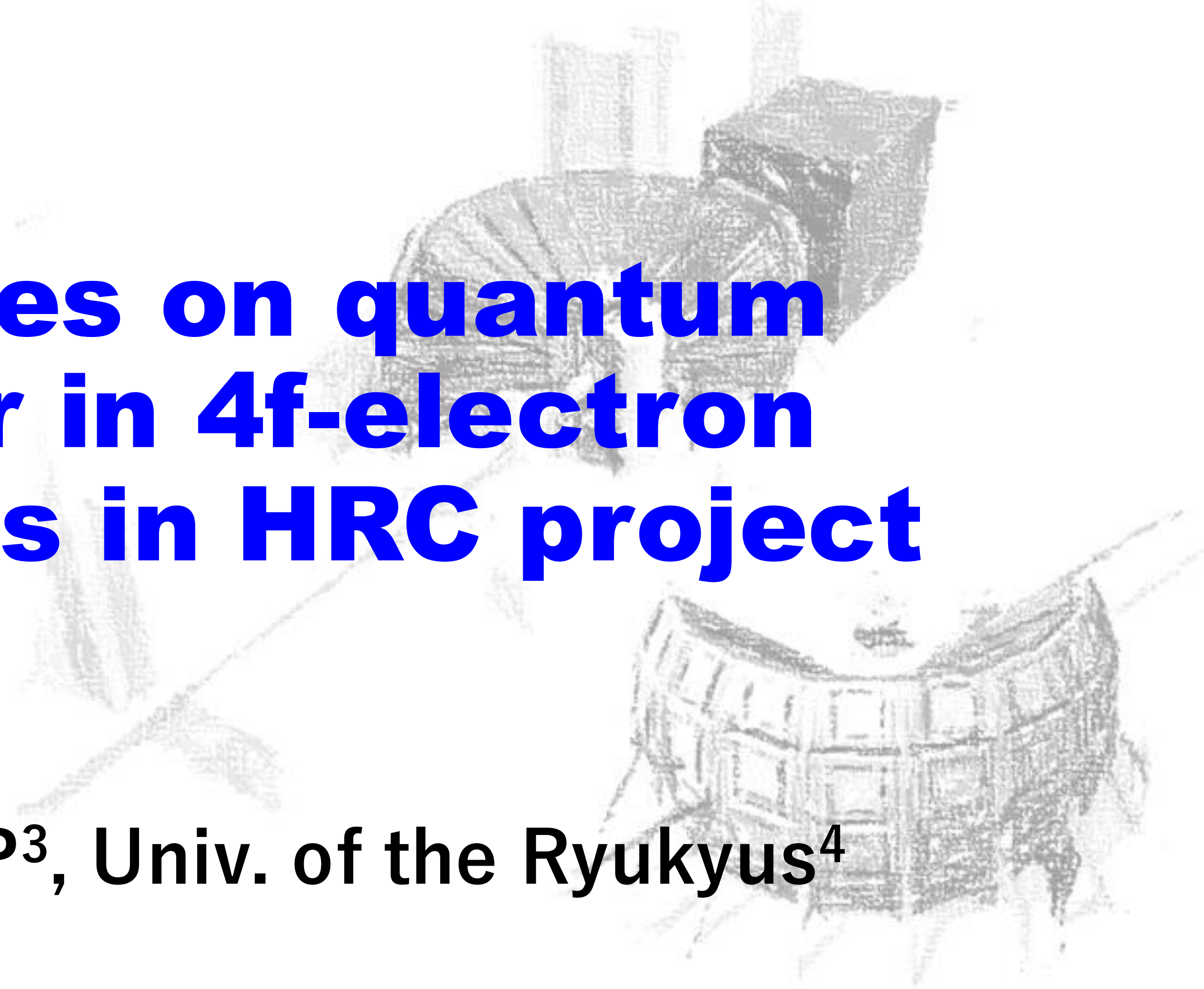
Dynamical studies on quantum critical behavior in 4f-electron frustrated systems in HRC project



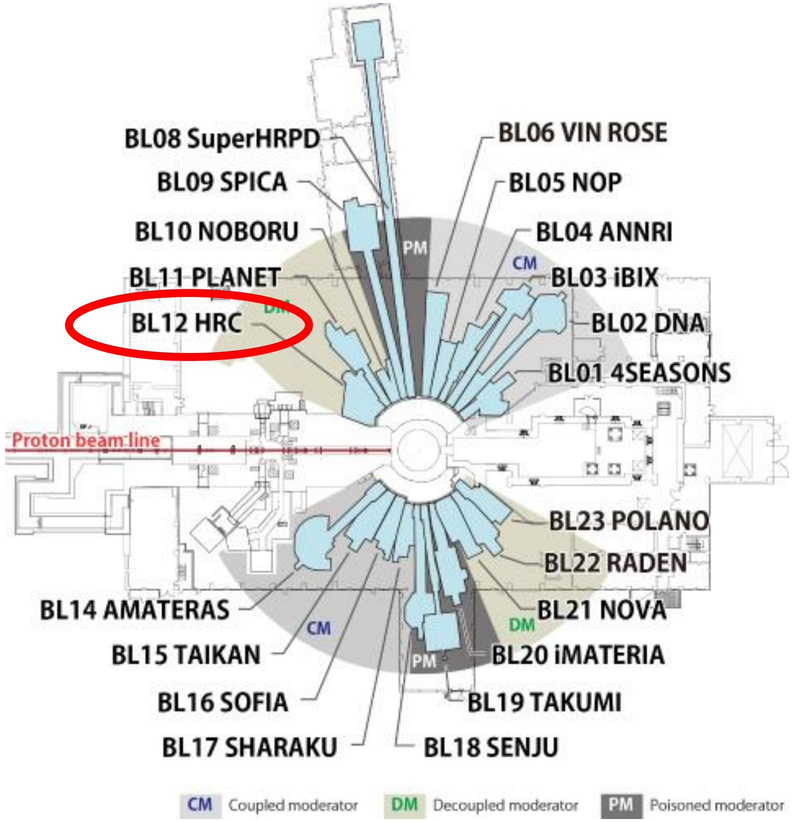
IMSS, KEK[1], J-PARC[2], ISSP[3], Univ. of the Ryukyus[4]

