Commensurate locking of magnetic skyrmion and crystal lattice in $Gd_3Ru_4Al_{12}$

Magnetic skyrmions are vortex-like, localized objects formed by the spin degree of freedom in solids. Their winding spin texture, when projected onto a sphere, forms a perfect hedgehog (Fig. 1(a)). Some high-symmetry models of frustrated magnetism show a tendency to form a skyrmions lattice (SkL): for example, Heisenberg models of localized spins with beyond-nearest neighbor interactions [1] or models of itinerant electrons coupled to localized spin moments [2]. In these types of models, the size of a magnetic vortex can be exceedingly small, measuring only a few lattice constants across; due to their localized nature, such nano-skyrmions may be suitable to carry and store information in future electronic circuitry. Previously, all skyrmion textures experimentally observed in nature were found to be incommensurate to the underlying lattice, that is, their period is not a multiple of the lattice constant of the atomic structure that supports them. However, relativistic spin-orbit coupling – which causes magneto-crystalline anisotropy (MCA) and anisotropic exchange energies – can lock the skyrmion to the lattice [3]. In this case, the period of the magnetic texture is a multiple of the lattice constant and the core of the spin vortex has a well-defined relationship with respect to the atomic lattice ('on-site' or 'off-site', for example).

Our target material is hexagonal $Gd_3Ru_4Al_{12}$, which crystallizes in space group $P6_3/mmc$ (#194). In this structure, the magnetic gadolinium ion forms a distorted ('breathing') Kagome network of triangles and hexagons, where certain gaps or holes in the lattice may favor the commensurate locking. The crystals were grown by the floating zone technique and thoroughly characterized by chemical and X-ray methods. Magnetization and electric resistivity are measured to verify the bulk magnetic phase transitions and phase diagram of the material. High quality pieces are polished into a regular shape (rectangular) and aligned by in-house Laue X-ray diffraction. Thus, flat and oriented surfaces are prepared for the synchrotron experiment.

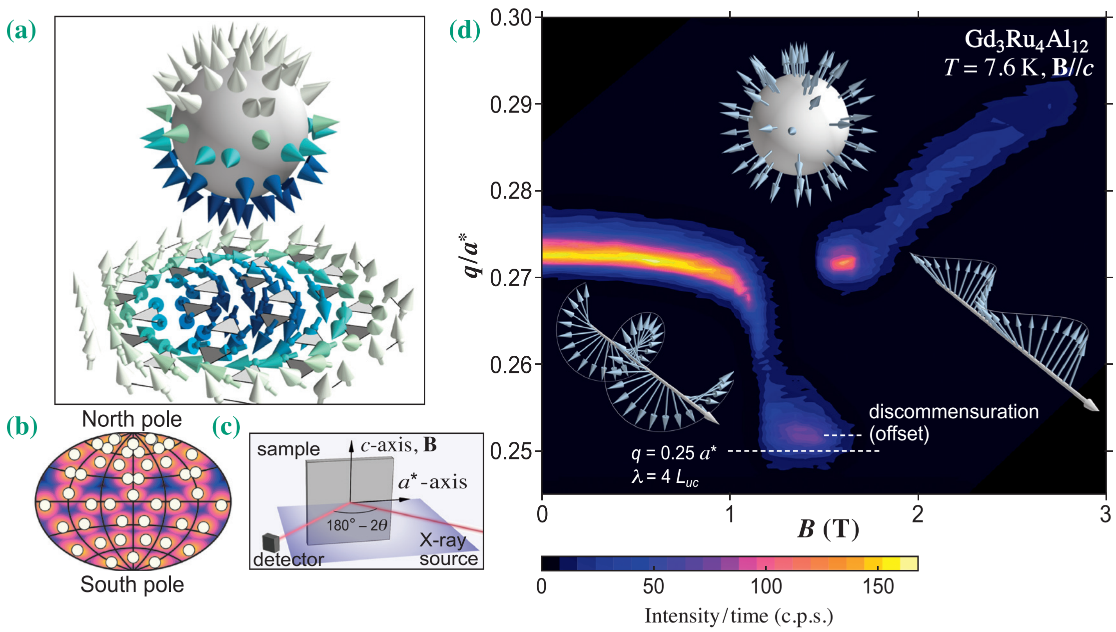

Fig. 1. Commensurate locking of the skyrmion lattice phase in $Gd_3Ru_4Al_{12}$. **(a)** Magnetic moments of a skyrmion winding a sphere (top), and corresponding two-dimensional magnetic moment texture in real space (bottom). Each arrow corresponds to a site of the magnetic sublattice. **(b)** Map projection showing a hemisphere of (a) unfolded, with white dots indicating directions of magnetic moments. The color code indicates the distance of each point on the sphere from the nearest moment on the sphere, in radians. **(c)** Experimental geometry of resonant elastic X-ray scattering (REXS), where the pink line is the trajectory of the X-ray beam. **(d)** Scattering intensity in REXS as a function of magnetic field (B) and momentum transfer q from $\boldsymbol{q} = (q, 0, 0)$; $q = 0.25$ corresponds to four times the projection of the lattice constant parallel to $\boldsymbol{q}$. Incommensurate proper screw (IC-PS), commensurate skyrmion lattice (C-SkL), and IC fan phases are illustrated by insets. In C-SkL, the periodicity of the magnetic texture is locked to the crystal lattice up to a weak discommensuration (offset) of $\Delta q = 0.0018$ r.l.u. B denotes magnetic induction after demagnetization correction.

