Mean-field analysis of a square skyrmion lattice in multi-orbital *f*-electron systems

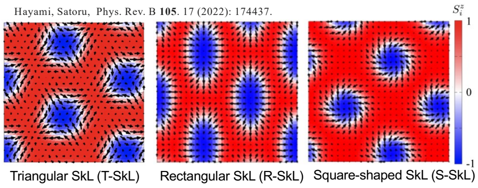
北海道大学
HOKKAIDO UNIVERSITY

Yan Zha (查言)[1], Satoru Hayami[1]
[1]*Graduate School of Science, Hokkaido University, Sapporo 060-0810, Japan*

Research Background

・Skyrmion lattice (SkL)

Hayami, Satoru, J. Magn. Magn. Mater, 553 (2022): 169220.
Hayami, Satoru, Phys. Rev. B **105**. 17 (2022): 174437.



Triangular SkL (T-SkL) Rectangular SkL (R-SkL) Square-shaped SkL (S-SkL)

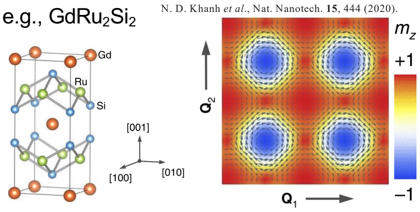
・ Merits

- ✓ A highly ordered structure with higher stability and orderliness as a thermodynamic phase.
- ✓ Potential candidates for next-generation recording media.

・Experimental Observation of the S-SkL

e.g., $GdRu_2Si_2$

N. D. Khanh *et al*., Nat. Nanotech. **15**, 444 (2020).



・Interactions provide Spiral

System without centrosymmetry

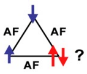
$$\mathcal{H}_{\rm DM} = \boldsymbol{D}\cdot(\boldsymbol{S}_1\times\boldsymbol{S}_2)$$

$$\boldsymbol{D}\cdot(\boldsymbol{S}_1\times\boldsymbol{S}_2) = \boldsymbol{D}\cdot(\boldsymbol{S}_2\times\boldsymbol{S}_1) = -\boldsymbol{D}\cdot(\boldsymbol{S}_1\times\boldsymbol{S}_2)$$

System with centrosymmetry

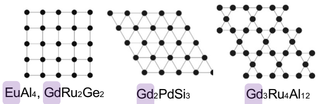
$$\sum_{\langle i,j\rangle}\mathcal{J}_{ij}\boldsymbol{S}_i\cdot\boldsymbol{S}_j$$

Frustration
e.g., J_1=1, J_2=-0.5, J_3=-0.25 on a square lattice



Geometrical frustration

・Existing research ($\boldsymbol{L}$=0)



Q: Why a square lattice?

Hayami, Satoru, Phys. Rev. B **105**. 17 (2022): 174437.

A: $2Q$ S-SkL needs additional magnetic anisotropy. (Energetic viewpoint)

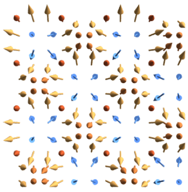
→ **Crystal filed splitting**

Aim

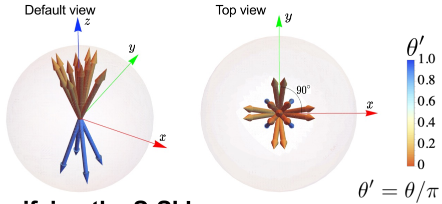
・ Explore whether the S-SkLs are possible in other $4f$-electron compounds with the **orbital angular momentum**.

S-SkL

・S-SkL's Configuration

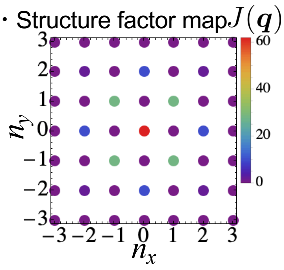


・S-SkL plotted on a unit sphere

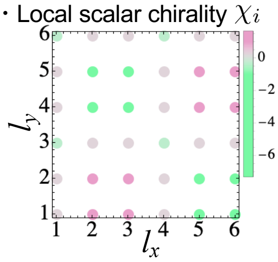


Physical quantities for classifying the S-SkL

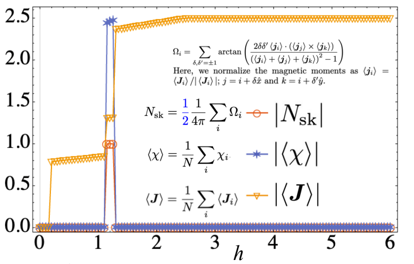
・Structure factor map $J(\boldsymbol{q})$