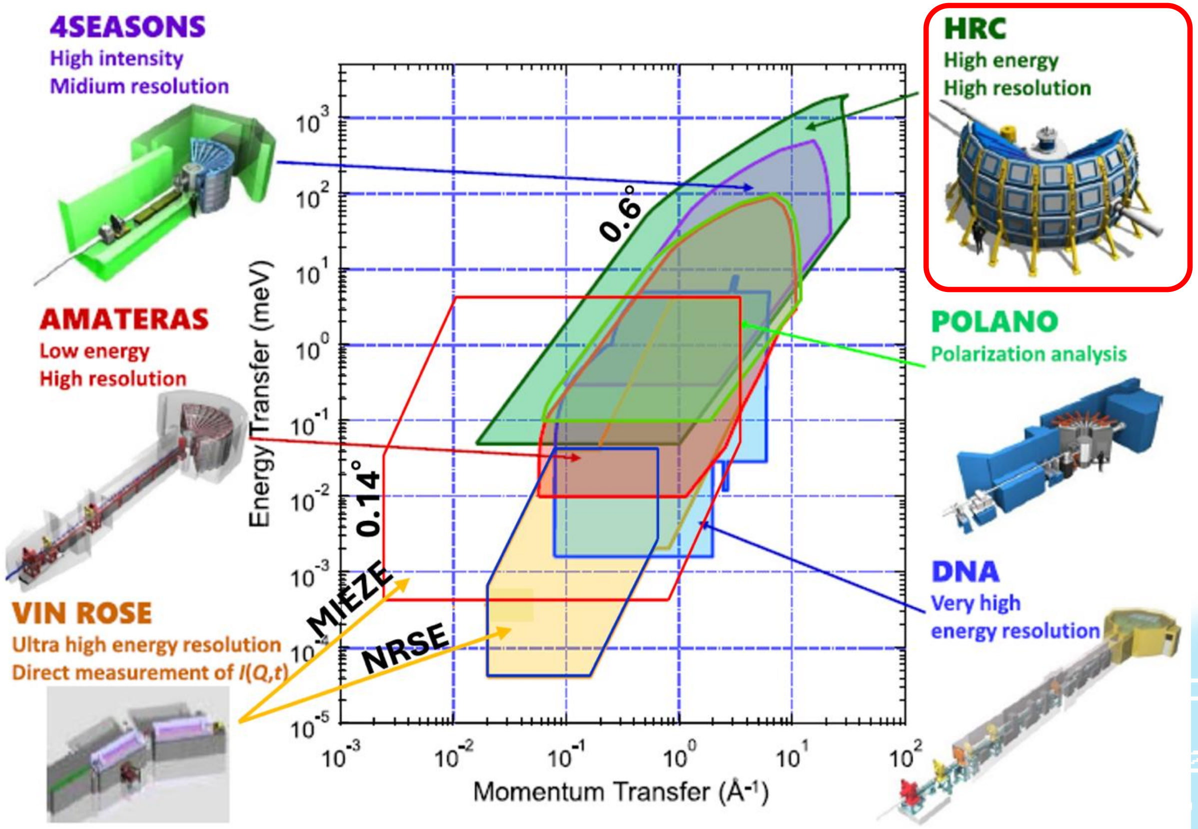
Daichi Ueta[A1 2], Takatsugu Masuda[3, 1], Taro Nakajima[3],
Tetsuya Yokoo[1, 2], Riki Kobayashi[4], Shinichi Itoh[1, 2]

High Resolution Chopper Spectrometer

高分解能チョッパー分光器
東京大学・高エネルギー加速器研究機構
High Resolution Chopper Spectrometer (HRC)
High Energy Accelerator Research Organization and The University of Tokyo

Inelastic neutron scattering instrument for probing the dynamics of materials at high resolution and over a wide range.





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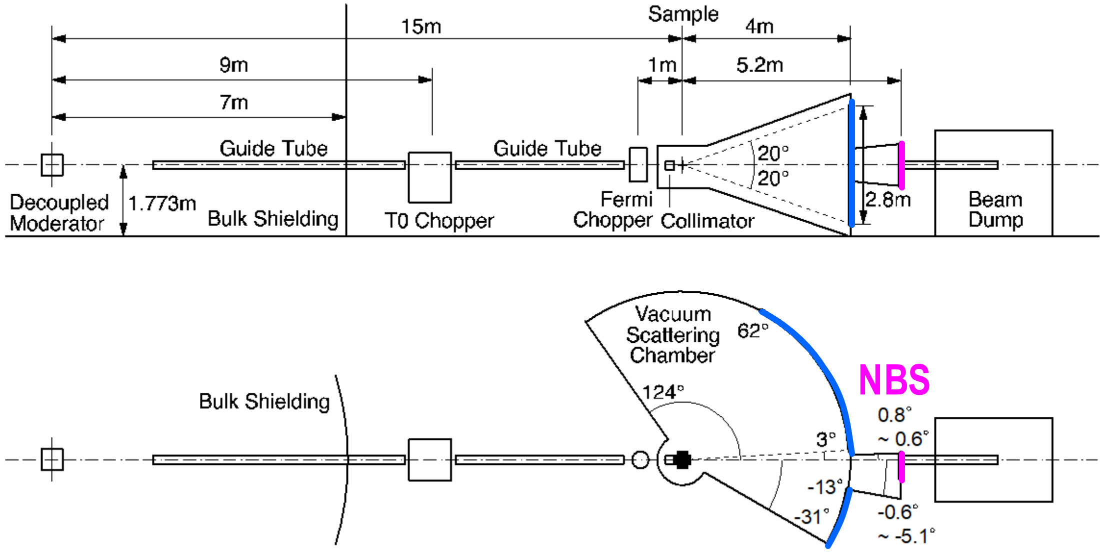
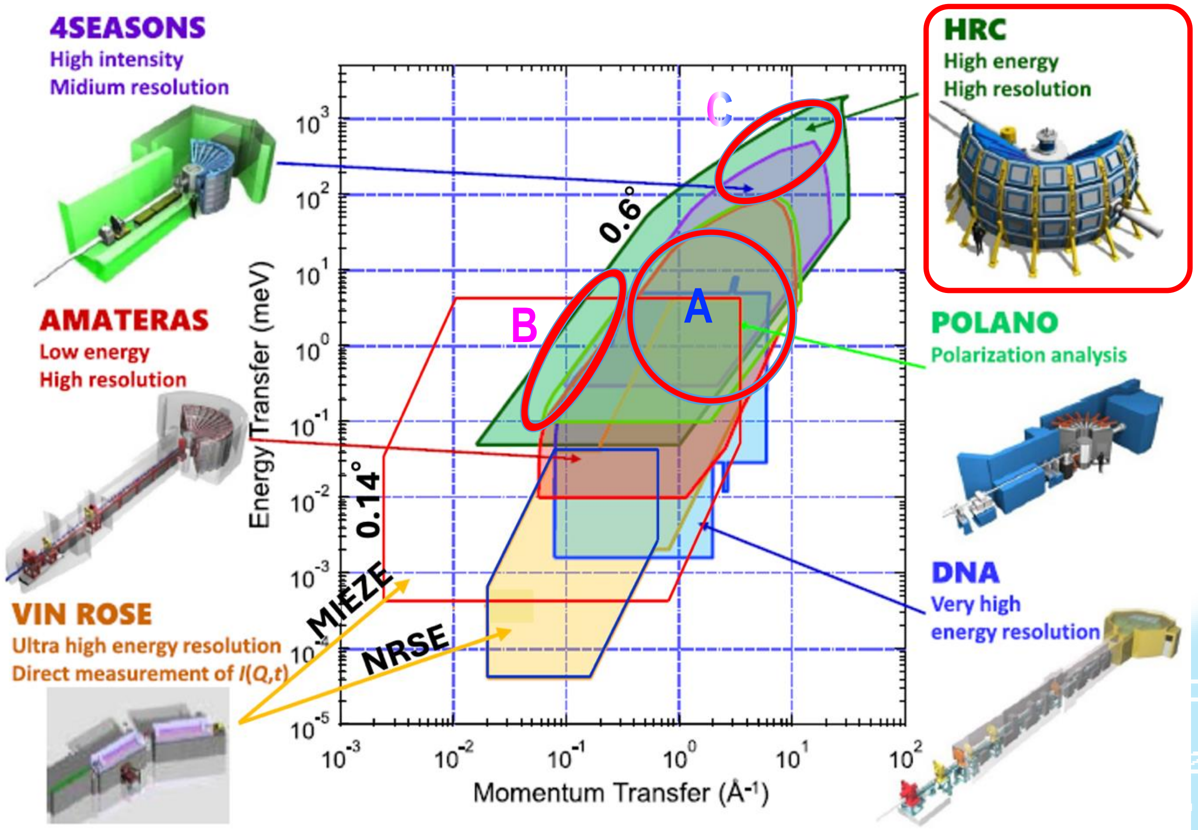


