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

Svetoslav Stankov

Institute for Photon Science and Synchrotron Radiation, Karlsruhe Institute of Technology, Germany

Acknowledgement

NANODYNAMICS
A Helmholtz-University Young Investigators Group at ANKA

KIT
Karlsruhe Institute of Technology

First-principles theory

P. Piekarz and *K. Parlinski*
Institute of Nuclear Physics,
Polish Academy of Sciences, Krakow, Poland

Nuclear resonance scattering

D.G. Merkel, *A.I. Chumakov* and *R. Rüffer*
ID18 of the ESRF, Grenoble, France

Inelastic X-ray scattering

A. Bosak and *M. Krisch*
ID28 of the ESRF, Grenoble, France

UHV-Analysis Laboratory at ANKA

B. Krause, A. Weißhardt, H.H. Gräfe

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HELMHOLTZ
GEMEINSCHAFT

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)*

Svetoslav.Stankov@kit.edu

Methods for probing lattice dynamics at the nanoscale

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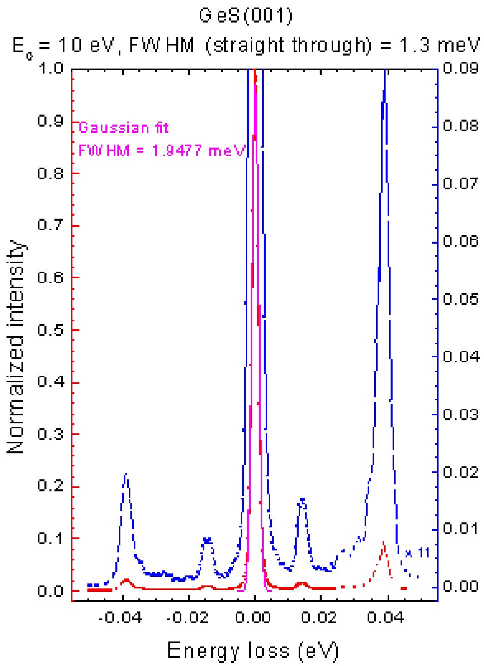
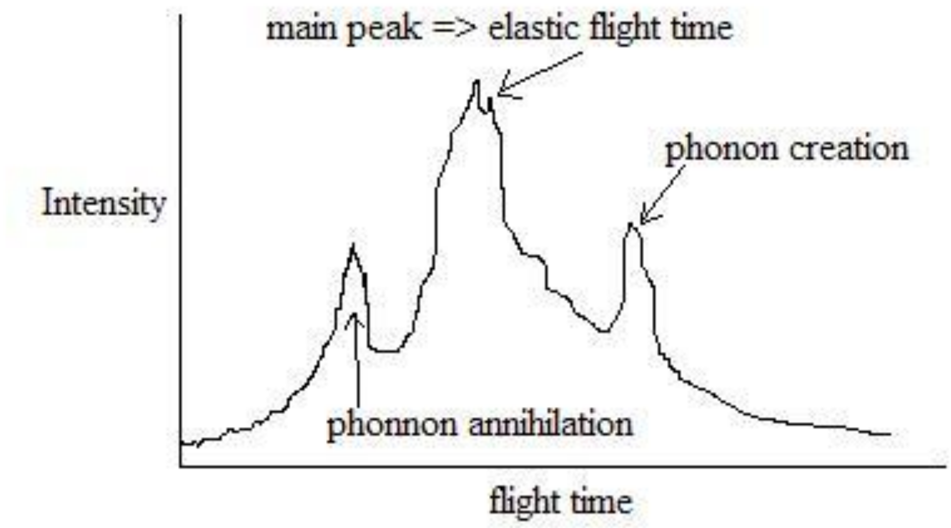
- Inelastic scattering of particles