・Local scalar chirality χ_i



・Absolute topological skyrmion number $|N_{\rm sk}|$
Absolute scalar chirality $|\langle\chi\rangle|$
Magnetization magnitude $|\langle\boldsymbol{J}\rangle|$ at Δ= 0



$$\Omega_i = \sum_{\delta,\delta'=\pm1}\arctan\left(\frac{2\delta\delta'\langle\boldsymbol{j}_i\rangle\cdot(\langle\boldsymbol{j}_j\rangle\times\langle\boldsymbol{j}_k\rangle)}{(\langle\boldsymbol{j}_i\rangle+\langle\boldsymbol{j}_j\rangle+\langle\boldsymbol{j}_k\rangle)^2-1}\right)$$

Here, we normalize the magnetic moments as $\langle\boldsymbol{j}_i\rangle = \langle\boldsymbol{J}_i\rangle/|\langle\boldsymbol{J}_i\rangle|$; $j=i+\delta\hat{x}$ and $k=i+\delta'\hat{y}$.

$$N_{\rm sk} = \frac{1}{2}\frac{1}{4\pi}\sum_i\Omega_i$$

$$\langle\chi\rangle = \frac{1}{N}\sum_i\chi_i$$

$$\langle\boldsymbol{J}\rangle = \frac{1}{N}\sum_i\langle\boldsymbol{J}_i\rangle$$

$$J(\boldsymbol{q}) = \frac{1}{N}\sum_{i,j}\langle\boldsymbol{J}_i\rangle\cdot\langle\boldsymbol{J}_j\rangle\exp\left[i\,\boldsymbol{q}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)\right]$$

$$\chi_i = \frac{1}{2}\sum_{\delta,\delta'=\pm1}\delta\delta'\langle\boldsymbol{J}_i\rangle\cdot(\langle\boldsymbol{J}_{i+\delta\hat{x}}\rangle\times\langle\boldsymbol{J}_{i+\delta'\hat{y}}\rangle)$$

・The S-SkL possesses **fourfold rotational symmetry**.

・ The **strong easy-axis magnetic anisotropy** tends to stabilize the S-SkL in a centrosymmetric system.

Model

• Frustrated classical Heisenberg model on a square lattice

N. Iwahara and L. F. Chibotaru, Phys. Rev. B **91**, 174438 (2015).

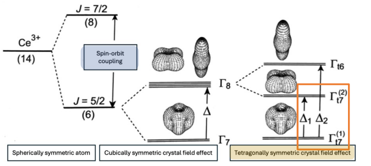
$$\mathcal{H}_{\rm tot} = \mathcal{H}_{\rm ex} + \mathcal{H}_\Delta + \mathcal{H}_Z,$$

$$\mathcal{H}_{\rm ex} = -\sum_{\langle i,j\rangle}\mathcal{J}_{ij}\boldsymbol{J}_i\cdot\boldsymbol{J}_j,$$ exchange interactions

$$\mathcal{H}_\Delta = \begin{pmatrix}0&0&0&0\\0&0&0&0\\0&0&\Delta&0\\0&0&0&\Delta\end{pmatrix},$$ crystal field splitting

$$\mathcal{H}_Z = -h\sum_i J_i^z,$$ Zeeman coupling

• Orbital splitting in a Ce^{3+} ion



Heavy Fermion Physics: Magnetism and Superconductivity, Nariaki Sato *et al.*

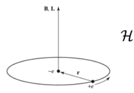
$$\begin{cases}|\Gamma_{t6\pm}\rangle = |\tfrac{5}{2},\pm\tfrac{1}{2}\rangle,\\ \left|\Gamma^{(1)}_{t7\pm}\right\rangle = \alpha|\tfrac{5}{2},\pm\tfrac{5}{2}\rangle - \beta|\tfrac{5}{2},\mp\tfrac{3}{2}\rangle,\\ \left|\Gamma^{(2)}_{t7\pm}\right\rangle = \beta|\tfrac{5}{2},\pm\tfrac{5}{2}\rangle + \alpha|\tfrac{5}{2},\mp\tfrac{3}{2}\rangle,\end{cases}$$