A: high-resolution inelastic experiments in conventional Q-E space.

B: accessible small-Q and high-E range.

C: sub-eV neutron spectroscopy.



S. Itoh et al., Physica B: Condensed Matter 568 76 (2019).

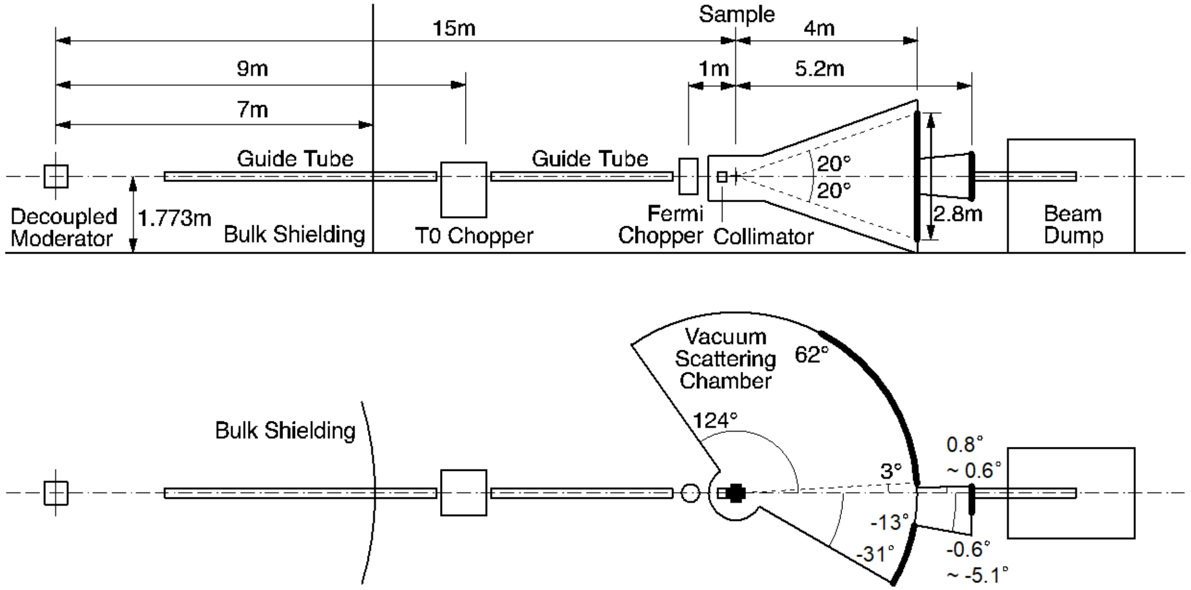
HRC Project

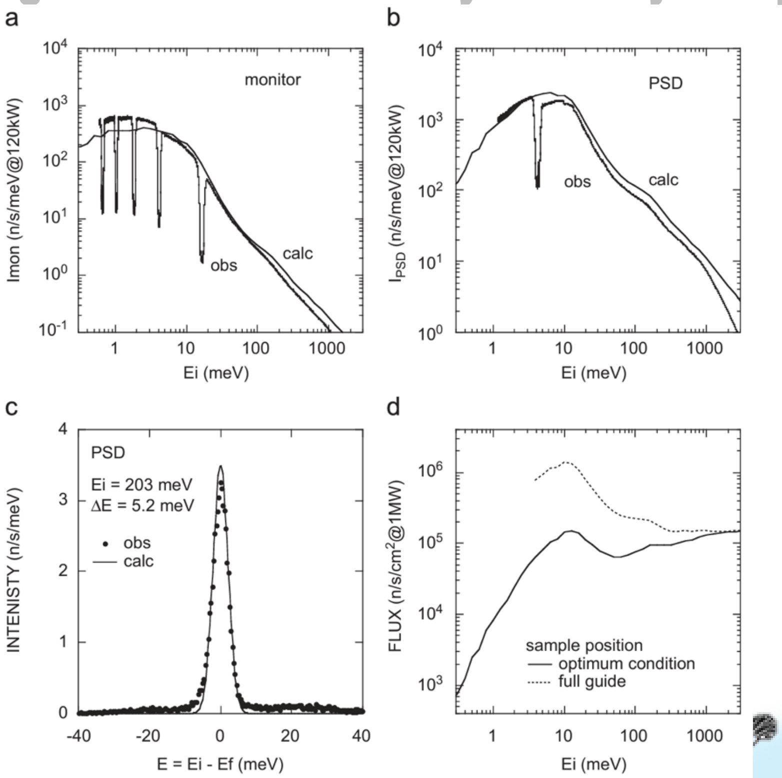
First stage (2008 - 2013): Construction of HRC and evaluation of its performance