HRHAS:
High Resolution He Atom Scattering

*A. P. Graham, Surf. Sci. Rep. **49**, 115 (2003)*





HREELS:
High Resolution Electron Energy Loss Spectroscopy

H. Ibach and D. L. Mills, Electron Energy Loss Spectroscopy and Surface Vibrations (Academic, New York,1982)

Methods for probing lattice dynamics at the nanoscale

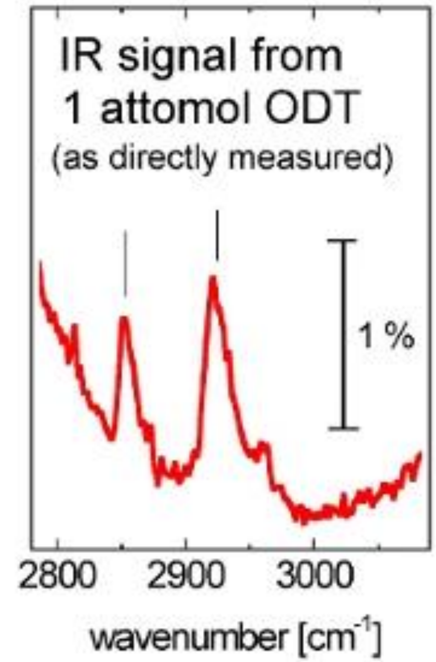
- Inelastic scattering of light

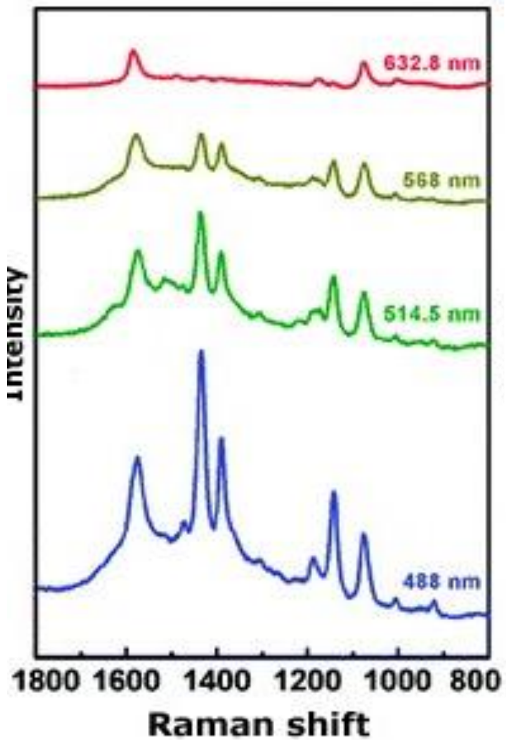
Surface Brillouin Scattering

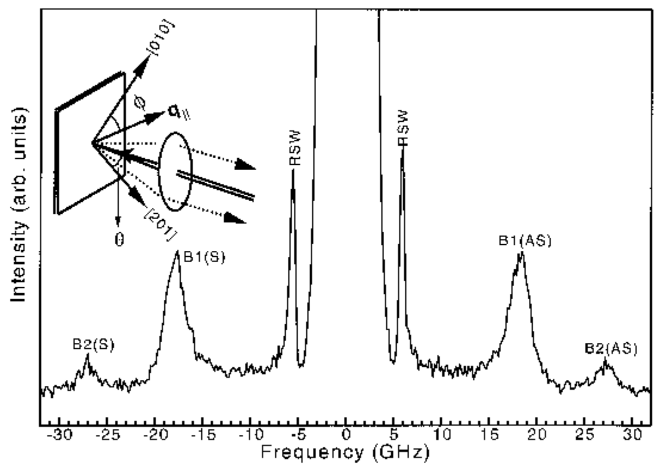
P. Murugavel et al., J. Phys.: Condens. Matter ***12*** *(2000)*

Surface Enhanced IR Absorption

F. Neubrech et al.,
Phys. Rev. Lett. , ***101*** *157403, (2008)*





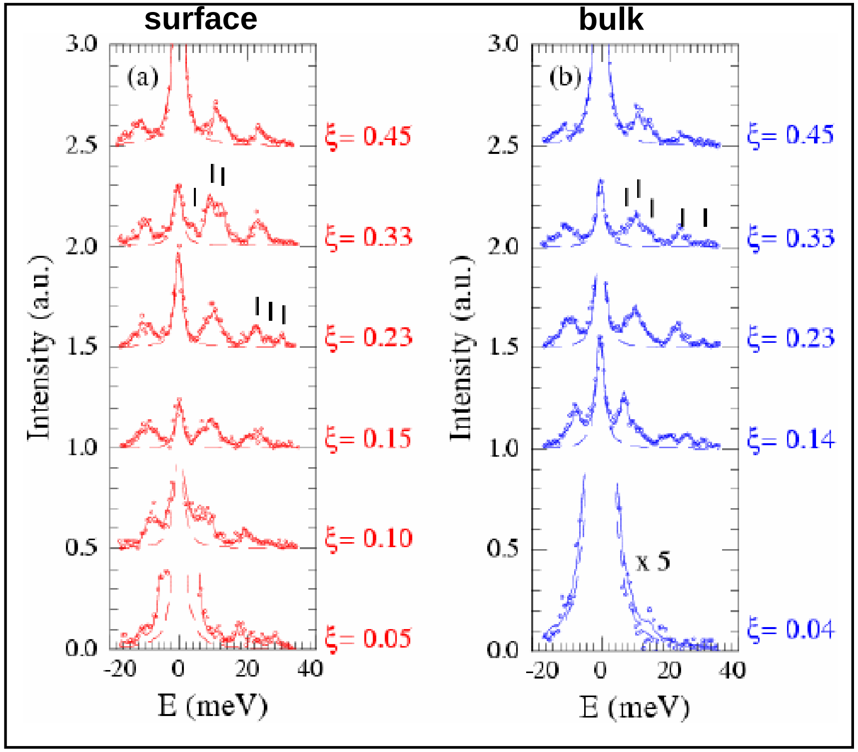


Surface Enhanced Raman Scattering

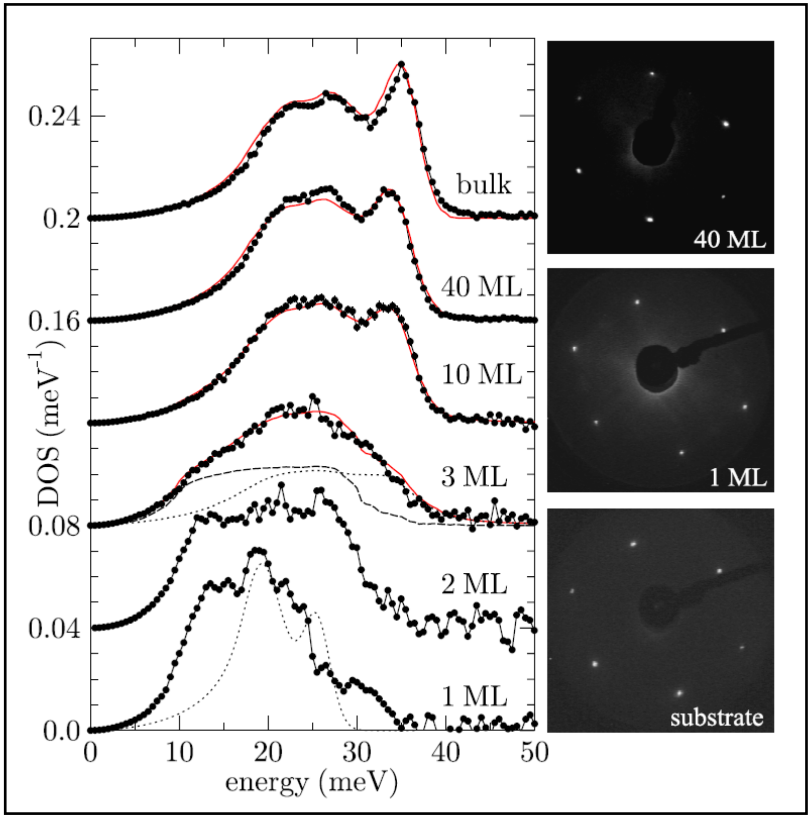
K. Kneipp, M. Moskovits, H. Kneipp (Edt.) Surface-Enhanced Raman Scattering: Physics and Applications, Springer (2006)

Methods for probing lattice dynamics at the nanoscale

- Inelastic scattering of light



Grazing Incidence Inelastic X-ray Scattering



(*In Situ*) Nuclear Inelastic Scattering

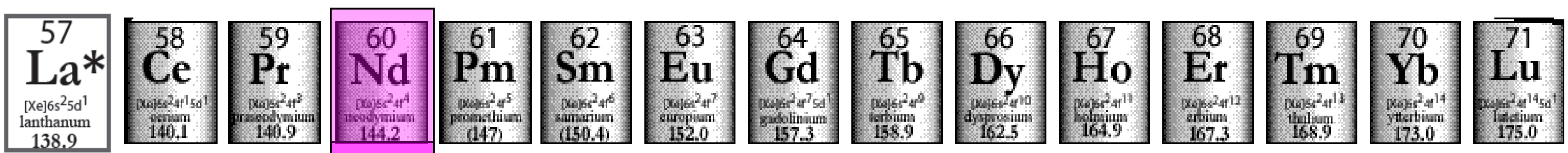
B. Murphy, et. al., Phys. Rev. Lett. ***95****, 256104* (2005)

