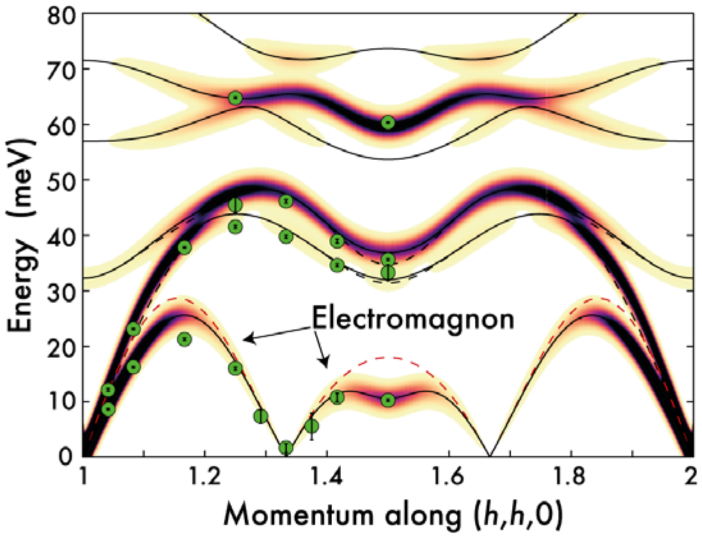
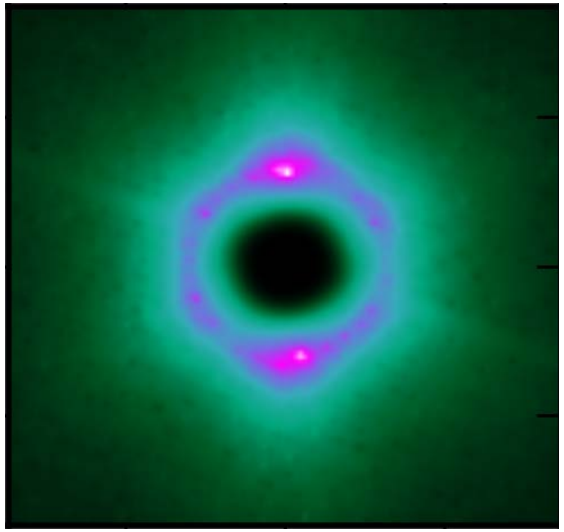
Spin and lattice correlations in quantum magnets and the full elasticity tensor from thermal diffuse scattering





Björn Wehinger
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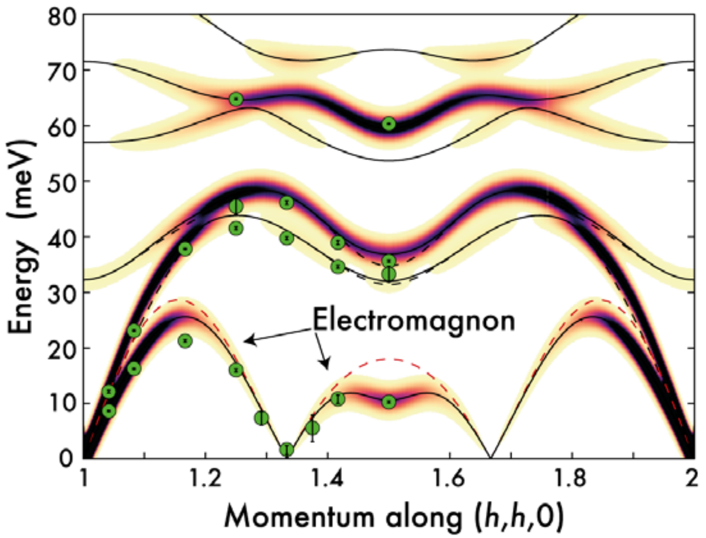
UNIVERSITÉ DE GENÈVE

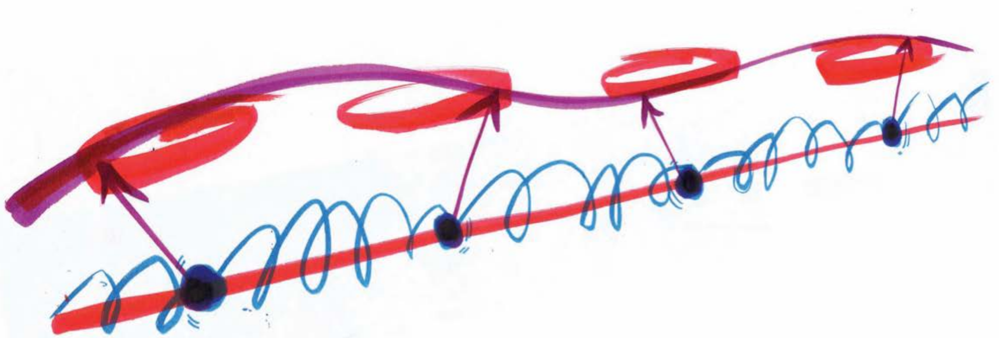
Part 1:

Magneto-elastic waves in quantum magnets



- **Strong spin-lattice coupling allows probing magnetic correlations using non-resonant IXS**

Magnon – Phonon coupling in Quantum Magnets



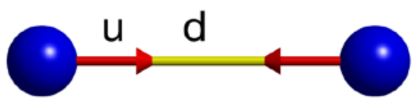
Electromagnons in real space

- 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

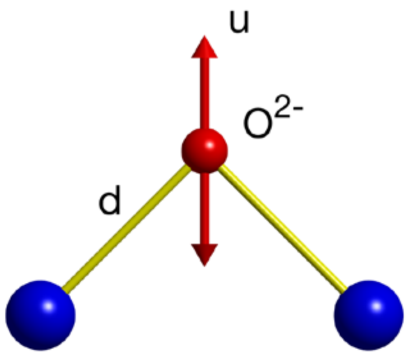
Modulation of direct exchange

- $\alpha = \frac{dJ}{dr} \cdot \frac{d}{J_0}$
- 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: $\alpha = 10$



