

First Inelastic Neutron Scattering from a SrRuO₃ Thin Film

SIKA makes it possible to investigate the magnetic property of single-crystal epitaxial thin film with inelastic neutron scattering

A bulk crystal or multiple crystals of mass of order gram are necessary for a successful detection of extremely weak signals. No successful inelastic neutron scattering (INS) measurement on films has been reported, but most materials are difficult to grow into large high-quality crystals, and some of them can be grown as high-quality single-crystal films. Pushing the INS measurement to the limit of a film is now extremely crucial. Hsiung Chou of National Sun Yat-sen University and his collaborators have used SrRuO₃ single-crystal epitaxial thin films (Fig. 1) to investigate the magnon dispersion curve in this ferromagnetic system to extend the limitation of INS measurements and to understand better the underlying mechanisms. This limitation imposes a strong constraint on the applicability of neutron inelastic-scattering instruments. Even though many materials can be grown as single-crystal films, their small mass makes this limitation a significant barrier. If the material has, however, a specific property that has a strong correlation with the low excitation phase along a specific direction and is measured with a highly sensitive and low-noise neutron inelastic-scattering instrument, this limitation might be overcome. Taiwan has built a state-of-the-art neutron inelastic-scattering instrument, **SIKA**, in Australian Nuclear Science and Technology Organisation (ANSTO), which has the potential to undertake neutron inelastic-scattering measurement on thin films.

Chou's group has published an example¹ on choosing SrRuO₃ (SRO), which is one of few itinerant ferromagnetic materials from the 4d-transition-metal group widely used as a conducting electrode in multilayer device applications, magnetic tunnel junctions, electronic transport tuning, dynamic random-access memory applications, switchable acoustic-wave resonators and spintronic devices, among others. Unexplained phenomena such as the non-saturated magnetization at a large magnetic field, changing of the sign of Hall coefficients, an unbelievable high upper-limit-temperature ($T \leq 30$ K) for Fermi liquid conduction and non-Fermi-liquid behavior within $90 \leq T/K \leq 150$, are, however, waiting to be explored. Chou's team has hence a great motivation to challenge the unbreakable limitation for inelastic neutron scattering on a thin film as well as to understand the exotic magnon dispersion profile of SrRuO₃ films.

To confirm that their observed signal is really contributed by a magnon and not by phonons or instrumental errors, it was necessary to compare the result at high temperature at which the material is in a paramagnetic state.

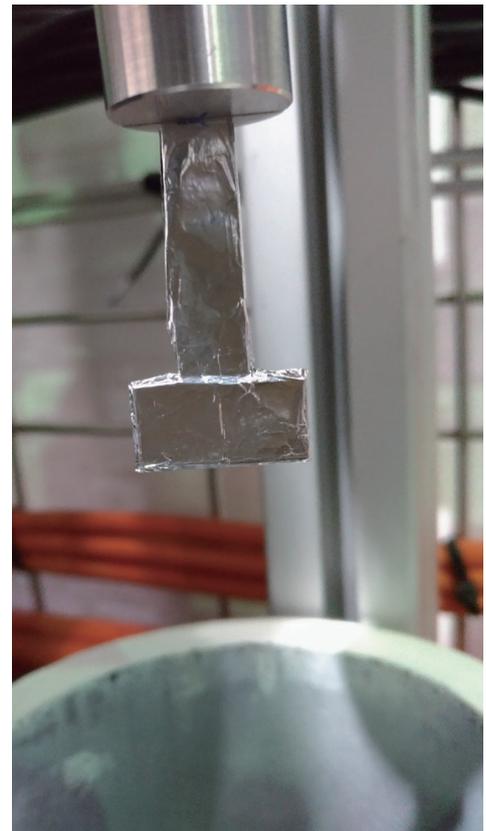


Fig. 1: Four 1×1 cm² SrRuO₃ films, stacked with the film's surface in contact with each other and placed side by side on an aluminium sample holder. [Courtesy of Chun-Ming Wu]

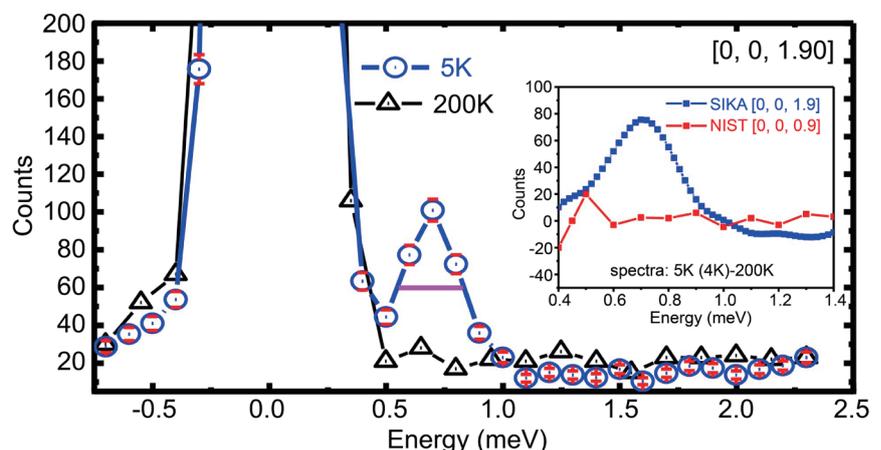


Fig. 2: Direction [002] shows the largest count difference, which indicates that the magnon dispersion curve that extends along this direction should be the most readily detectable. INS spectra of $L = 1.90$ at 5 K and 200 K. [Reproduced from Ref. 1]

