Spin and lattice correlations in quantum magnets and the full elasticity tensor from thermal diffuse scattering
Part 1:

Magneto-elastic waves in quantum magnets

- Strong spin-lattice coupling allows probing magnetic correlations using non-resonant IXS
Magnon – Phonon coupling in Quantum Magnets

- Coupling between magnetic degrees of freedom and lattice vibrations
- Enables control mechanism for magnetic and lattice excitations
- Phonon measurements for probing magnetic correlations
- Critical fluctuations: Collective excitations near quantum critical points in low dimensional systems
Spin-lattice coupling by exchange striction

Modulation of direct exchange

- $\alpha = \frac{dJ}{dr} \cdot \frac{d}{J_0}$
- chromates: $\alpha = 30$

Modulation of super exchange

- $\alpha = \frac{dJ}{dr} \cdot \frac{2d}{J_0}$
- cuprates: $\alpha = 2 - 7$
- manganates: $\alpha = 10 - 20$
- chromates: $\alpha = 10$
Magnon-Phonon Spectrum

Assuming linear coupling

\[ H = J_0 \left[ \frac{\omega_{PH}(Q)}{\alpha} + \omega_{MAG}(Q) \right] \]

Change of dispersion:
- Anti-crossing (small \(\alpha\))
- Energy shift (large \(\alpha\))
- Mixed (hybridized) excitations
LiCrO$_2$ probed by IXS

- Triangular lattice Heisenberg antiferromagnet
- Spin 3/2
- 120° helical magnetic order $T_N = 61.2$ K
- ID28, ESRF, France

Excitation spectra along HHL

\[ H = J_1 \sum_{ij} S_i \cdot S_j + J_{mp} \sum_{ij} \hat{d}_{ij} \cdot (u_i - u_j)S_i \cdot S_j + H_p \]


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Excitation width measurements

- Finite life-time of excitations
- Momentum dependence
- Coupling to two-magonon continuum

- Enhanced decay due to phonon hybridization
- Brake-down of non-interaction magnon picture

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Metallic Antiferromagnet PdCrO$_2$

- Triangular lattice Heisenberg antiferromagnet
- Strongly correlated CrO$_2$ layers carry localized magnetic moments
- High electric conductivity due to Pd layers.
- Unconventional anomalous Quantum Hall effect

- Strong interplay between itinerant electrons and localized magnetic moments

Hybridized excitations in PdCrO$_2$

- Hybridized excitations due to spin-lattice coupling

- Coupling to conducting electrons entangled in decay of magneto-elastic excitations with temperature

- BL35XU, SPring-8, Japan
Hybrid excitations at high pressures

IXS at multi-extreme conditions
ID28, ESRF, France

4K flow cryostat for membrane-driven
Diamond anvil cells

$\alpha = \frac{dJ}{dr} \cdot \frac{d}{J_0}$
LiCrO$_2$ at high pressures

LiCrO$_2$ at high pressure and low temperature:
- Pressure strongly affects energy and linewidth of hybridized excitation

➤ Significant increase of magnetic exchange and exchange striction with pressure

Pressure control of magnetic exchange:
Part 2:

Full elasticity tensor from thermal diffuse scattering

New method for simultaneous study of structure and anisotropic elasticity
Elasticity tensor

- Mechanical properties of materials
- Phase stability
- Electron-phonon interaction
- Magnon-phonon interaction
- Sound wave propagation
- Improved refinements in crystallography


Quantum magnet

SrCu$_2$(BO$_3$)$_2$

Seismic wave propagation

Exploring the Earth using seismology

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Thermal diffuse scattering

β-tin

Experimental requirements

Incoming beam:
- Monochromatic X-rays
- Collimation
- Good background shielding

Diffractometer:
- 4-Circle Kappa Goniometer
- Flexible sample environment

Detector:
- Single photon counting area detector
- Large dynamical range
- No electronic noise
- Adjustable energy threshold

ID28 at ESRF: new side station dedicated for diffuse scattering
Model calculations

Thermal diffuse scattering of $\beta$-tin: experiment versus calculation

![Graph showing model calculations](image)


**Ab2tds** – open source program for calculating inelastic and diffuse scattering intensities

Full elasticity tensor from thermal diffuse scattering

- Elastic waves, equation of motion
  \[ \rho \omega^2 u_i = c_{ijkl} u_j k_l u_m \]
- Conditions for momentum transfer
- Fitting intensity difference
- Simultaneous fit in 3D reciprocal space of
  \( \sim 10^7 \) pixels with GPU acceleration

Calcite

Reference: Ultrasound measurements
Calc. (i) Acoustic phonons, single phonon scattering
Calc. (ii) All phonon branches
Calc. (iii) Acoustic phonons including two-phonon scattering

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Diffuse scattering at extreme conditions

- Simultaneous measurement of elasticity and crystal structure
- Precise measurement of incoming flux and absorption
- Temperature dependent study
- Application to interdisciplinary problems

High-pressure diffuse scattering ID27 at ESRF
High-pressure elasticity tensor

MgO at high pressures


MgO elastic moduli compared to Brillouin scattering
Diffuse scattering in quantum magnets

Magnetic diffuse scattering in resonant condition:
- Critical scattering in vicinity of quantum phase transitions
- Quantitative study of disorder and correlation
- Full anisotropic tensor of magnon-phonon coupling

Na$_2$IrO$_3$


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Summary

Spin – lattice coupling in quantum magnets:
- Hybrid excitations visible by non-resonant IXS
- Dispersive electromagnons with distinct momentum-dependent width
- Coupling to two-magnon continuum and conducting electrons
- Pressure control of magnetic correlations

Full elasticity tensor from thermal diffuse scattering:
- Absolute values of full tensor to high precision
- Application to high pressure
- Crystal structure and elasticity in single measurement

Perspectives:
- Critical fluctuations: Collective excitations near quantum critical points in low dimensional systems
- Magnetic diffuse scattering in resonant conditions
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