$$\alpha = \frac{2\sqrt{5}}{\sqrt{\frac{\left(\sqrt{X^2+20(B_4^4)^2}+X\right)^2}{(B_4^4)^2}+20}},\quad X = (B_2^0+20\,B_4^0),\quad \beta^2 = 1-\alpha^2,$$

Kramers doublets

• Spin-orbit coupling

Griffiths, David J *et al.*, *Introduction to quantum mechanics*. Cambridge university press, 2019.



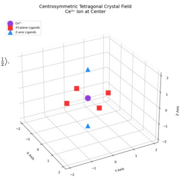
$$\mathcal{H}_{LS} = \lambda\boldsymbol{L}\cdot\boldsymbol{S}$$

we obtain a 14 × 14 sparse matrix $\mathcal{H}_{LS}$ naturally, in the ordered basis from $|3,3\rangle\otimes|\frac{1}{2},\pm\frac{1}{2}\rangle$ down to $|3,-3\rangle\otimes|\frac{1}{2},\pm\frac{1}{2}\rangle$.

$$\mathcal{H}_{LS} = \begin{pmatrix}
\frac{3\lambda}{2}&0&0&0&0&0&0&0&0&0&0&0&0&0\\
0&-\frac{3\lambda}{2}&\sqrt{\frac{3}{2}}\lambda&0&0&0&0&0&0&0&0&0&0&0\\
0&\sqrt{\frac{3}{2}}\lambda&\lambda&0&0&0&0&0&0&0&0&0&0&0\\
0&0&0&-\lambda&\sqrt{\frac{5}{2}}\lambda&0&0&0&0&0&0&0&0&0\\
0&0&0&\sqrt{\frac{5}{2}}\lambda&\frac{\lambda}{2}&0&0&0&0&0&0&0&0&0\\
0&0&0&0&0&-\frac{\lambda}{2}&\sqrt{3}\lambda&0&0&0&0&0&0&0\\
0&0&0&0&0&\sqrt{3}\lambda&0&0&0&0&0&0&0&0\\
0&0&0&0&0&0&0&0&\sqrt{3}\lambda&0&0&0&0&0\\
0&0&0&0&0&0&0&\sqrt{3}\lambda&-\frac{\lambda}{2}&0&0&0&0&0\\
0&0&0&0&0&0&0&0&0&\frac{\lambda}{2}&\sqrt{\frac{5}{2}}\lambda&0&0&0\\
0&0&0&0&0&0&0&0&0&\sqrt{\frac{5}{2}}\lambda&-\lambda&0&0&0\\
0&0&0&0&0&0&0&0&0&0&0&\lambda&\sqrt{\frac{3}{2}}\lambda&0\\
0&0&0&0&0&0&0&0&0&0&0&\sqrt{\frac{3}{2}}\lambda&-\frac{3\lambda}{2}&0\\
0&0&0&0&0&0&0&0&0&0&0&0&0&\frac{3\lambda}{2}
\end{pmatrix}$$

$J=\frac{5}{2}$ basis

• Tetragonal crystalline electric field effect



$$\mathcal{H}_{\rm cry} = B_2^0O_2^0 + B_4^0O_4^0 + B_4^4O_4^4$$

$$O_2^0 = 3J_z^2 - \boldsymbol{J}^2,$$
$$O_4^0 = 35J_z^4 - 30\boldsymbol{J}^2J_z^2 + 25J_z^2 - 6\boldsymbol{J}^2 + 3\boldsymbol{J}^4,$$
$$O_4^4 = \frac{1}{2}\left[(J^+)^4 + (J^-)^4\right].$$