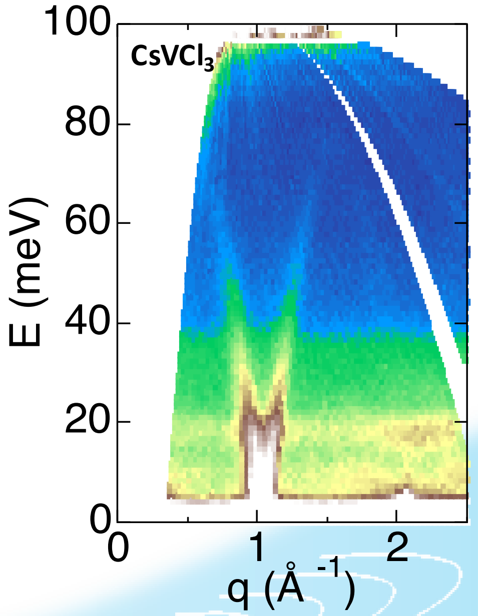
Second stage (2014 - 2018): Advancement of neutron Brillouin scattering with small scattering angle

Third stage (2019 - 2023): Expansion of external environment

Fourth stage (2024 -): Promoting the study of topological and frustrated systems by complex environments







S. Itoh et al., Physica B: Condensed Matter 568 76 (2019).
S. Itoh et al., Nucl. Instrum. Methods Phys. Res., Sect. A 631 90 (2011).
S. Itoh et al., JPSJ 81 084706 (2012).

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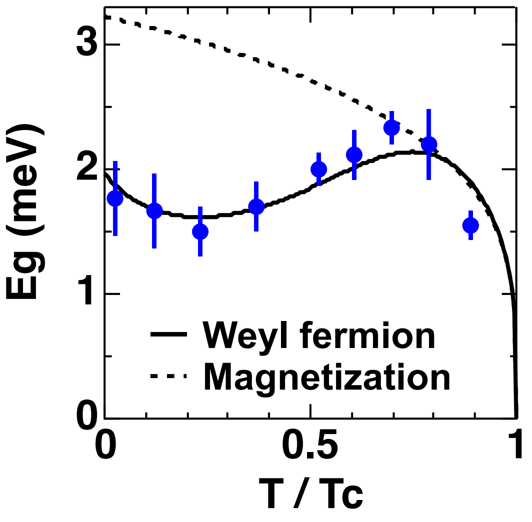
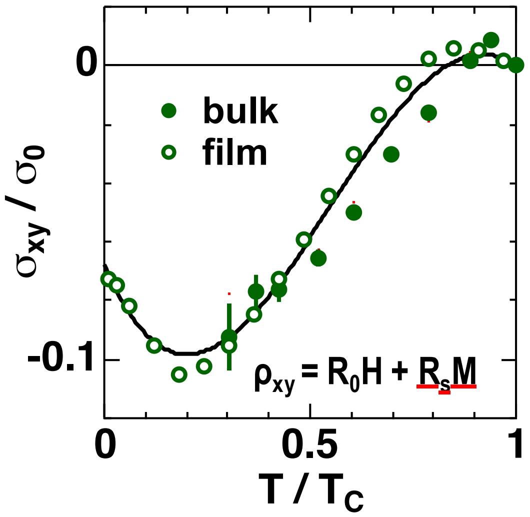
$SrRuO_3$

S. Itoh et al., Nat. Comm. 7, 11788 (2016).

Ferromagnetic powder sample





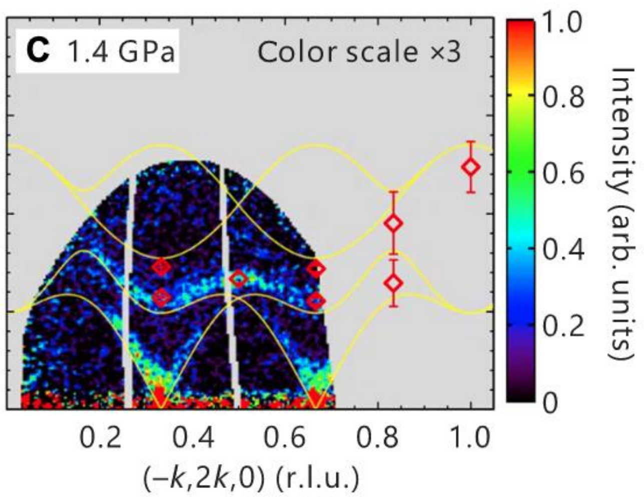
$E=Dq^2+E_g$

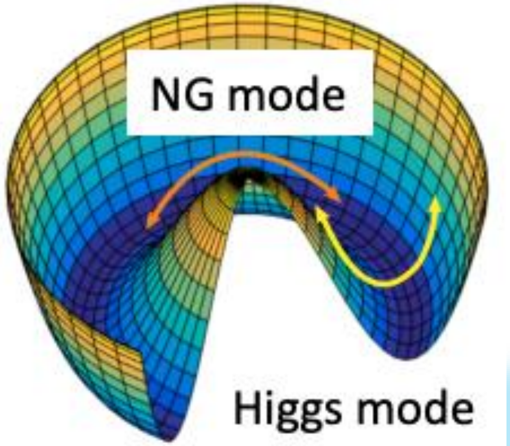
$$E_g(T)=\frac{a\left(M(T)/M_0\right)}{1+b\left(M(T)/M_0\right)\left(s_{xy}(T)/s_0\right)}$$