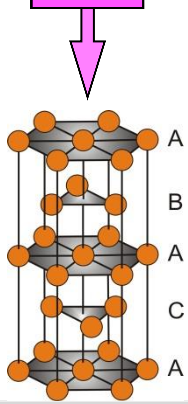
S. Stankov et al., Phys. Rev. Lett. ***99****, 185501 (2007)*

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





DHCP

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)

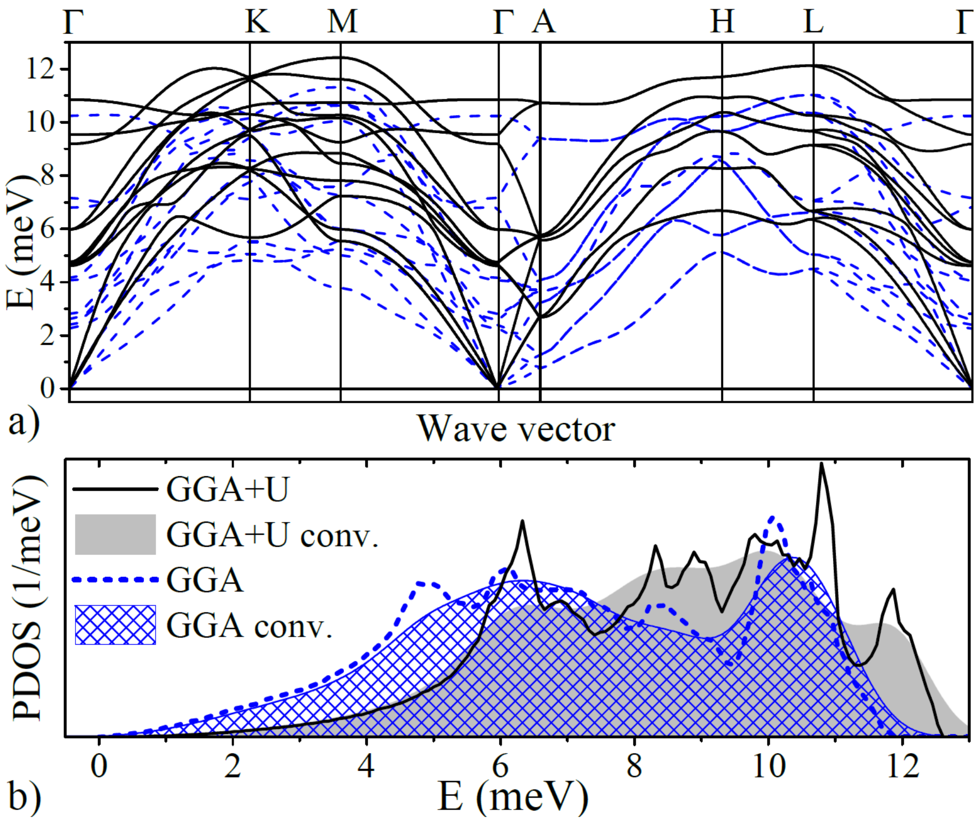


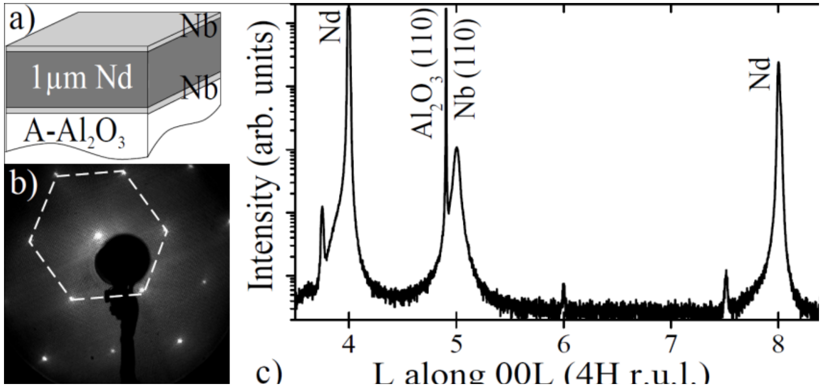
TABLE I. Lattice constants (a, c), volume per one atom (V), and thermoelastic properties (bulk modulus B, derivative of the bulk modulus B', elastic constants c_{xy}, and lattice specific heat C_V at 300 K) of Nd.

Property	GGA_0	GGA	GGA+U(SOC)	Expt.
a (Å)	3.690	3.528	3.669 (3.670)	3.658[a]
c (Å)	11.870	11.277	11.804 (11.824)	11.797[a]
V (Å^3/atom)	34.997	30.389	34.398 (34.470)	34.18[a]
B (GPa)	34.7	18.6	31.4 (32.15)	31.8[a]
B' (GPa)	3.09	2.41	3.05 (3.04)	2.9[b]
c_{11} (GPa)	59.9	31.4	55.2	58.78[c]
c_{33} (GPa)	72.2	39.1	65.1	65.13[c]
c_{12} (GPa)	29.8	14.9	27.9	24.58[c]
c_{13} (GPa)	16.5	11.1	14.2	16.20[c]
c_{44} (GPa)	18.8	6.3	18.5	16.20[c]
C_V (J/mol K)	24.67	24.75	24.69	24.68[d]

