusions and outlook
Motivation to study phonons in nanoscale materials

The fundamental understanding of the atomic vibrations in low dimensional systems is essential for the elucidation of phenomena such as:

- superconductivity
- thermoelectricity
- propagation of sound and heat

and for designing new devices like:

- thermal logic gates, thermal memory
  \( PRL \ 99, \ 177208 \ (2007); \ PRL \ 101, \ 267203 \ (2008) \)

- phononic waveguides, resonators and switches
  \( PCCP \ 16, \ 23355 \ (2014) \)
Methods for probing lattice dynamics at the nanoscale

- Inelastic scattering of particles

HRHAS:
High Resolution He Atom Scattering


HREELS:
High Resolution Electron Energy Loss Spectroscopy

Methods for probing lattice dynamics at the nanoscale

- Inelastic scattering of light

Surface Brillouin Scattering


Surface Enhanced IR Absorption


Surface Enhanced Raman Scattering

Methods for probing lattice dynamics at the nanoscale

- Inelastic scattering of light

Grazing Incidence Inelastic X-ray Scattering


(In Situ) Nuclear Inelastic Scattering

Lattice dynamics in thin films and nanostructures

- Lattice dynamics of Nd: spin-orbit coupling and 4f electron correlations
  - Represents the light lanthanides
  - More delocalized 4f electrons compared to the heavy lanthanides
  - Important material for building strong permanent magnets
  - Unknown lattice dynamics
Lattice dynamics in thin films and nanostructures

- Lattice dynamics of Nd: spin-orbit coupling and 4f electron correlations

*Ab initio* calculated lattice dynamics of Nd: VASP + PHONON by P. Piekarz, K. Parlinski (Krakow)

![Graph of lattice dynamics](image)

**TABLE I.** Lattice constants \((a, c)\), volume per one atom \((V)\), and thermoelastic properties (bulk modulus \(B\), derivative of the bulk modulus \(B'\), elastic constants \(c_{xy}\), and lattice specific heat \(C_V\) at 300 K) of Nd.

<table>
<thead>
<tr>
<th>Property</th>
<th>GGA₀</th>
<th>GGA</th>
<th>GGA+U (SOC)</th>
<th>Expt.</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a) (Å)</td>
<td>3.690</td>
<td>3.528</td>
<td>3.669 (3.670)</td>
<td>3.658&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>(c) (Å)</td>
<td>11.870</td>
<td>11.277</td>
<td>11.804 (11.824)</td>
<td>11.797&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>(V) (Å³/atom)</td>
<td>34.997</td>
<td>30.389</td>
<td>34.398 (34.470)</td>
<td>34.18&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>(B) (GPa)</td>
<td>34.7</td>
<td>18.6</td>
<td>31.4 (32.15)</td>
<td>31.8&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>(B') (GPa)</td>
<td>3.09</td>
<td>2.41</td>
<td>3.05 (3.04)</td>
<td>2.9&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>(c_{11}) (GPa)</td>
<td>59.9</td>
<td>31.4</td>
<td>55.2</td>
<td>58.78&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>(c_{33}) (GPa)</td>
<td>72.2</td>
<td>39.1</td>
<td>65.1</td>
<td>65.13&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>(c_{12}) (GPa)</td>
<td>29.8</td>
<td>14.9</td>
<td>27.9</td>
<td>24.58&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>(c_{13}) (GPa)</td>
<td>16.5</td>
<td>11.1</td>
<td>14.2</td>
<td>16.20&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>(c_{44}) (GPa)</td>
<td>18.8</td>
<td>6.3</td>
<td>18.5</td>
<td>16.20&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>(C_V) (J/mol K)</td>
<td>24.67</td>
<td>24.75</td>
<td>24.69</td>
<td>24.68&lt;sup&gt;d&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

Lattice dynamics in thin films and nanostructures

- Lattice dynamics of Nd: spin-orbit coupling and 4f electron correlations

IXS experiment at ID28, ESRF (A. Bosak):
- $E = 17.8$ keV
- FWHM = 3 meV

- 4f el. correlations have an impact on the lattice dynamics
- They can be correctly described by the GGA+U formalism
- The spin-orbit coupling has a negligible influence on the lattice parameters

Lattice dynamics in thin films and nanostructures

- Lattice dynamics of EuO: evidence for giant spin-phonon coupling
  - Semiconducting ferromagnet & model system for Heisenberg ferromagnets
  - Exceptionally high magnitudes of Faraday and Kerr effects
  - Insulator-to-metal transition
  - Proposed as spin injector for future spintronic devices