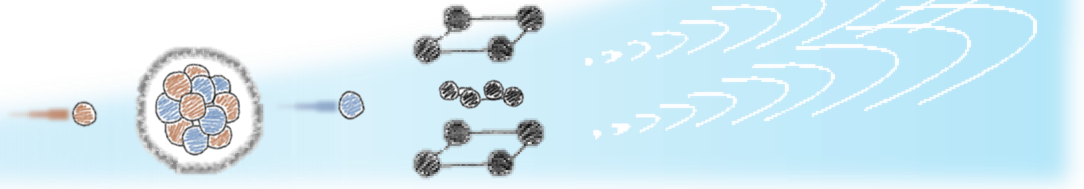
$CsFeCl_3$

S. Hayashida, et al., Sci. Adv. 5, eaaw5639 (2019).

Neutron experiments under pressure







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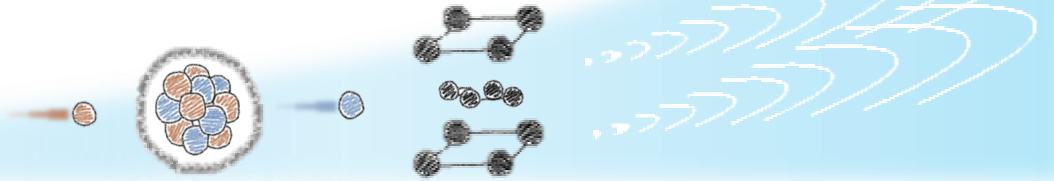
Sample environment	Control range
GM refrigerator	T = 4 – 300 K
1K refrigerator	T = 0.6 – 300 K
Superconducting magnet	H ≤ 5 T, T = 0.3 – 300 K
Cryofurnace	T = 4 – 700 K
^{3}He sorption-type ref.	T = 0.3 – 300 K (100 h)
Cylindrical pressure cell	P ≤ 1.2 GPa

- **High temperature measurements of stoner excitation FeMn**
- **Low temperature measurements of spin dimer excitation Ce_5Si_3**
- **High energy neutron diffraction measurements of magnetic skyrmion $GdRu_2Ge_2$**
- **Field control of quasiparticle decay in $RbFeCl_3$**



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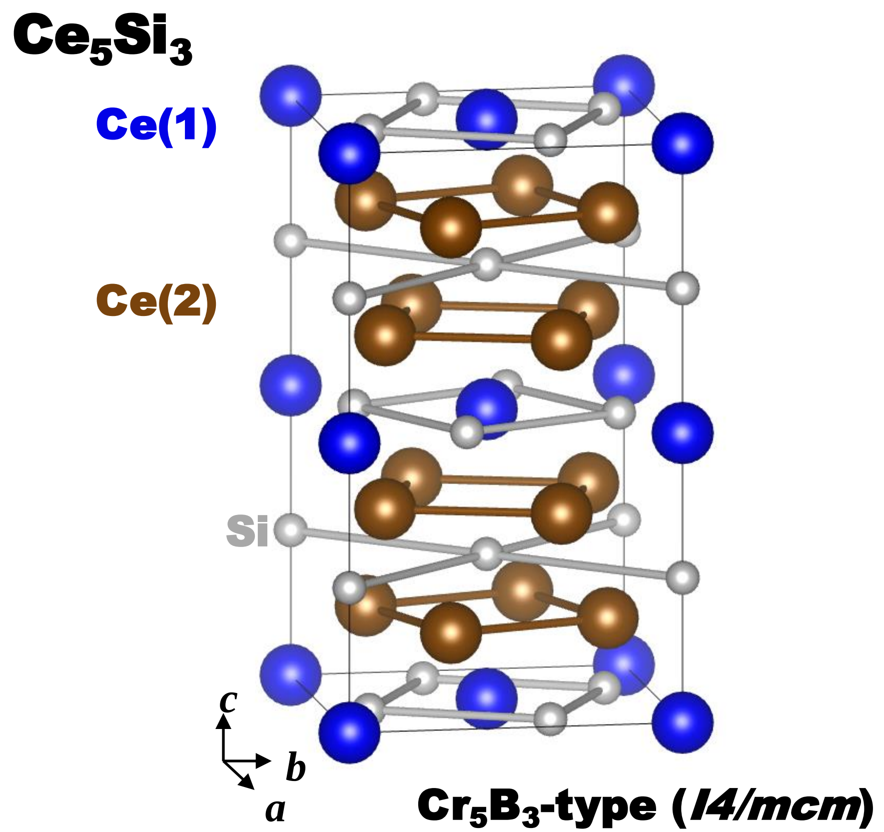
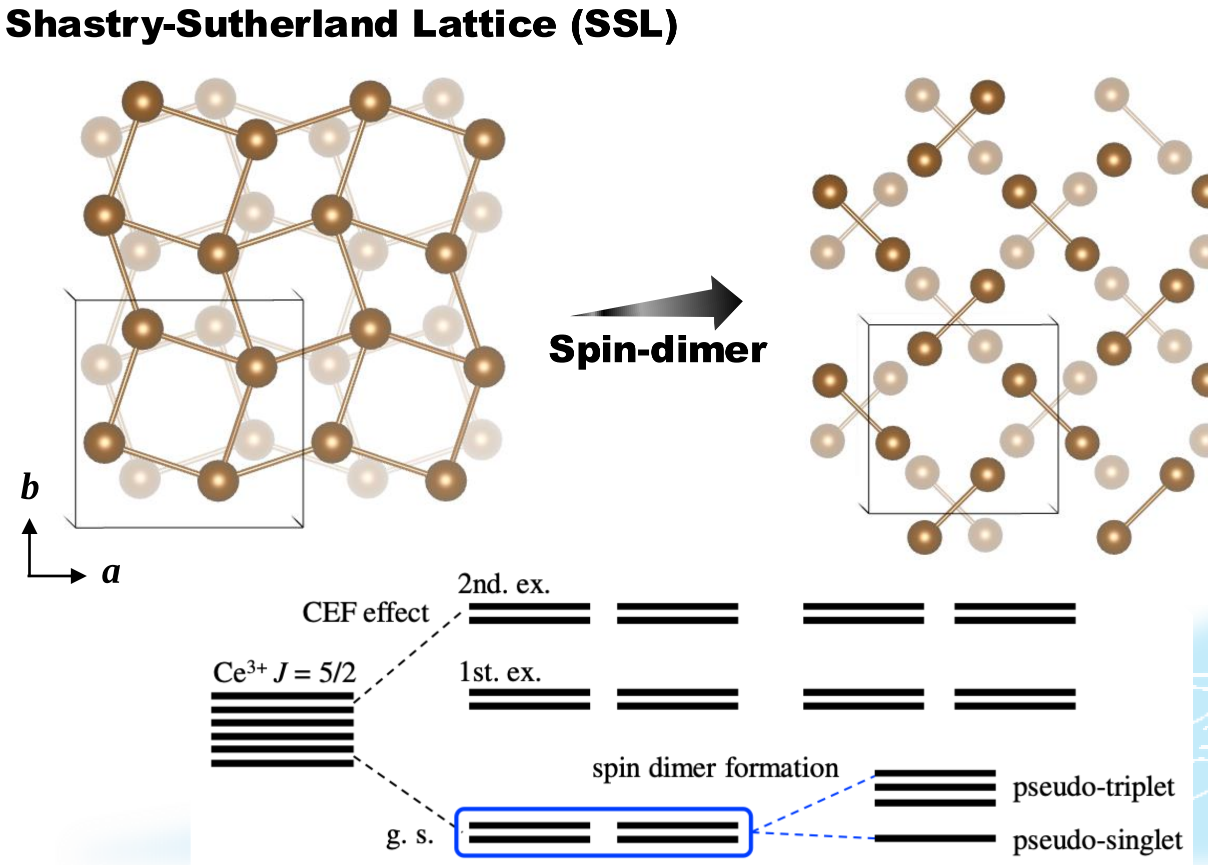