*O. Waller et al., Phys. Rev. B **94**, 014303 (2016)*

Lattice dynamics in thin films and nanostructures

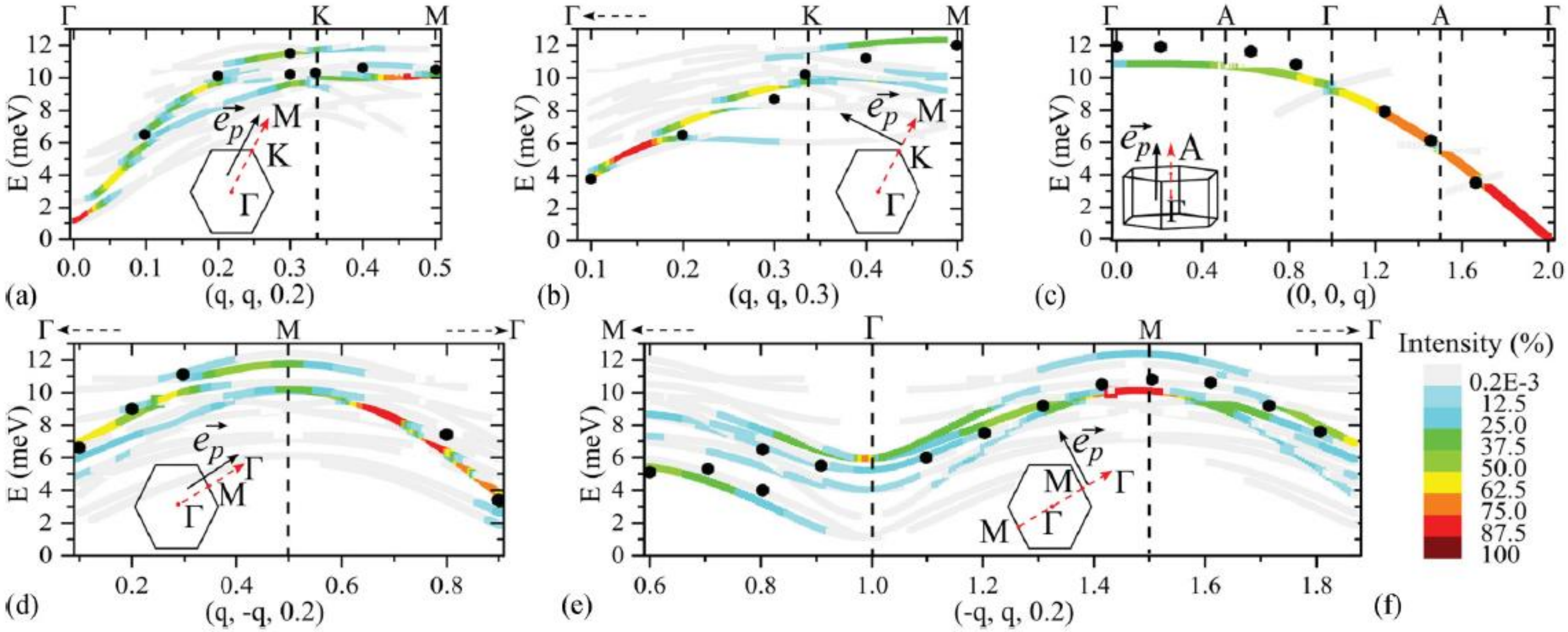
- Lattice dynamics of Nd: spin-orbit coupling and *4f* electron correlations



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

O. Waller et al., Phys. Rev. B ***94****, 014303 (2016)*

Lattice dynamics in thin films and nanostructures

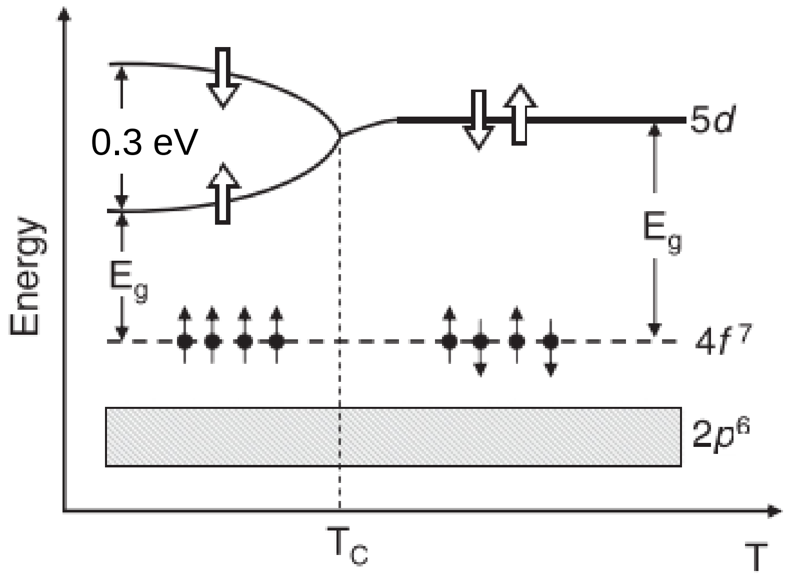
- Lattice dynamics of EuO: evidence for giant spin-phonon coupling

- 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 K

Band gap =1.1 eV

Electronic config: [Xe] $4f^7$ $6s^2$



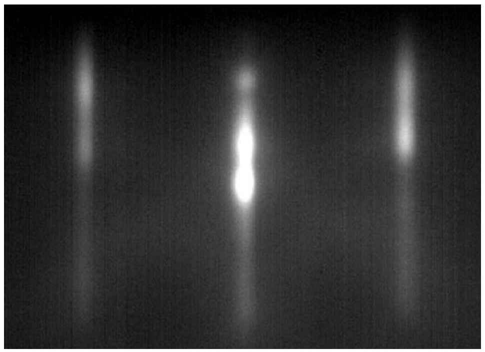
*A. Schmehl et al., Nat. Mater. **6**, 882 (2007)*
*K. Y. Ahn et al., J. Appl. Phys. **39**, 5061 (1968)*
*J. H. Greiner et al., Appl. Phys. Lett. **9**, 27 (1966)*
*Y. Shapira et al., Phys. Rev. B **8**, 2316 (1973)*
*T. S. Santos et al., Phys. Rev. Lett. **101**, 147201 (2008)*

Lattice dynamics in thin films and nanostructures

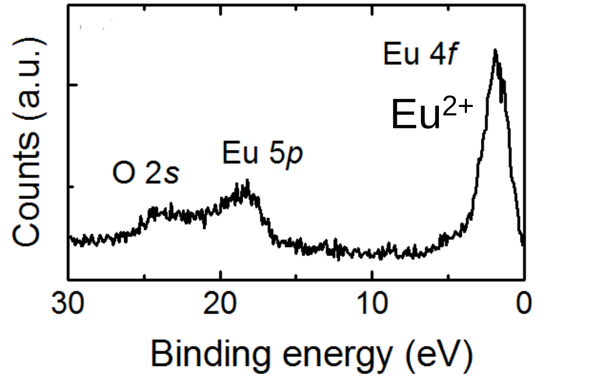
- Lattice dynamics of EuO: evidence for giant spin-phonon coupling