Stevens operator

K. Stevens, Proceedings of the Physical Society. Section A **65**, 209 (1952).

$$\langle\mathcal{H}_{\rm cry}\rangle = \begin{pmatrix}
10(B_2^0+6B_4^0)&0&0&0&12\sqrt{5}B_4^4&0\\
0&-2(B_2^0+90B_4^0)&0&0&0&12\sqrt{5}B_4^4\\
0&0&-8(B_2^0-15B_4^0)&0&0&0\\
0&0&0&-8(B_2^0-15B_4^0)&0&0\\
12\sqrt{5}B_4^4&0&0&0&-2(B_2^0+90B_4^0)&0\\
0&12\sqrt{5}B_4^4&0&0&0&10(B_2^0+6B_4^0)
\end{pmatrix}$$

• Full angular momentum operator in 4×4 Hilbert space (e.g., J^z)

-----------------------------<Jz> 4x4-----------------------------

	$\left\lvert\Gamma^{(1)}_{t7}+\right\rangle$	$\left\lvert\Gamma^{(1)}_{t7}-\right\rangle$	$\left\lvert\Gamma^{(2)}_{t7}+\right\rangle$	$\left\lvert\Gamma^{(2)}_{t7}-\right\rangle$
$\left\langle\Gamma^{(1)}_{t7}+\right\rvert$	$\frac{1}{2}\times(5\alpha^2-3\beta^2)$	0	$4\alpha\beta$	0
$\left\langle\Gamma^{(1)}_{t7}-\right\rvert$	0	$\frac{1}{2}\times(-5\alpha^2+3\beta^2)$	0	$-4\alpha\beta$
$\left\langle\Gamma^{(2)}_{t7}+\right\rvert$	$4\alpha\beta$	0	$\frac{1}{2}\times(-3\alpha^2+5\beta^2)$	0
$\left\langle\Gamma^{(2)}_{t7}-\right\rvert$	0	$-4\alpha\beta$	0	$\frac{1}{2}\times(3\alpha^2-5\beta^2)$

• Mean-field calculations

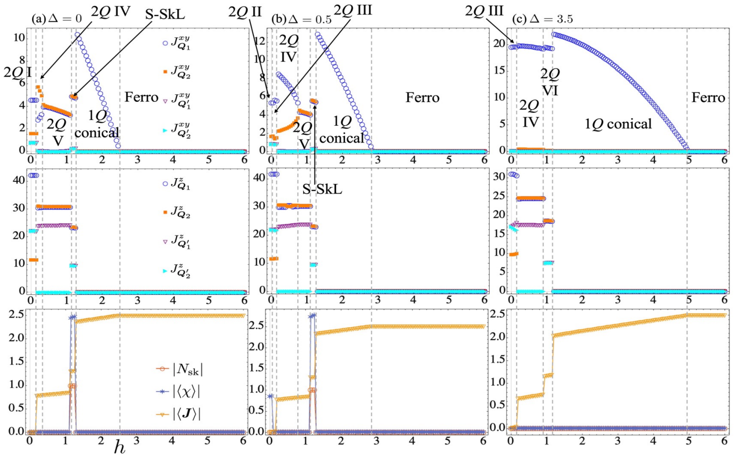
$$\mathcal{H}_{\rm ex}^{\rm MF} = -\sum_{\langle i,j\rangle}\mathcal{J}_{ij}\left(\boldsymbol{J}_i\cdot\langle\boldsymbol{J}_j\rangle + \boldsymbol{J}_j\cdot\langle\boldsymbol{J}_i\rangle - \langle\boldsymbol{J}_i\rangle\cdot\langle\boldsymbol{J}_j\rangle\right)$$

operator
mean magnetic moment at the *i*-th site
constant term

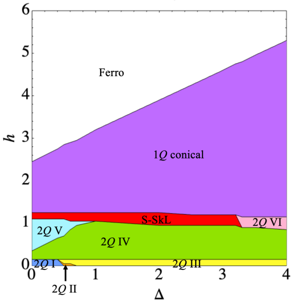
$$\langle\mathcal{O}\rangle = \frac{1}{Z}\sum_n\exp(-\epsilon_n/T)\langle n|\mathcal{O}|n\rangle$$

$$Z = \sum_n\exp(-\epsilon_n/T)$$

Phase Classification



・Δ-h Phase diagram



・Nonzero components of $J_{\boldsymbol{Q}_\eta}$ and $J_{\boldsymbol{Q}'_\eta}$ $(\eta = 1,2)$ in each phase.