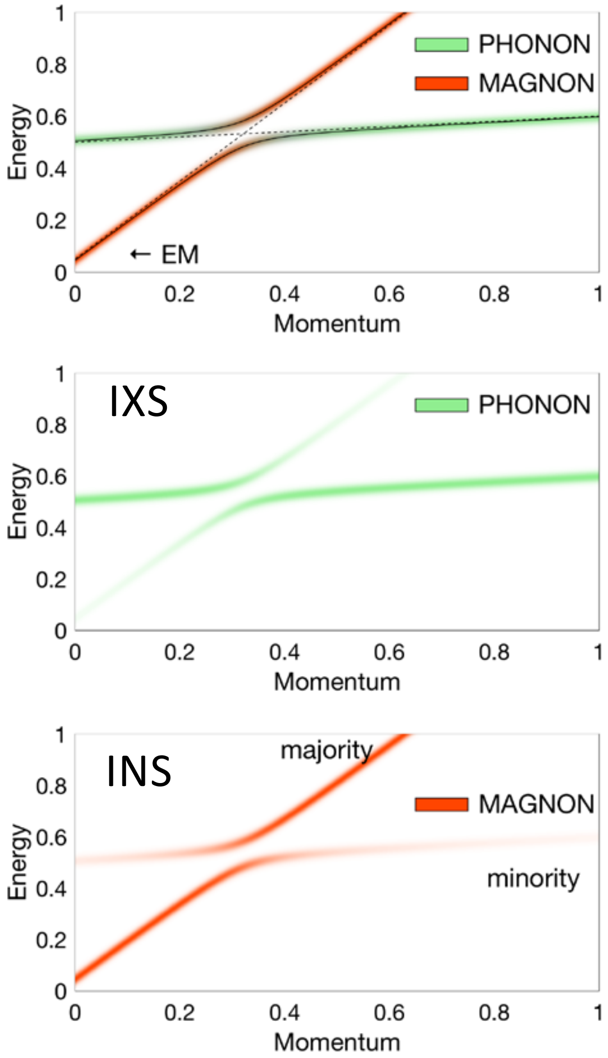
Magnon-Phonon Spectrum

Assuming linear coupling

$$H = J_0 \begin{bmatrix} \omega_{PH}(Q)/J_0 & \alpha \\ \alpha & \omega_{MAG}(Q) \end{bmatrix}$$

Change of dispersion:

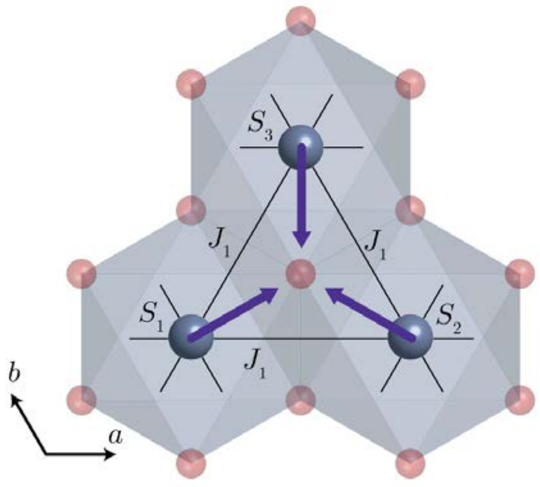
- Anti-crossing (small α)
- Energy shift (large α)
- Mixed (hybridized) excitations