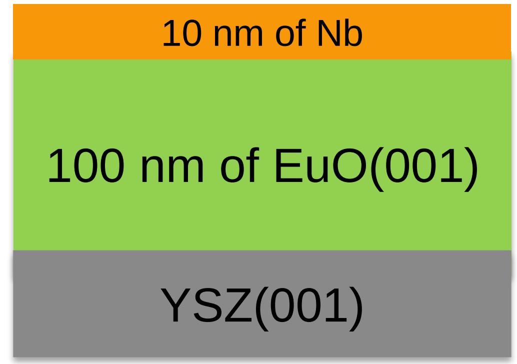
100 nm EuO(001) on YSZ(001) using Reactive Molecular Beam Epitaxy

RHEED along EuO(110)

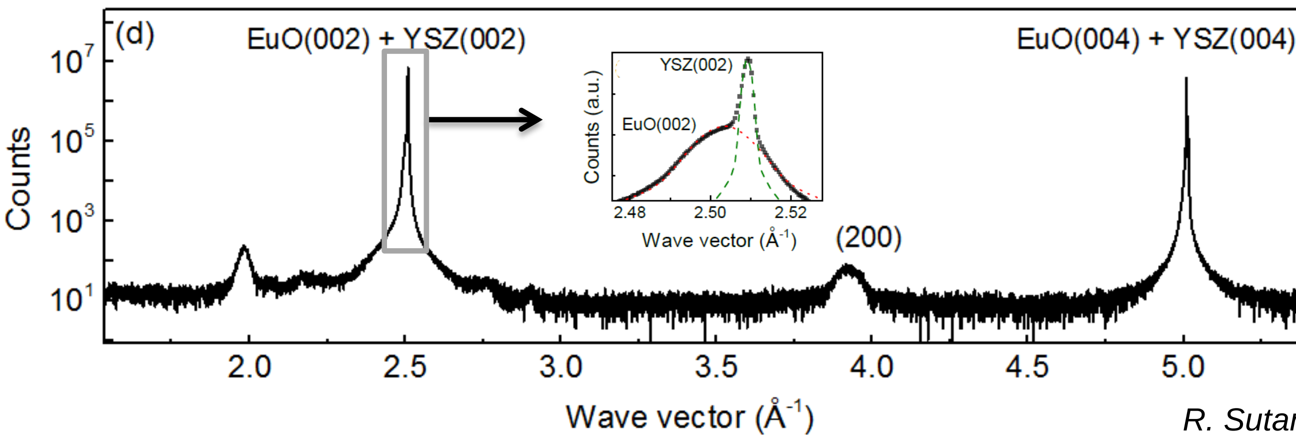


X-ray photoelectron spectroscopy





X-ray diffraction



Lattice parameters

a_EuO = 5.142 Å

a_YSZ = 5.144 Å

*R. Sutarto et al., Phys. Rev. B **79**, 205318 (2009)*

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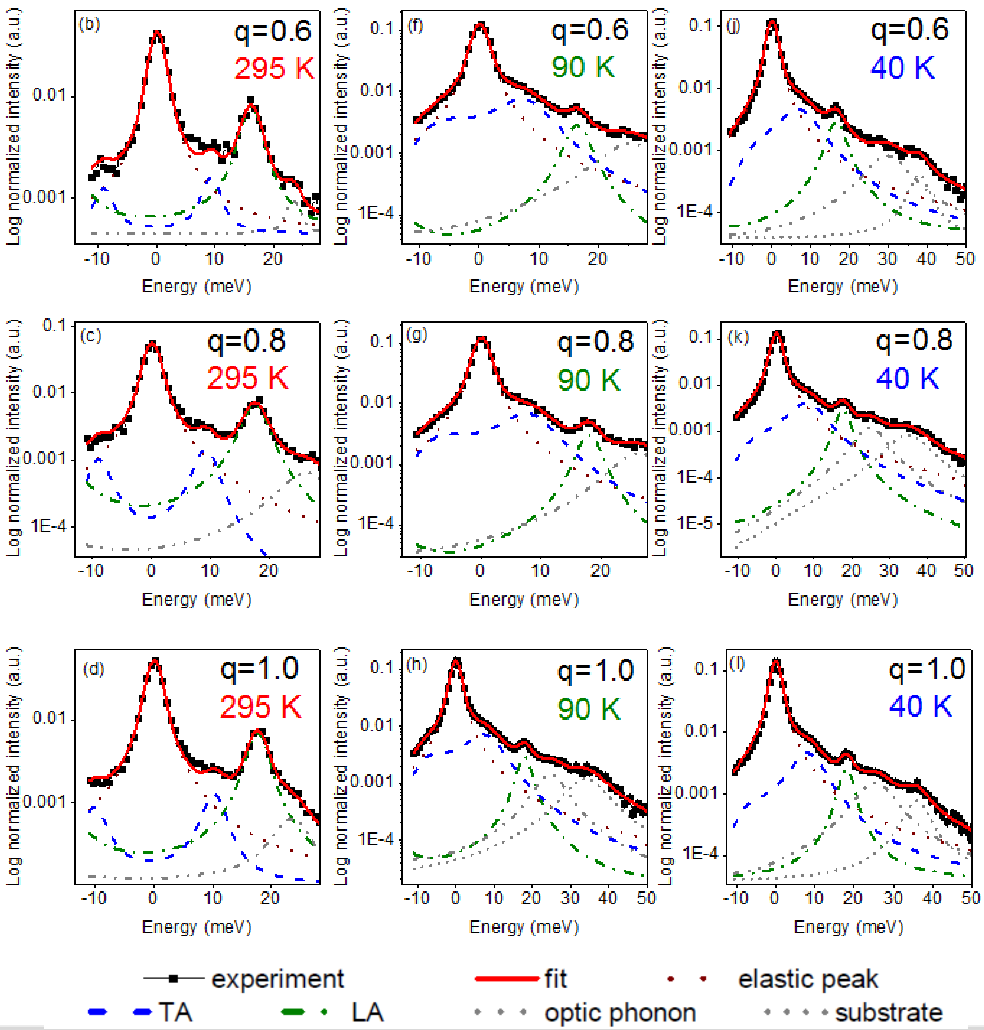
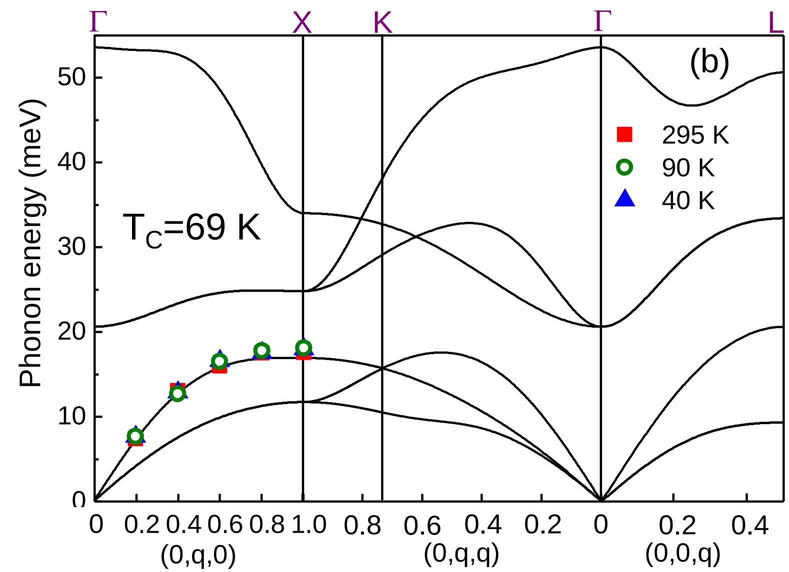
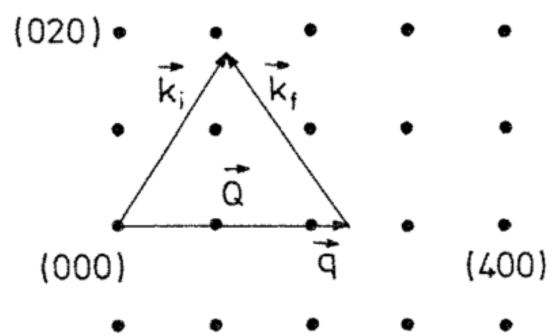
IXS scans for the LA phonons