Characteristics of Ce_5Si_3

$SrCu_2(BO_3)_2$

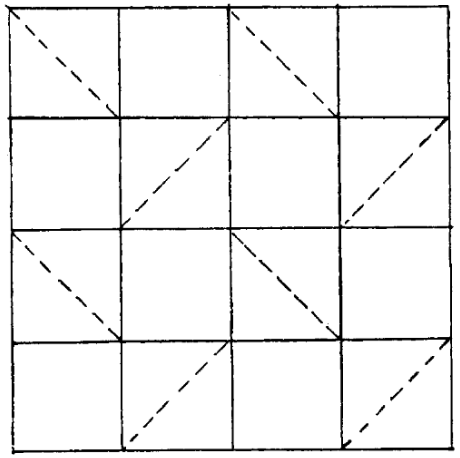
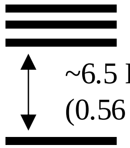
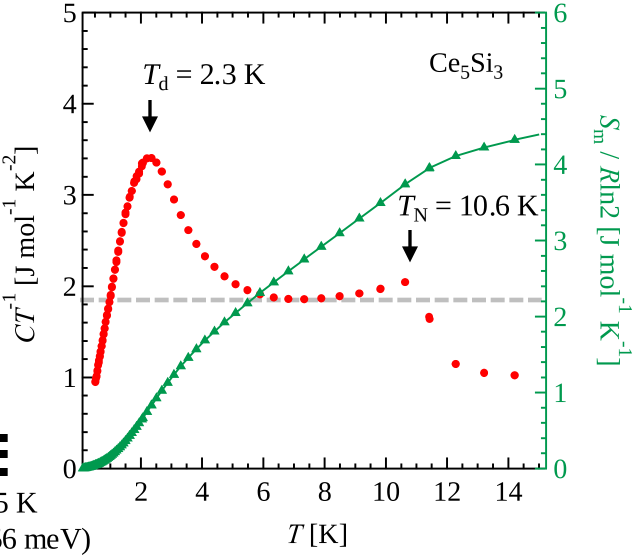
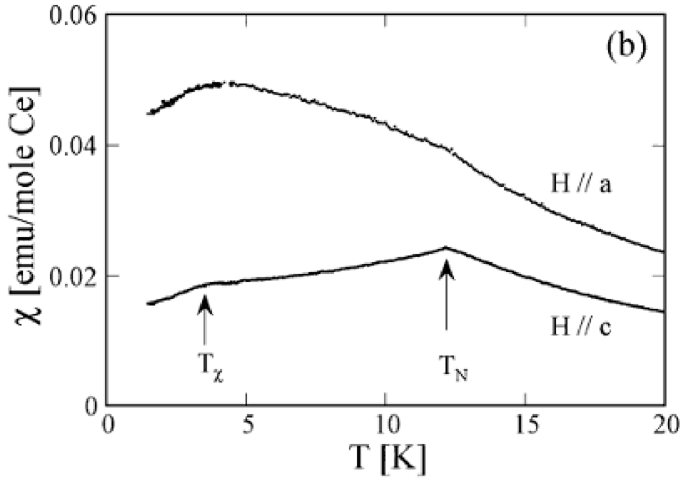


Fig. 1. The Lattice

1.B. S. Shastry and B. Sutherland, Physica (Amsterdam) **108B**, 1069 (1981).







Y. Ushida, et al., JPSJ, 70 (2001) 513.

- Anomalies caused by magnetic transition around 10.6 K.

➔ Magnetic entropy indicates that the Ce(1) site is magnetically ordered.

- The bending of the magnetic susceptibility when a magnetic field is applied in the interplanar direction is remarkable.

➔ Suggests that the magnetic moment is oriented in the interplane direction.

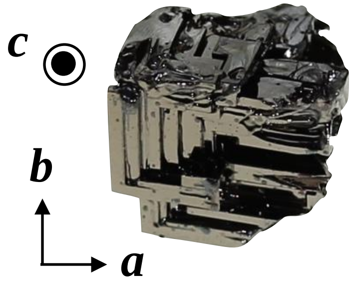
- Schottky-type anomaly near 2.3 K .

➔ Suggests a singlet ground state associated with spin dimer formation.

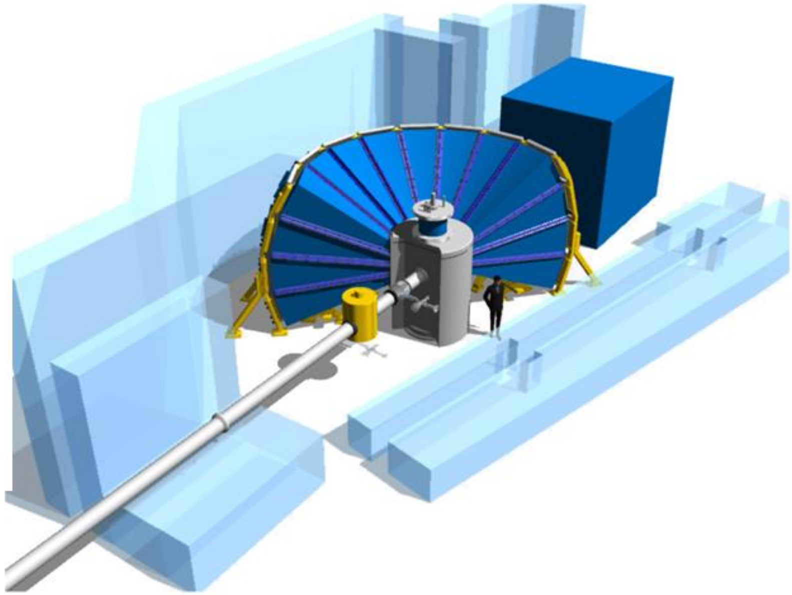
Microscopic evidence of spin dimer formation by inelastic neutron scattering experiments.