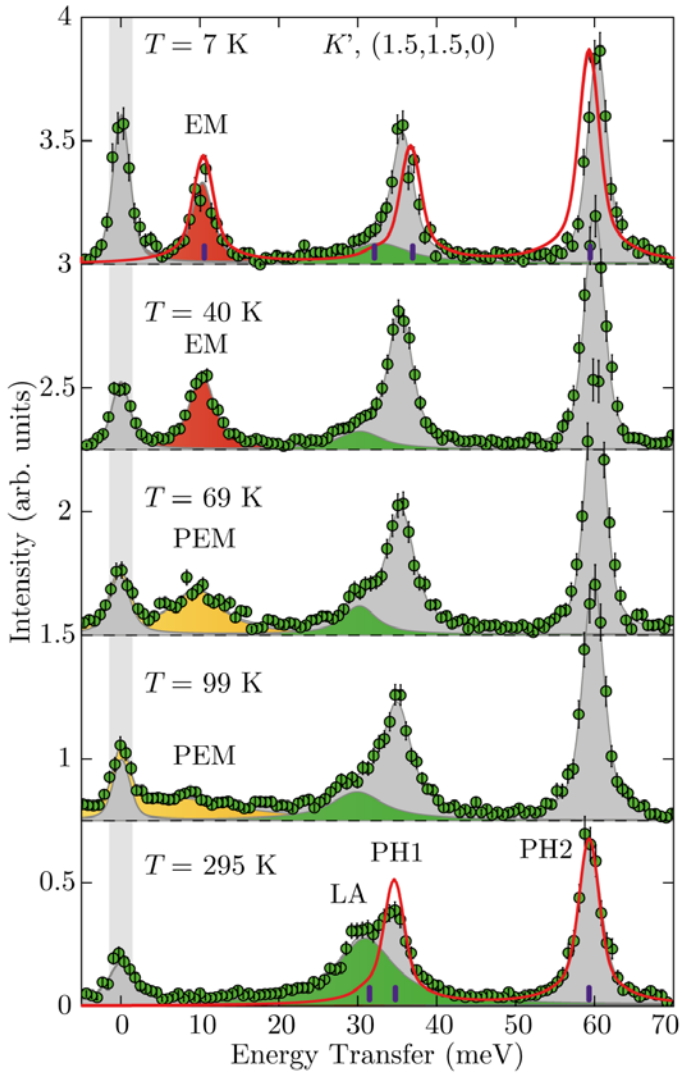
$LiCrO_2$ probed by IXS

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

- ID28, ESRF, France

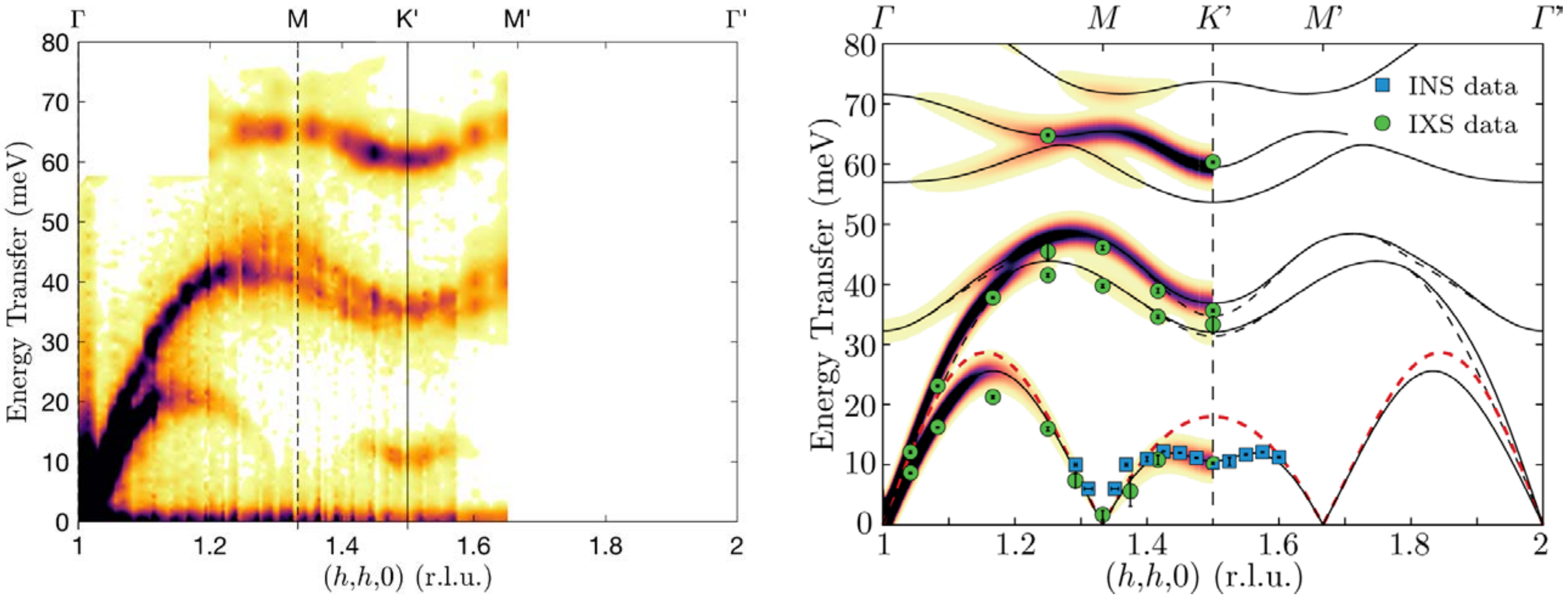


$LiCrO_2$



Toth and Wehinger et al. *Nat. Comm.* **7**, 13547 (2016).

Excitation spectra along HHL



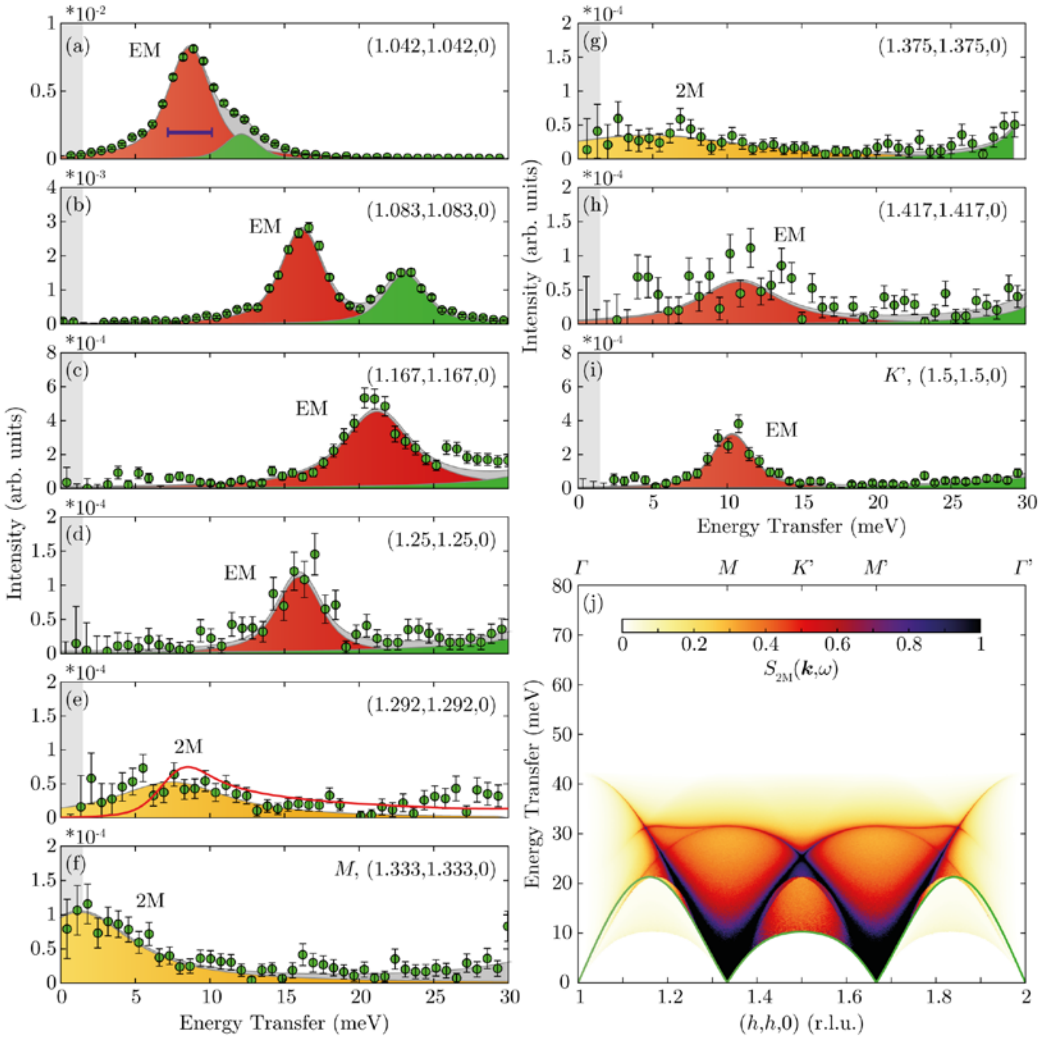
$LiCrO_2$ – IXS measurement T = 7K

IXS and INS versus calculation

$$\mathcal{H} = J_1 \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + J_{\mathrm{mp}} \sum_{ij} \hat{\mathbf{d}}_{ij} \cdot (\mathbf{u}_i - \mathbf{u}_j) \mathbf{S}_i \cdot \mathbf{S}_j + \mathcal{H}_{\mathrm{p}}$$