Curie temperature \( T_c = 69 \text{ K} \)
Band gap = 1.1 eV
Electronic config: \([\text{Xe}] 4f^7 6s^2\)

\[ E_g \]
\[ 0.3 \text{ eV} \]

A. Schmehl et al., Nat. Mater. 6, 882 (2007)
J. H. Greiner et al., Appl. Phys. Lett. 9, 27 (1966)
Lattice dynamics in thin films and nanostructures

- Lattice dynamics of EuO: evidence for giant spin-phonon coupling

100 nm EuO(001) on YSZ(001) using Reactive Molecular Beam Epitaxy

RHEED along EuO(110)  X-ray photoelectron spectroscopy

![RHEED image]

- Counts (a.u.)
  - Eu 4f
  - Eu 2+ O 2s
  - Eu 5p

Binding energy (eV)

X-ray diffraction

![X-ray diffraction spectrum]

10 nm of Nb
100 nm of EuO(001)
YSZ(001)

Lattice parameters

- $a_{\text{EuO}} = 5.142 \text{ Å}$
- $a_{\text{YSZ}} = 5.144 \text{ Å}$

Lattice dynamics in thin films and nanostructures

- Lattice dynamics of EuO: evidence for giant spin-phonon coupling

IXS scans for the LA phonons

- T_C = 69 K

scattering geometry
Lattice dynamics in thin films and nanostructures

- Lattice dynamics of EuO: evidence for giant spin-phonon coupling

IXS scans for the TA phonons

![Graph showing phonon energy vs. temperature for different wavevectors](image)

- $T_C = 69$ K

Scattering geometry
Lattice dynamics in thin films and nanostructures

- Lattice dynamics of EuO: evidence for giant spin-phonon coupling

\[ \Gamma - X \]

\[ \Gamma - K - X \]

\[ \Gamma - K - \Lambda \]

\[ (0,q,q) \]

\[ (0,0,q) \]

\[ (0,q,0) \]

\[ T_C = 69 \text{ K} \]

Lattice dynamics in thin films and nanostructures

- Lattice dynamics of EuO: evidence for giant spin-phonon coupling

Eu-partial phonon DOS from NIS experiment

FWHM of the peaks in the DOS

FWHM of the TA along Γ-X from the IXS exp.

Lattice dynamics in thin films and nanostructures

- Lattice dynamics of EuO: evidence for giant spin-phonon coupling

\[ \Delta \text{FWHM} \approx \hbar \omega \left( \frac{zJ^2}{K} \right) \chi_s''(\hbar \omega) \]

For \( \Delta \text{FWHM} = 8 \text{ meV} \),
spin-phonon coupling constant, \( \alpha \approx 10! \)

\( \omega \): phonon energy; \( J \): exchange energy, \( K \): force constant;
\( z \): coordination number; \( X \): spectral density of spin waves

\[ C. \text{Ulrich et al.}, \text{Phys. Rev. Lett. 115, 156403 (2015)} \]

<table>
<thead>
<tr>
<th>Material</th>
<th>( \alpha )</th>
</tr>
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<tbody>
<tr>
<td>CuGeO(_3)</td>
<td>5.9</td>
</tr>
<tr>
<td>YBaCuO</td>
<td>10.4</td>
</tr>
<tr>
<td>NaOsO(_3)</td>
<td>( \approx 11.8 )</td>
</tr>
</tbody>
</table>

\[ R. \text{Werner et al.}, \text{PRB 59, 14356 (1999)} \]

\[ J. \text{P. Carbotte et al.}, \text{Nature 401, 354 (1999)} \]

\[ S. \text{Calder et al.}, \text{Nat. Commun. 6, (2015)} \]

\[ R. \text{Pradip et al.}, \text{Phys. Rev. Lett. 116, 185501 (2016)} \]
Conclusions and outlook

- Reaching a comprehensive understanding of the lattice dynamics modifications by nanostructuring is a challenge.

- New experimental methods are clearly needed for mapping phonon dispersions of nanostructures in particular of ultrathin buried layers (nanoscale interfaces):
  - electron correlations, superconductivity
  - electron-phonon, spin-phonon interactions,
  - thermal conductivity etc.
    → phonon nanoengineering

- The X-ray Echo Spectroscopy has the potential to provide deep insights into the lattice dynamics of nanoscale materials.

Thank you for the attention!