Sample preparation and instrument

Single crystal



HRC (BL12) J-PARC, MLF



- **Self-flux method**

Single crystal; 5 × 5 × 5 mm^3
Total amount; 10 g

- **Sample environment**

^{3}He refrigerator (0.3 – 2.0 K)
1K refrigerator (2.0 – 12.5 K)
GM refrigerator (5.2 – 15.3 K)

- **CEF**

Fermi: 500 Hz, E_i = 150 meV, ~32 h (La; ~24 h)

- **dimer**

Fermi: 100 Hz, E_i = 3 meV, ~32 h (La; ~24 h)

Crystalline electric field

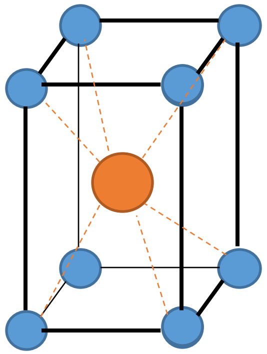
D. Ueta et al., PRB, 109 205127 (2024).

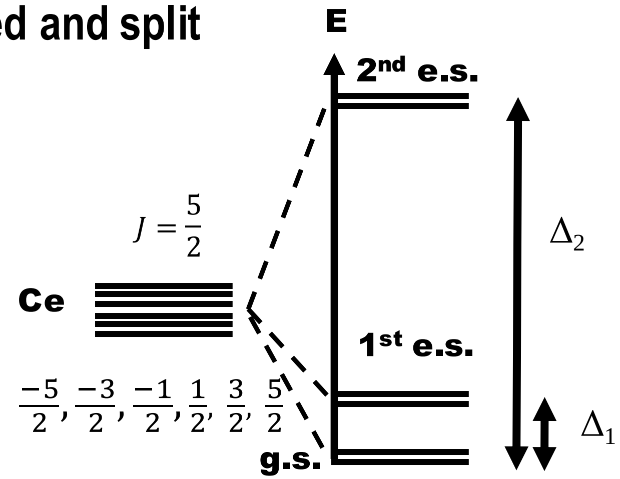
E_i = 150 meV, 500 Hz

Produced by a surrounding charge distribution

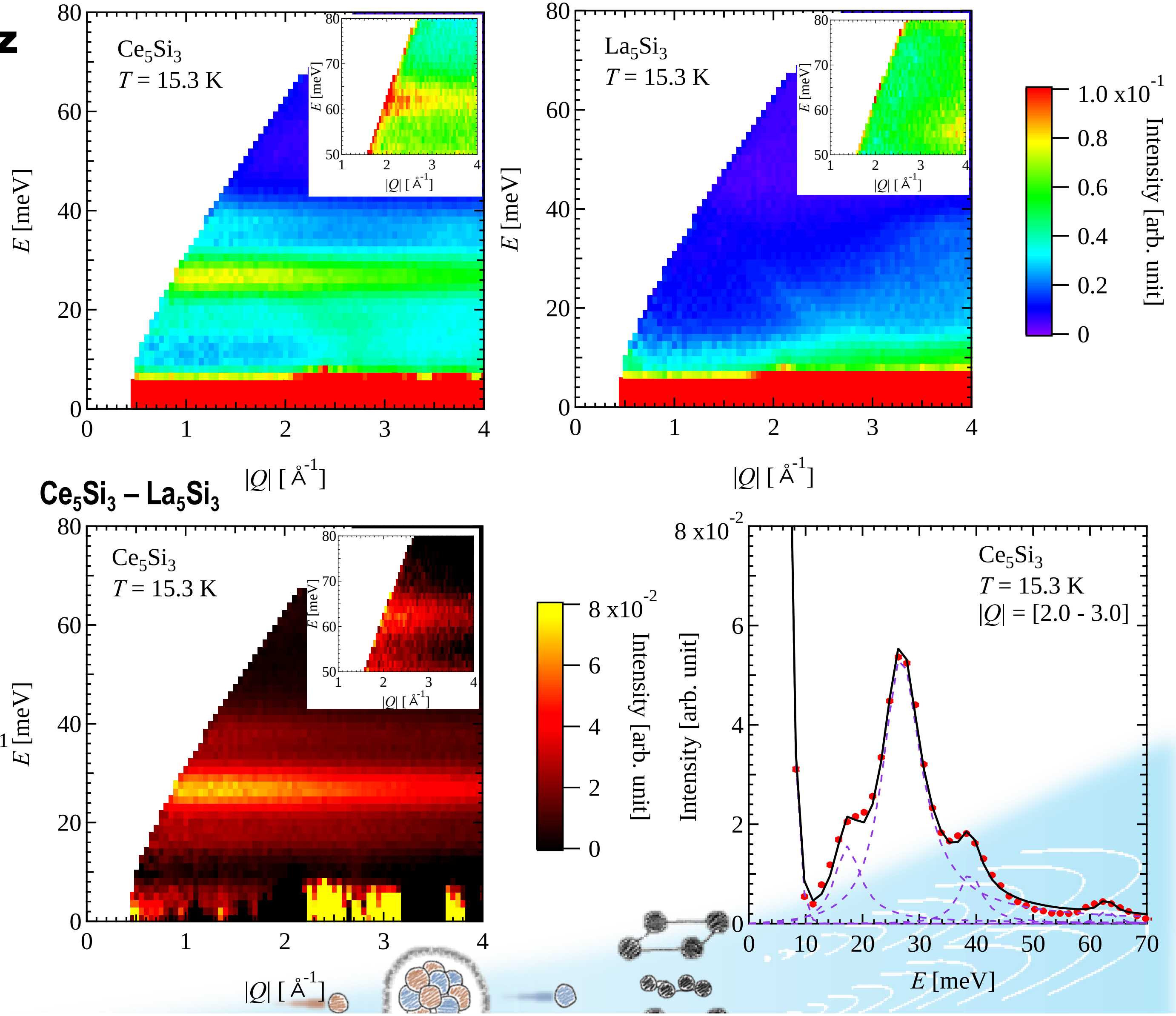
↓

Degeneracy of f - electron orbital states is removed and split





$$H_{\mathrm{CEF}}^{\mathrm{tetra}} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4$$