S. Tóth and B. Wehinger *et al.*, *Nat. Commun.*, **7**, 13547 (2016).

Excitation width measurements

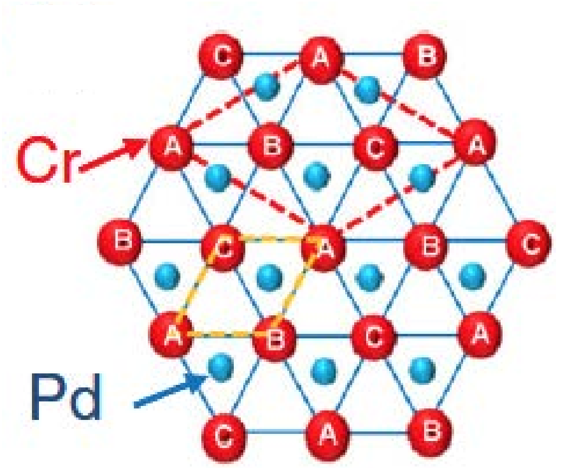


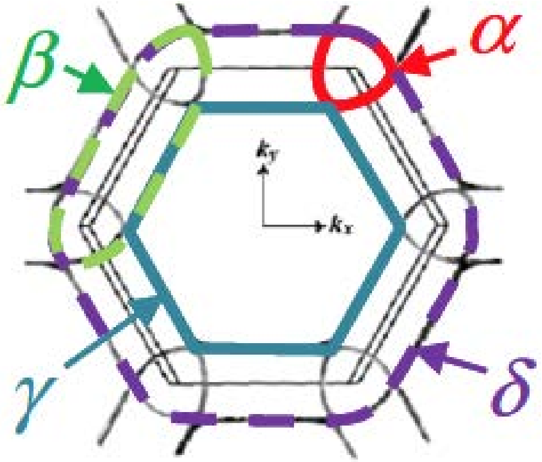
- Finite life-time of excitations
- Momentum dependence
- Coupling to two-magonon continuum