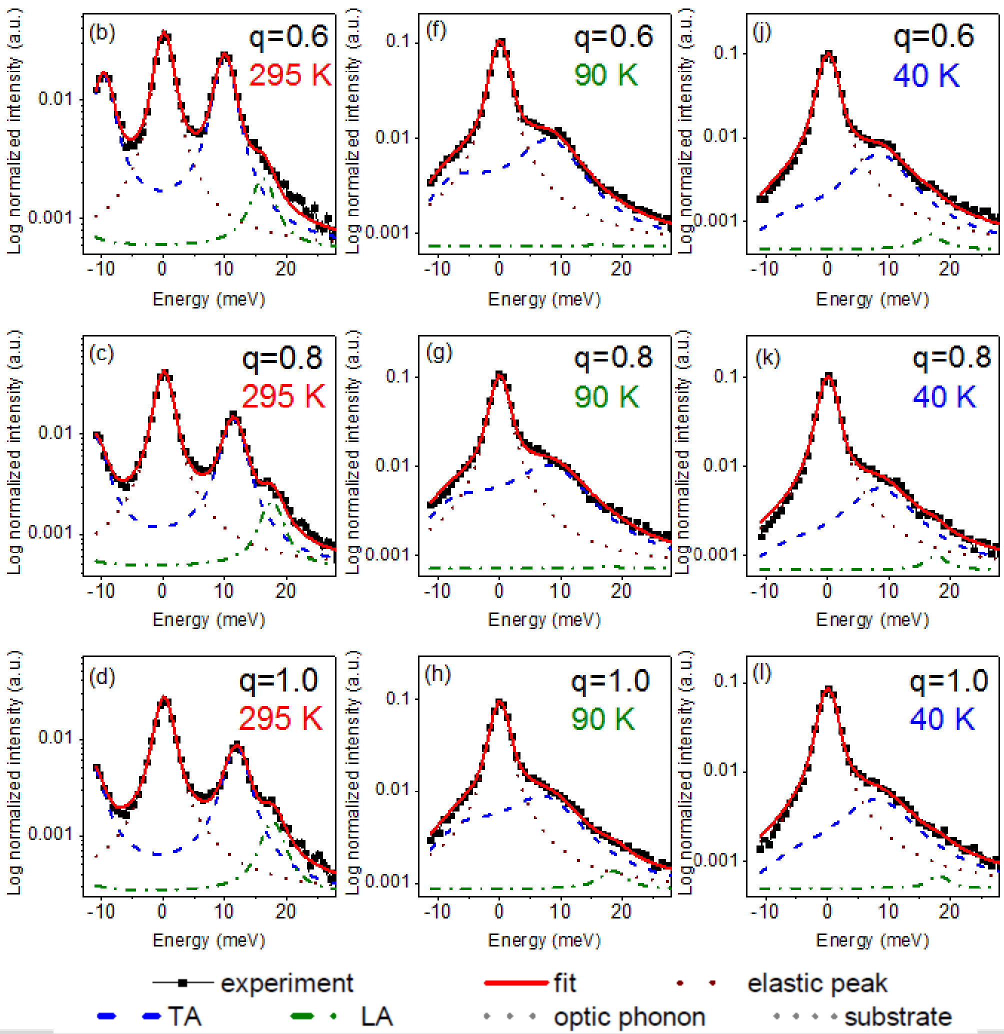
scattering geometry

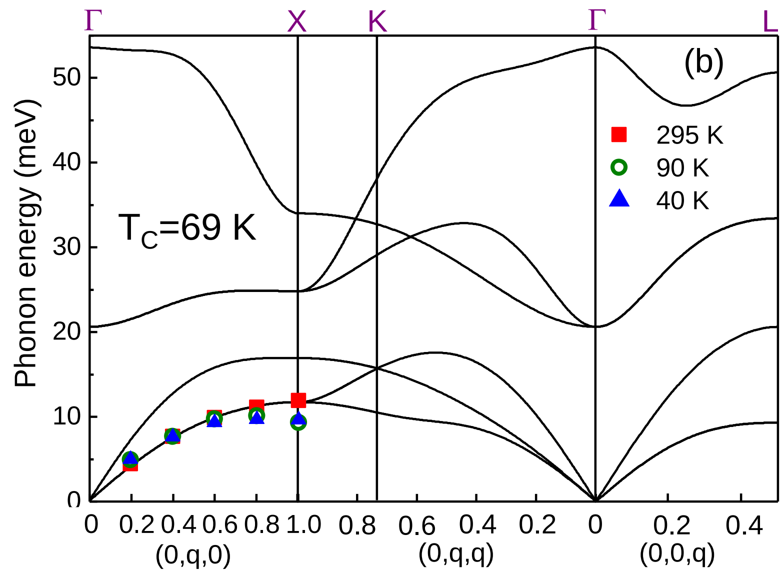
Svetoslav.Stankov@kit.edu

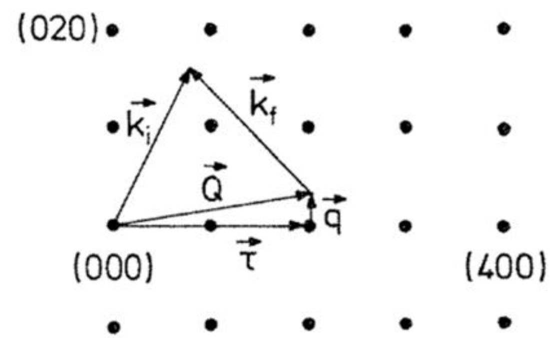
Lattice dynamics in thin films and nanostructures