We studied the magnetic structure of the intermetallic $Gd_3Ru_4Al_{12}$ by resonant elastic X-ray scattering (REXS) at SPring-8 **BL19LXU**. The energy of SPring-8's powerful synchrotron X-ray beam was set to 7.9 keV, around the L_2 absorption edge of Gadolinium. Focusing on the (electric) dipole component of the resonant scattering process to detect magnetic textures, we find that the SkL in this material is nearly perfectly locked to the underlying crystal lattice, barring some defects every 100 unit cells or so [4]. More specifically, we discovered that the magnetic phases that surround the SkL in the *B*–*T* phase diagram – proper screw spiral, transverse cone cycloidal, and fan-type – are incommensurate, while the SkL is commensurate. In particular, Fig. 1(d) shows the wavevector $q = 2\pi/\lambda$ measured in units of the reciprocal lattice constant a^*. It shows a sharp dip when entering the skyrmion lattice phase, to a commensurate value, and remains flat as long as the SkL is stable. This type of study is enabled by the flexible sample environment of BL19LXU, where various magnets, a four-circle diffractometer, etc. can be installed at-will to support specific and promising scattering experiments.

The spin at the core of a skyrmion points parallel to the magnetic field, and the corresponding magnetic moment – which is antiparallel to the spin – is opposed to the magnetic field. Therefore, the core spin of a skyrmion is generally not favored by the Zeeman energy, but still stable when an SkL is incommensurate to the crystal lattice, due to the exchange energy. We studied the commensurate SkL of $Gd_3Ru_4Al_{12}$ by measurements of the single-ion anisotropy via electron spin resonance, combined with model calculations. The analysis shows that the commensurate SkL in $Gd_3Ru_4Al_{12}$ likely arranges itself on the lattice in such a way that the core spin is located on a "hole" of the lattice, due to the magneto-crystalline anisotropy.

Among materials with twisted spin textures, so-called 'noncoplanar magnets', the commensurate SkL in $Gd_3Ru_4Al_{12}$ represents a link between short-period, commensurate antiferromagnets – on the left side in Fig. 2 – and longer-period, incommensurate skyrmion crystal lattices (right side). On the ordinate, this plot illustrates the largest solid angle that remains uncovered by arrows when the magnetic structure is projected onto a sphere, as in Fig. 1(a). Thus, the observation of a commensurate SkL represents an essential link between various classes of magnetic materials, with many opportunities for future discoveries: For example, studies of the magnetic excitation spectrum, which likely consists of coupled phonon and spin wave modes under the influence of Berry's quantum phase [5].

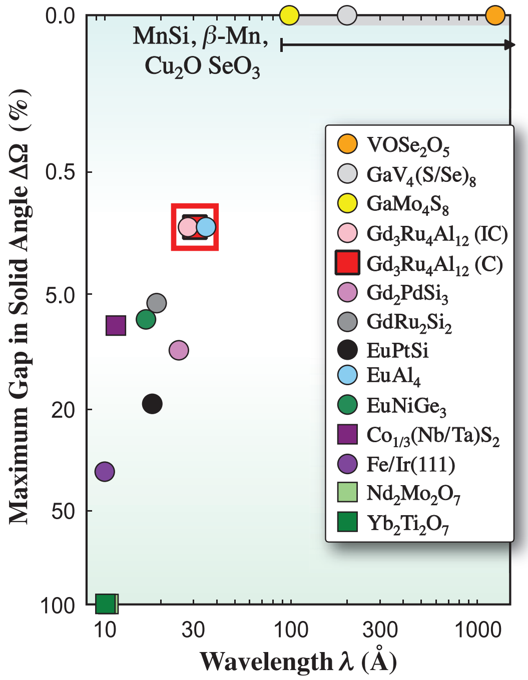

Fig. 2. Relationship of magnetic texture dimensions (λ) and coverage of directions on the sphere for various materials with noncoplanar textures and spin chirality. At the center of the plot, the commensurate C-SkL state in $Gd_3Ru_4Al_{12}$ is highlighted by a red box. On the *y*-axis, a continuous magnetic texture has zero uncovered solid angle, i.e., we assign a value of 0%. Note the stretched scale of the *y*-axis.

Max Hirschberger[a,b,*], Yoshikazu Tanaka[c] and Hiroyuki Ohsumi[c]

[a] Department of Applied Physics and Quantum-Phase Electronics Center, The University of Tokyo
[b] RIKEN Center for Emergent Matter Science (CEMS)/Wako
[c] RIKEN SPring-8 Center

*Email: hirschberger@ap.t.u-tokyo.ac.jp

References

[1] T. Okubo *et al.*: Phys. Rev. Lett. **108** (2012) 017206.
[2] S. Hayami *et al.*: Phys. Rev. B **95** (2017) 224424.
[3] S. Hayami and R. Yambe: Phys. Rev. Res. **3** (2021) 043158.
[4] *M. Hirschberger, B. G. Szigeti, M. Hemmida, M. M. Hirschmann, S. Esser, H. Ohsumi, Y. Tanaka, L. Spitz, S. Gao, K. K. Kolincio, H. Sagayama, H. Nakao, Y. Yamasaki, L. Forró, H.-A. K. von Nidda, I. Kezsmarki, T. Arima and Y. Tokura: npj Quantum Mater.* ***9*** *(2024) 45.*
[5] T. Weber *et al.*: Science **375** (2022) 1025.