➢ **Enhanced decay due to phonon hybridization**

➢ **Brake-down of non-interaction magnon picture**

Metallic Antiferromagnet $PdCrO_2$





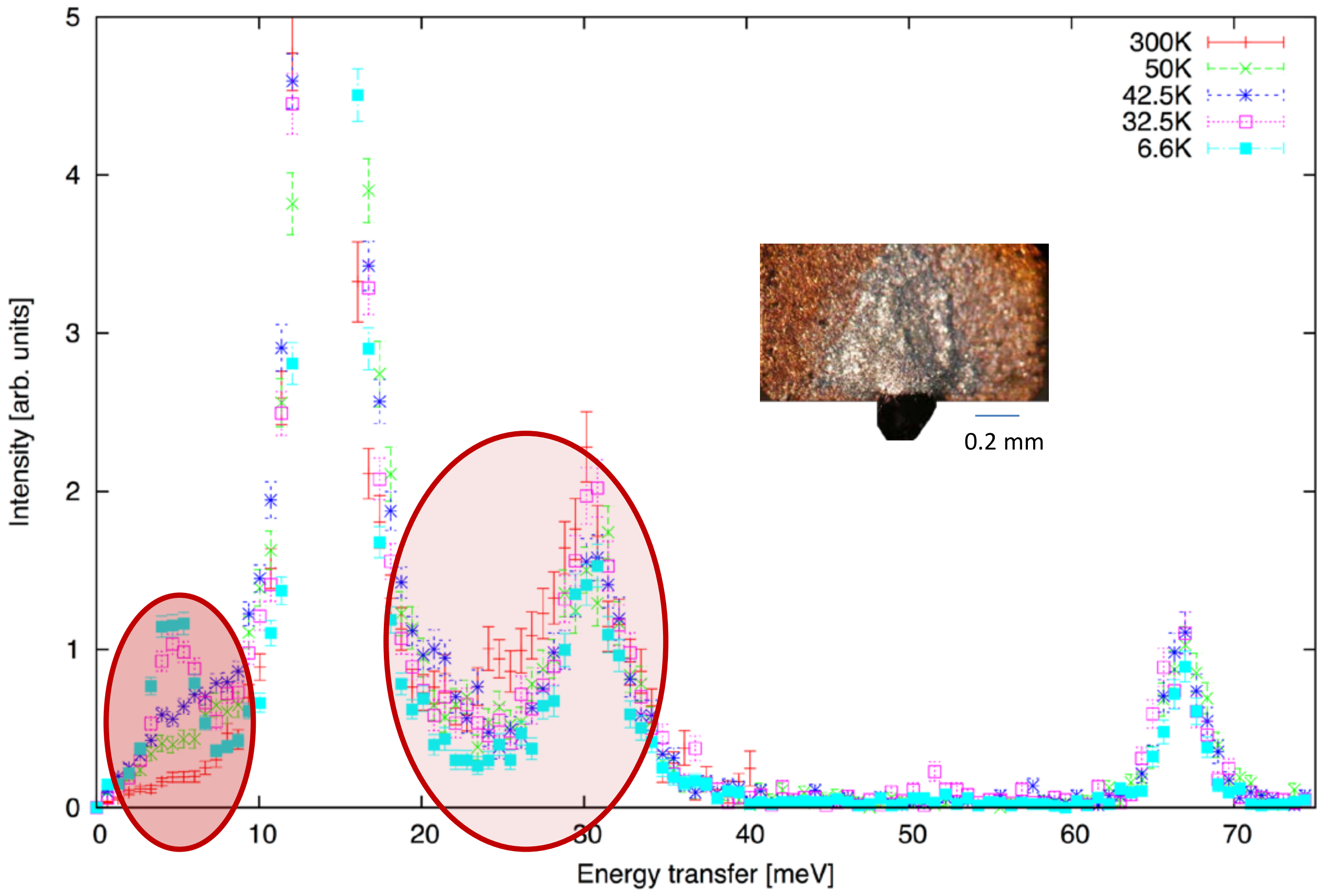
$PdCrO_2$

- 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**

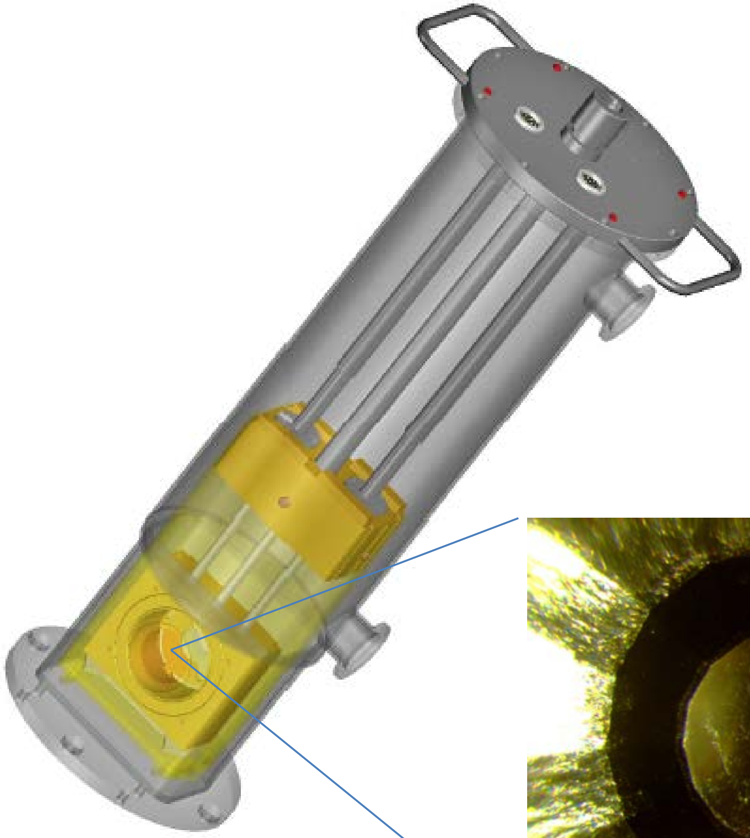
Ok et al., *Phys. Rev. Lett.* **111**, 176405 (2013).

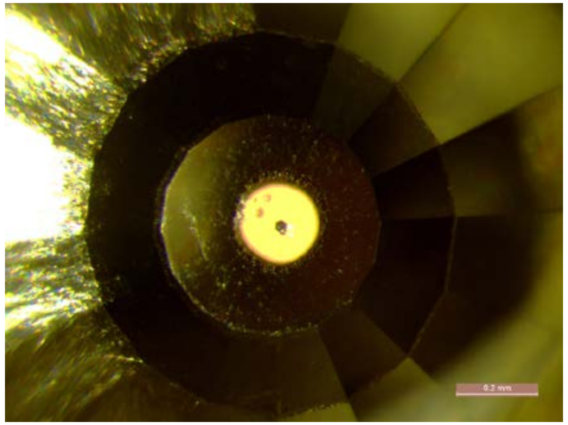
Hybridized excitations in $PdCrO_2$

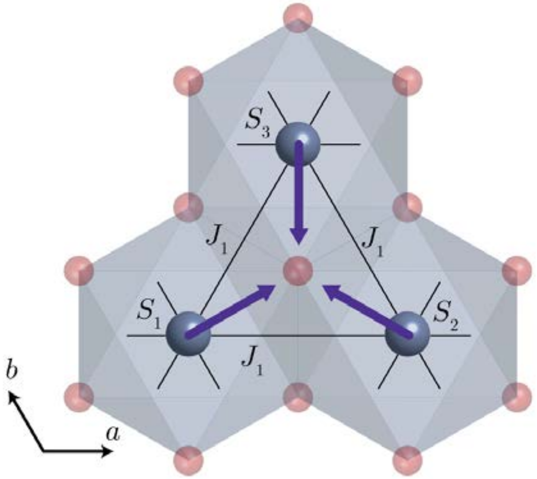


- Hybridized excitations due to spin-lattice coupling
- Coupling to conducting electrons entangled in decay of magneto-elastic excitations with temperature
- BL35XU, SPring-8, Japan

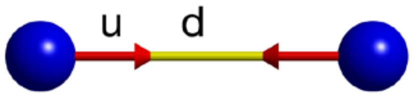
Hybrid excitations at high pressures







LiCrO$_2$

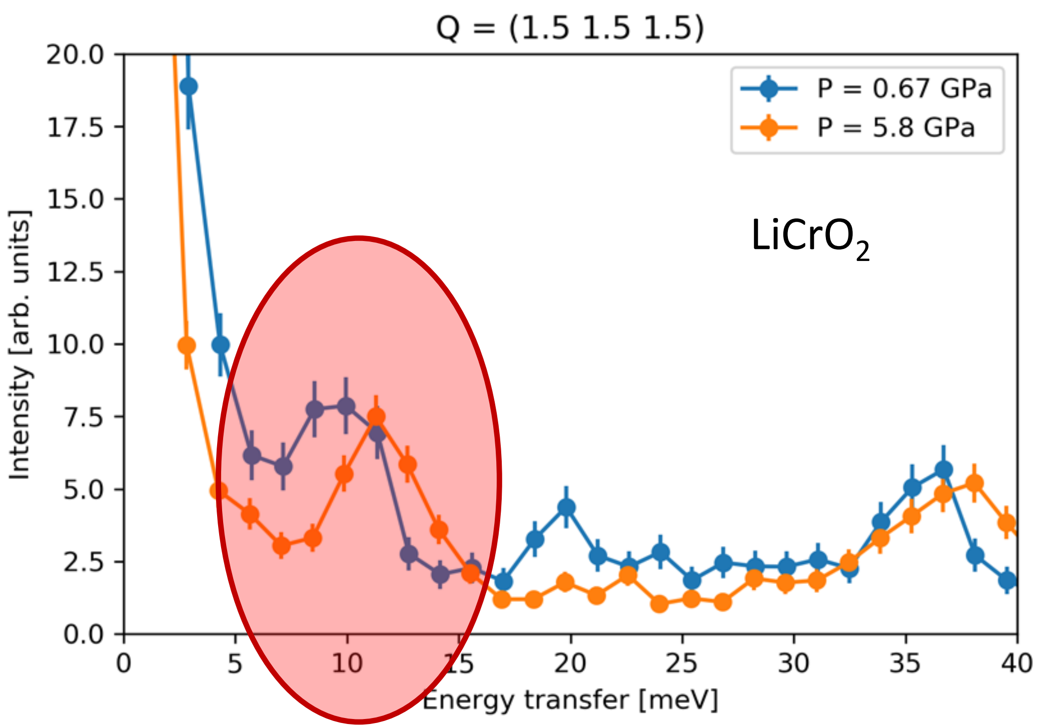


$$\alpha = \frac{dJ}{dr} \cdot \frac{d}{J_0}$$

IXS at multi-extreme conditions
ID28, ESRF, France

4K flow cryostat for membrane-driven
Diamond anvil cells

$LiCrO_2$ at high pressures



$LiCrO_2$ at high pressure and low temperature:

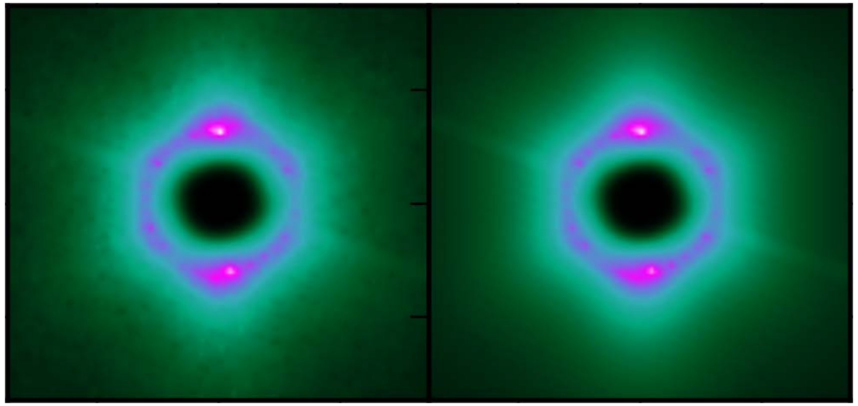
- Pressure strongly affects energy and linewidth of hybridized excitation

➢ **Significant increase of magnetic exchange and exchange striction with pressure**

Pressure control of magnetic exchange:
B. Wehinger et al., *Phys. Rev. Lett.* **121**, 117201 (2018).

Part 2:

Full elasticity tensor from thermal diffuse scattering

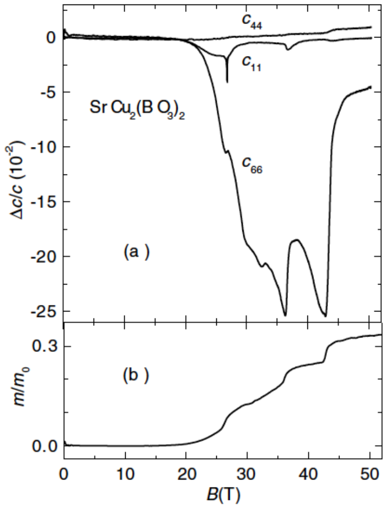


New method for simultaneous study of structure and anisotropic elasticity

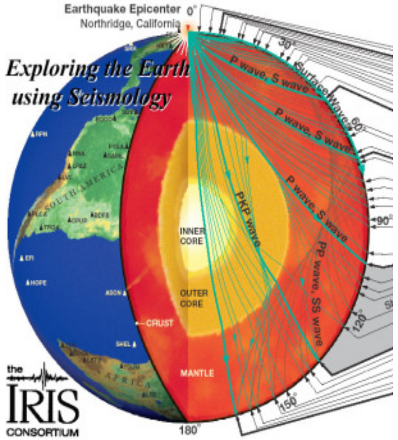
Elasticity tensor

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

B. Wehinger *et al*. *Phys. Rev. Lett.* **118**, 035502 (2017).

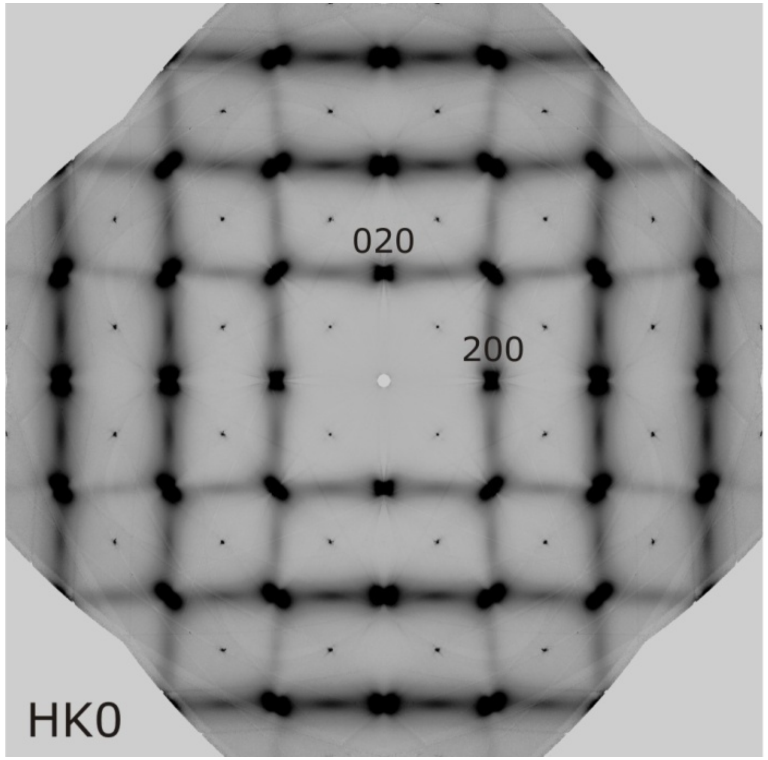


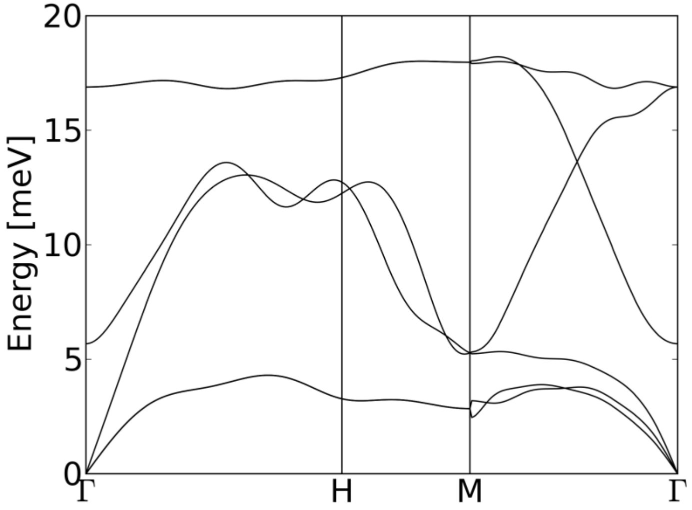
Quantum magnet $SrCu_2(BO_3)_2$

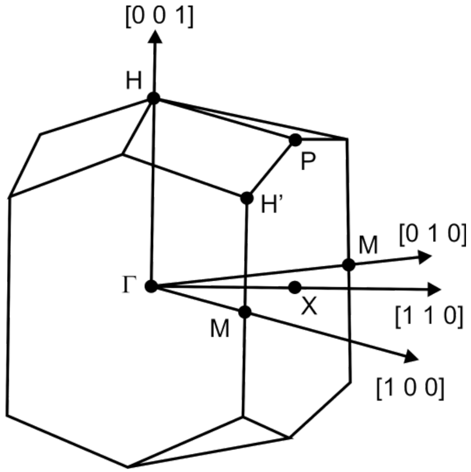


Seismic wave propagation

Thermal diffuse scattering







β-tin

B. Wehinger *et al. J. Phys.: Condens. Matter* **26** 115401 (2014).

Experimental requirements

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

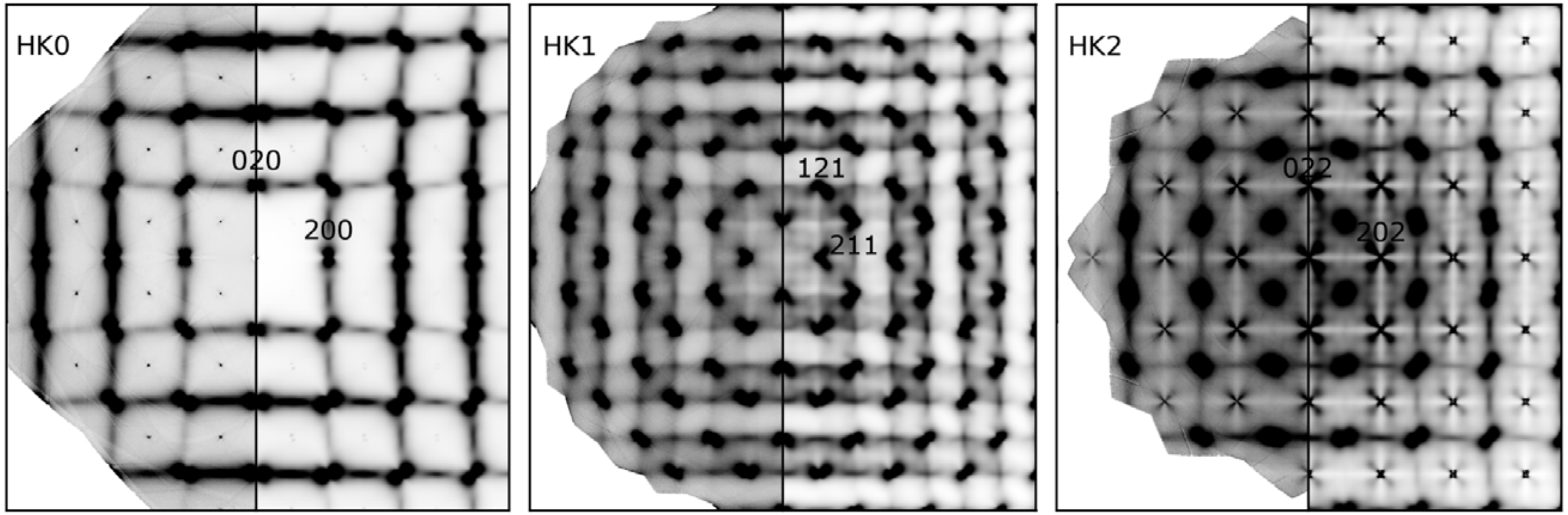
Diffractometer:
- 4-Circle Kappa Goniometer
- Flexible sample environment

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 β-tin: experiment versus calculation



B. Wehinger *et al. J. Phys.: Condens. Matter* **26** 115401 (2014).

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

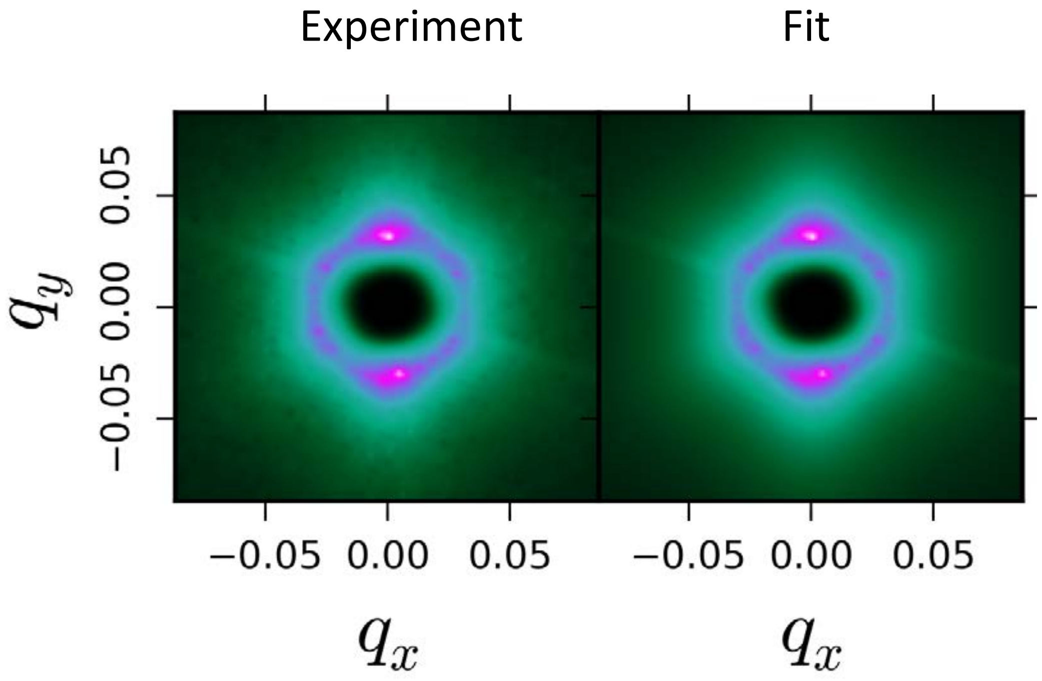
B. Wehinger and A. Mirone, http://ftp.esrf.fr/scisoft/AB2TDS/ (2013).

Full elasticity tensor from thermal diffuse scattering

- Elastic waves, equation of motion

$$\rho\omega^2 u_i = c_{ijlm} k_j k_l u_m$$

- Conditions for momentum transfer
- Fitting intensity difference
- Simultaneous fit in 3D reciprocal space of $\sim 10^7$ pixels with GPU acceleration



Calcite

Björn Wehinger *et al*. *Phys. Rev. Lett.* **118**, 035502 (2017).
A. Mirone and B. Wehinger, TDS2EL, http://ftp.esrf.fr/scisoft/TDS2EL (2017).