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

IXS scans for the TA phonons







scattering geometry

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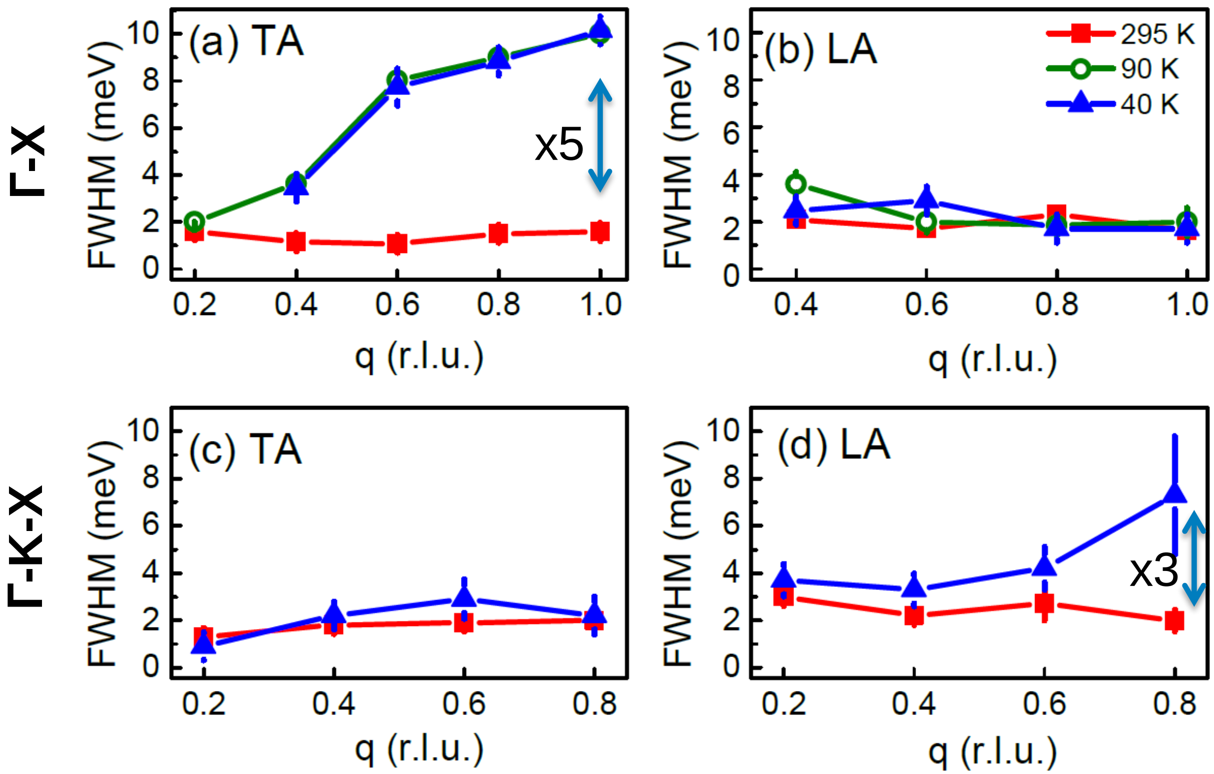