phase	$J_{\boldsymbol{Q}'_1}$, $J_{\boldsymbol{Q}'_2}$ ($\boldsymbol{Q}'_\eta \parallel [100]$)	$J_{\boldsymbol{Q}_1}$, $J_{\boldsymbol{Q}_2}$ ($\boldsymbol{Q}_\eta \parallel [110]$)
S-SkL	$J^{xy}_{\boldsymbol{Q}'_1} = J^{xy}_{\boldsymbol{Q}'_2}$, $J^{z}_{\boldsymbol{Q}'_1} = J^{z}_{\boldsymbol{Q}'_2}$	$J^{xy}_{\boldsymbol{Q}_1} = J^{xy}_{\boldsymbol{Q}_2}$, $J^{z}_{\boldsymbol{Q}_1} = J^{z}_{\boldsymbol{Q}_2}$
$1Q$ conical	–	$S^{xy}_{\boldsymbol{Q}_1}$
$2Q$ I	$J^{xy}_{\boldsymbol{Q}'_1} = J^{xy}_{\boldsymbol{Q}'_2}$, $J^{z}_{\boldsymbol{Q}'_1} = J^{z}_{\boldsymbol{Q}'_2}$	$J^{xy}_{\boldsymbol{Q}_1}$, $J^{xy}_{\boldsymbol{Q}_2}$, $J^{z}_{\boldsymbol{Q}_1}$, $J^{z}_{\boldsymbol{Q}_2}$
$2Q$ II	$J^{xy}_{\boldsymbol{Q}'_1} = J^{xy}_{\boldsymbol{Q}'_2}$, $J^{z}_{\boldsymbol{Q}'_1} = J^{z}_{\boldsymbol{Q}'_2}$	$J^{xy}_{\boldsymbol{Q}_1}$, $J^{xy}_{\boldsymbol{Q}_2}$, $J^{z}_{\boldsymbol{Q}_1}$, $J^{z}_{\boldsymbol{Q}_2}$
$2Q$ III	$J^{xy}_{\boldsymbol{Q}'_1} = J^{xy}_{\boldsymbol{Q}'_2}$, $J^{z}_{\boldsymbol{Q}'_1}$, $J^{z}_{\boldsymbol{Q}'_2}$	$J^{xy}_{\boldsymbol{Q}_1}$, $J^{xy}_{\boldsymbol{Q}_2}$, $J^{z}_{\boldsymbol{Q}_1}$, $J^{z}_{\boldsymbol{Q}_2}$
$2Q$ IV	$J^{z}_{\boldsymbol{Q}'_1}$	$J^{xy}_{\boldsymbol{Q}_1}$, $J^{xy}_{\boldsymbol{Q}_2}$, $J^{z}_{\boldsymbol{Q}_1} = J^{z}_{\boldsymbol{Q}_2}$
$2Q$ V	$J^{z}_{\boldsymbol{Q}'_1}$	$J^{xy}_{\boldsymbol{Q}_1} = J^{xy}_{\boldsymbol{Q}_2}$, $J^{z}_{\boldsymbol{Q}_1} = J^{z}_{\boldsymbol{Q}_2}$
$2Q$ VI	$J^{z}_{\boldsymbol{Q}'_1} = J^{z}_{\boldsymbol{Q}'_2}$	$J^{xy}_{\boldsymbol{Q}_1}$, $J^{z}_{\boldsymbol{Q}_1} = J^{z}_{\boldsymbol{Q}_2}$

・Except for the **S-SkL** state, we also classify one **$1Q$ conical** state and six types of $2Q$ states.

Summary

・We have investigated the emergence of the S-SkL on a centrosymmetric square lattice by employing **mean-field calculations** with an emphasis on the **multi-orbital degree of freedom**.

・We derived the total angular momentum in a 4×4 Hilbert space using Kramers' doublets from **crystal field effects**.

・We systematically introduced several physical quantities for classifying the S-SkL and other multi-Q states.

・Our study reveals the possibility of stabilizing S-SkLs in $4f$-electron systems with a **finite orbital angular momentum**.