Calcite

	TDS	Ref.[23]	rel. diff.	Calc. (i)	Calc. (ii)	Calc. (iii)
c_{11}	156	155.1	0.6%	154.3	153.5	154.2
c_{13}	57.0	57.69	1.1%	57.74	57.86	57.81
c_{15}	21.2	21.51	1.6%	21.11	20.86	20.94
c_{33}	87.6	87.12	0.6%	86.91	86.88	86.80
c_{44}	35.7	34.42	3.7%	33.95	33.60	33.75
c_{66}	48.0	47.80	0.4%	46.53	46.06	46.36

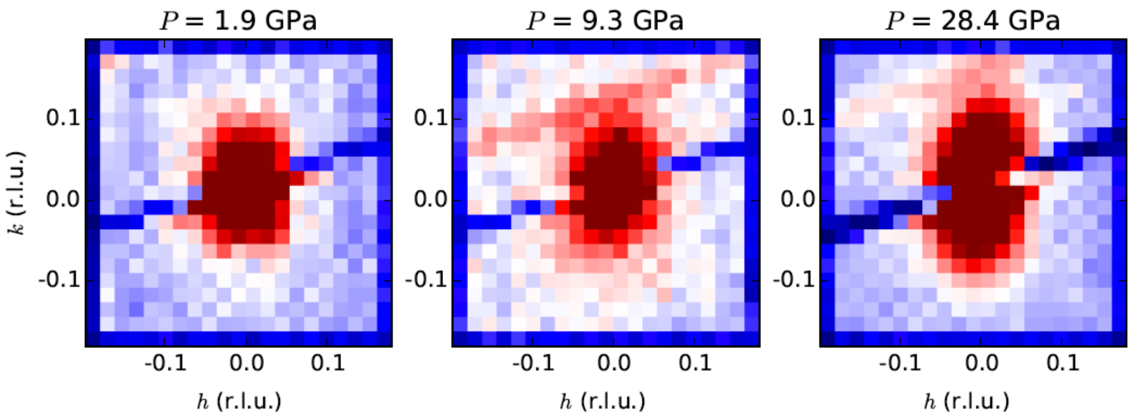
Reference: Ultrasound measurements
Calc. (i) Acoustic phonons, single phonon scattering
Calc. (ii) All phonon branches
Calc. (iii) Acoustic phonons including two-phonon scattering

Diffuse scattering at extreme conditions

High-pressure diffuse scattering
ID27 at ESRF

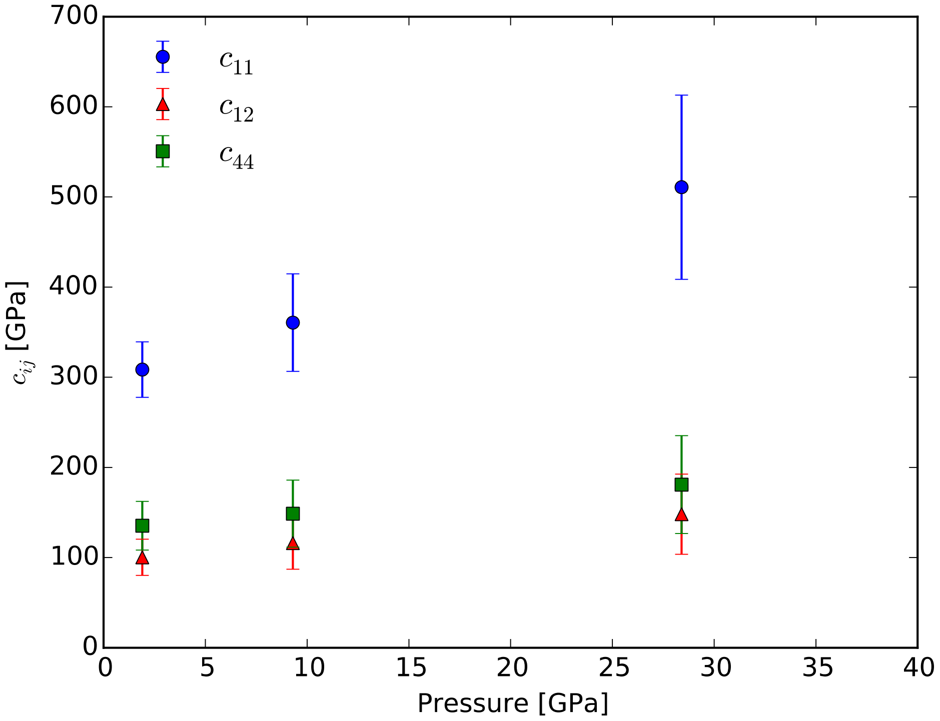
- Simultaneous measurement of elasticity and crystal structure
- Precise measurement of incoming flux and absorption
- Temperature dependent study
- Application to interdisciplinary problems

High-pressure elasticity tensor



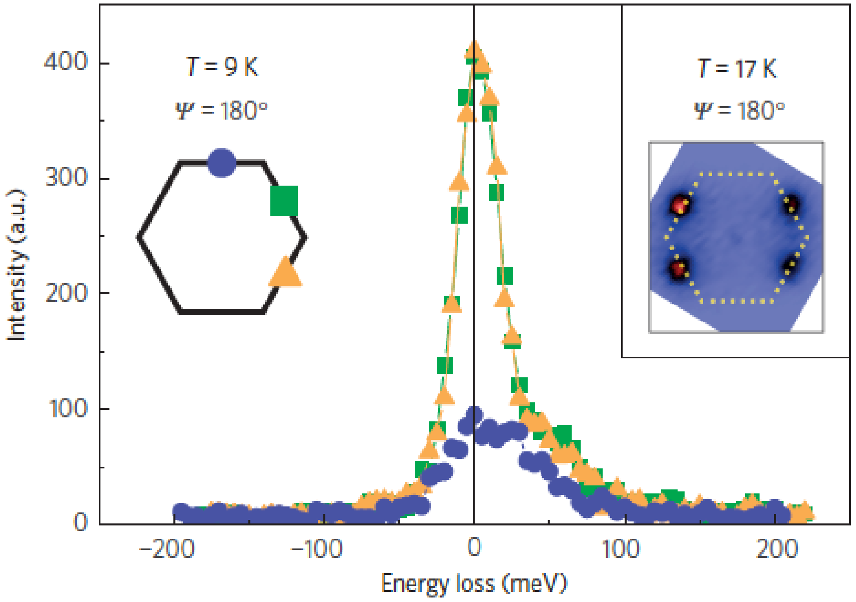
MgO at high pressures

B. Wehinger *et al. arXiv:*1808.04595 (2018).



MgO elastic moduli compared to Brillouin scattering

Diffuse scattering in quantum magnets



Na_2IrO_3

Chun et al., *Nat. Phys.* **11**, 462 (2015).

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

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

Acknowledgement

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