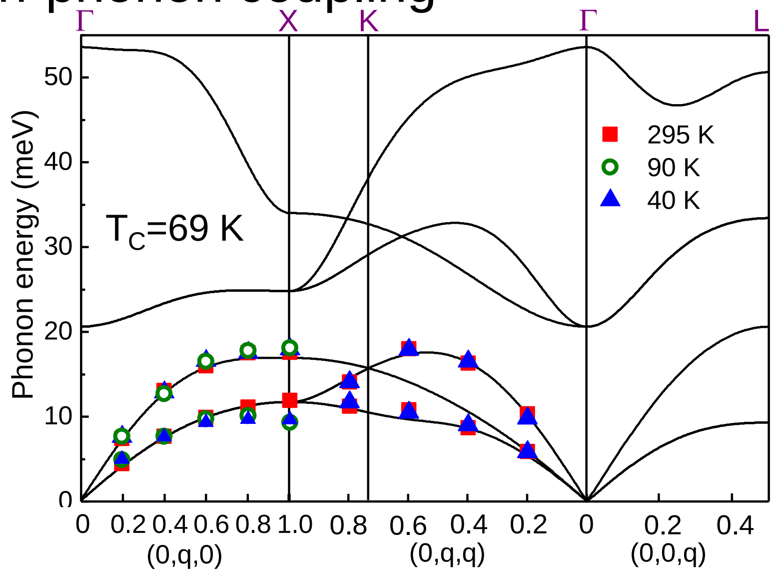

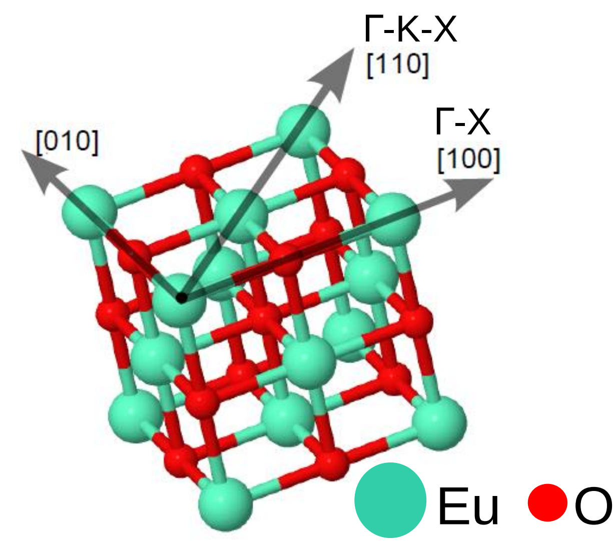

R. Pradip et al., Phys. Rev. Lett. ***116****, 185501 (2016)*

Lattice dynamics in thin films and nanostructures

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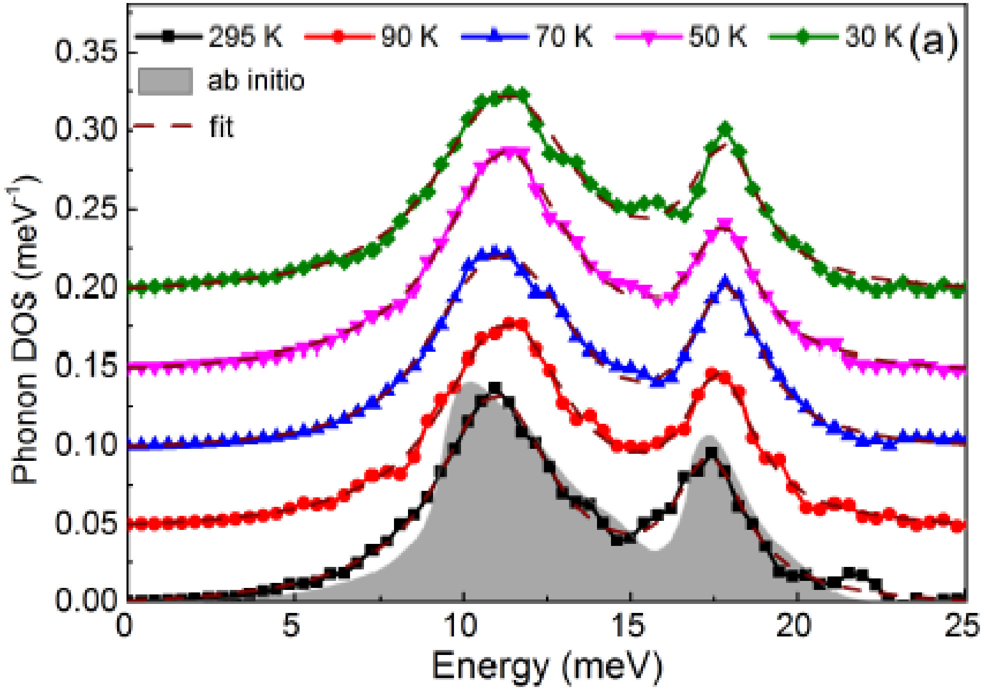
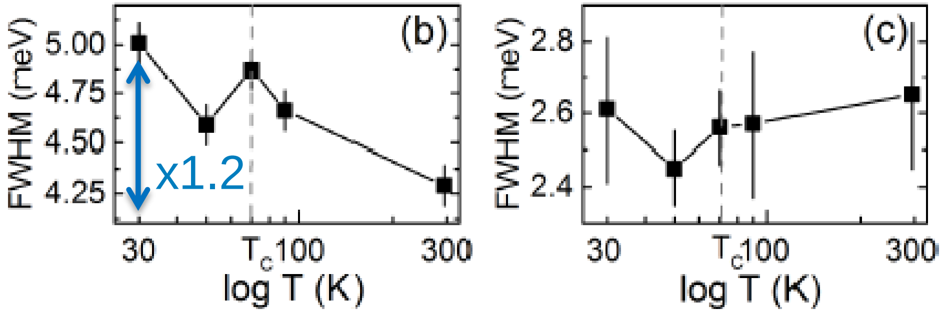
- Lattice dynamics of EuO: evidence for giant spin-phonon coupling

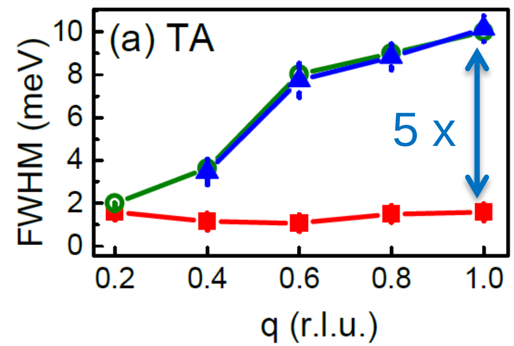
Eu-partial phonon DOS from NIS experiment



FWHM of the peaks in the DOS



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



R. Pradip et al., Phys. Rev. Lett. ***116****, 185501 (2016)*

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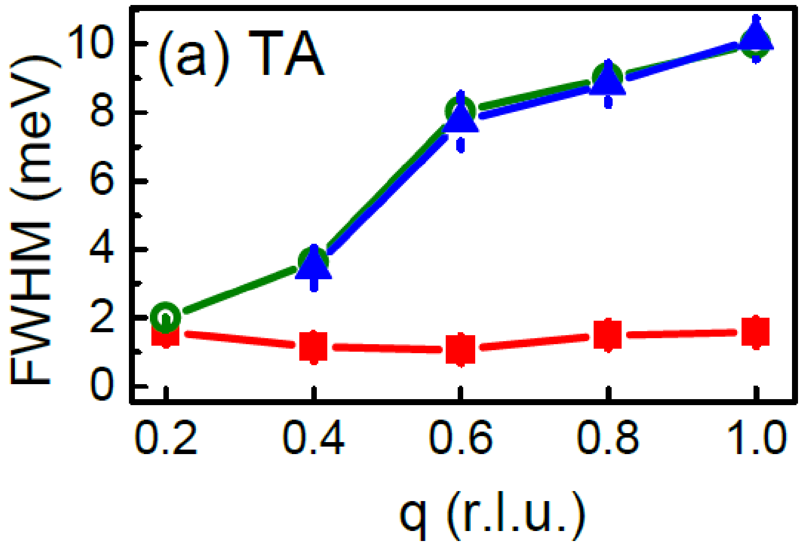

$$\Delta \mathrm{FWHM} \approx \hbar\omega \left(\frac{z(\alpha J)^2}{K} \right) \chi_s''(\hbar\omega)$$

For Δ FWHM =8 meV,
spin-phonon coupling constant, $\alpha \approx 10$!

ω: phonon energy; J: exchange energy, K: force constant;
z: coordination number; X: spectral density of spin waves

C. Ulrich et al., Phys. Rev. Lett. ***115****, 156403 (2015)*

$CuGeO_3$ α=5.9	YBaCuO α=10.4	$NaOsO_3$ $\alpha \approx$11.8

R. Pradip et al.,
Phys. Rev. Lett. ***116****, 185501 (2016)*

R. Werner et al.,
PRB ***59****, 14356 (1999)*

J. P. Carbotte et al.,
Nature ***401****, 354 (1999)*

S. Calder et al.,
Nat. Commun. ***6****, (2015)*

Conclusions and outlook

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- 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 !