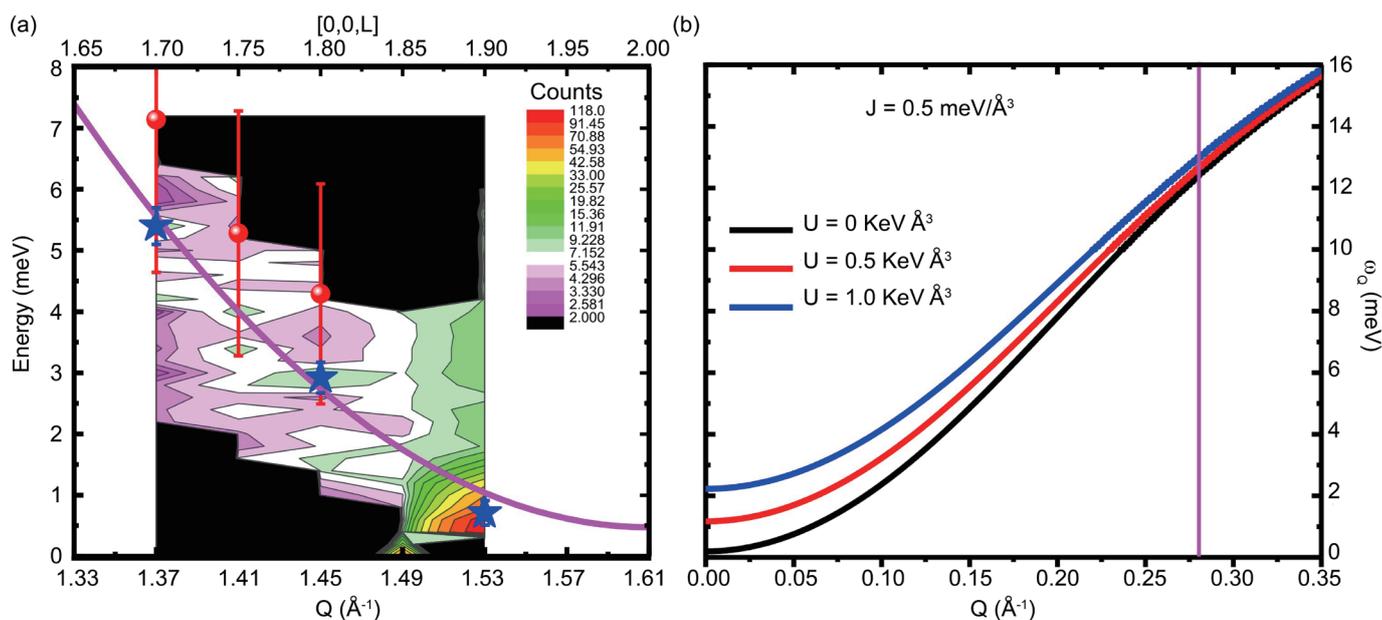


Fig. 3: (a) Relation between E and L (or Q). The magnon spectrum follows the dispersion relation ($E \propto Q^2$). The data points presented as red circles are obtained from Ref. 2. The data points obtained from Chou's measurements match well with Ref. 2 and are situated well within their FWHM. (b) Magnon dispersions for varied Coulomb U at temperature $T = 0$. Magnetic coupling $J = 0.5 \text{ meV \AA}^3$, conduction-band electron density $n_e = 1 \times 10^4 \text{ \AA}^{-3}$ and Ru occupation $c_{\text{Ru}} = 10^{-3} \text{ \AA}^{-3}$. [Reproduced from Ref. 1]

The same measurement results at 5 K and 200 K are plotted on a linear scale (Fig. 2). The center signals at 0 meV overlap perfectly whereas the signal about 0.7 meV at 5 K disappears at 200 K. This observation confirmed a magnon peak and also showed a successful breaking of the concept of an inability to measure INS of films.

Using the present identified magnon signal, they further obtained a dispersion curve, in Fig. 3. Fitting the data points with the dispersion relation yielded $J = 1.88 \text{ meV}$. This magnon band gap 0.32 meV is much smaller than Itoh's 2 meV^2 and slightly smaller than 1 meV estimated from a FMR measurement calculated from the anisotropy energy estimated with bulk magnetization.

In summary, Chou and his collaborators successfully demonstrated that measuring a low-energy excitation with inelastic neutron scattering on a single crystal film is possible. Because of the strong strain effect at the STO/SRO interface, the crystal structure of SRO was tuned to a tetragonal structure with out-of-plane lattice parameter slightly larger than the in-plane lattice parameter, about 0.1%. This change suppresses the structural distortion and symmetry and produces a smaller magnon gap. (Reported by Chun-Ming Wu)

This report features the work of Hsiung Chou and his collaborators published in Physical Review B 101, 054403 (2020).

ANSTO SIKA – Cold Neutron Triple-axis Spectrometer

- INS
- Ferromagnetism, Magnon, Inelastic Neutron Scattering, Epitaxial Single-Crystal Thin Film

References

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2. S. Itoh, Y. Endoh, T. Yokoo, S. Ibuka, J.-G. Park, Y. Kaneko, K.-S. Takahashi, Y. Tokura, N. Nagaosa, *Nat. commun* **7**, 11788 (2016).