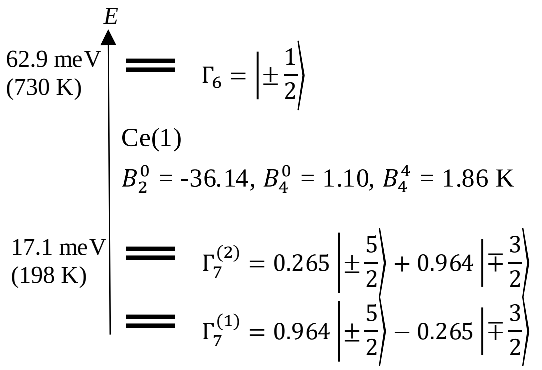


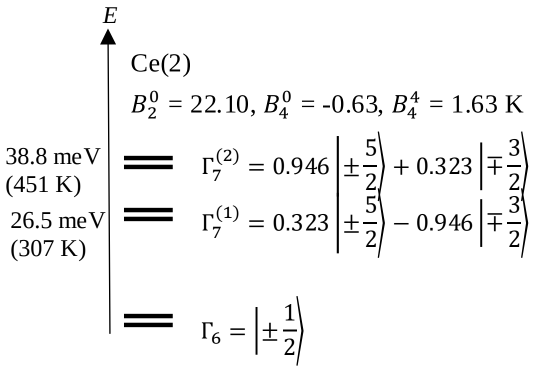
Magnetic excitations were observed around E = 17, 26, 39, 63 meV.

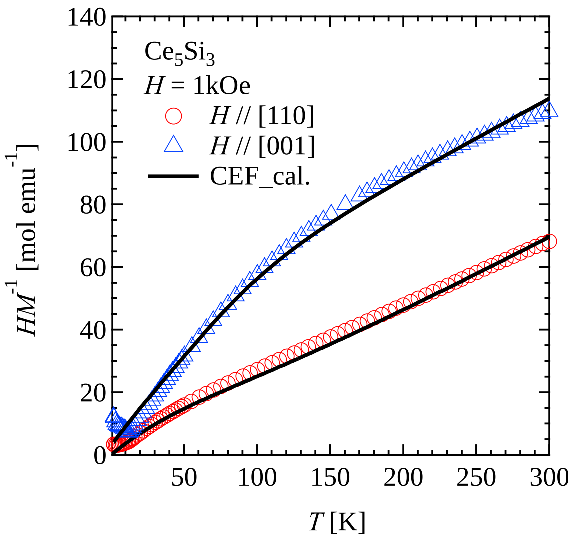
Crystalline electric field

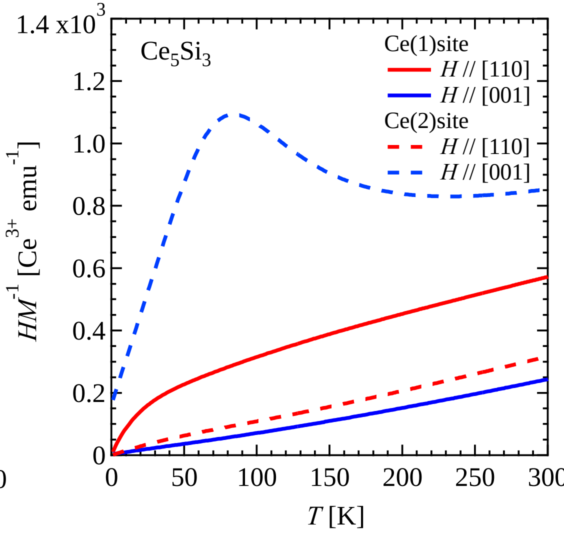
D. Ueta et al., PRB, 109 205127 (2024).

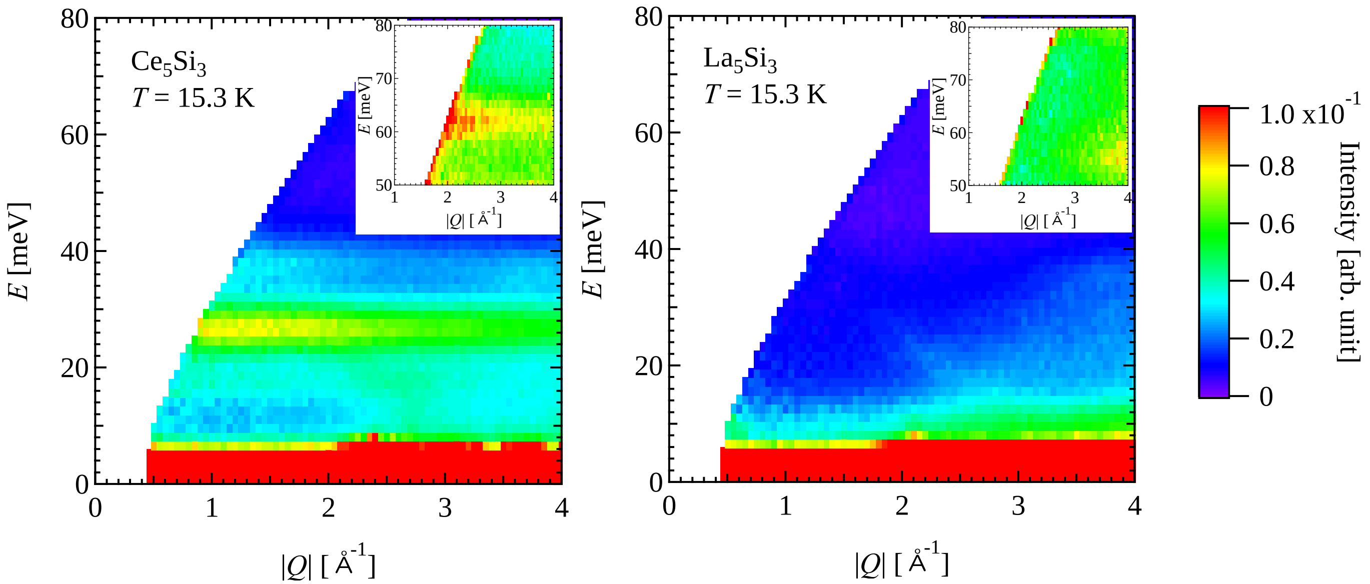
$$H_{\mathrm{CEF}}^{\mathrm{tetra}} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4 \qquad H_{CEF}^{tot} = H_{CEF}^{Ce1} + 4H_{CEF}^{Ce2}$$

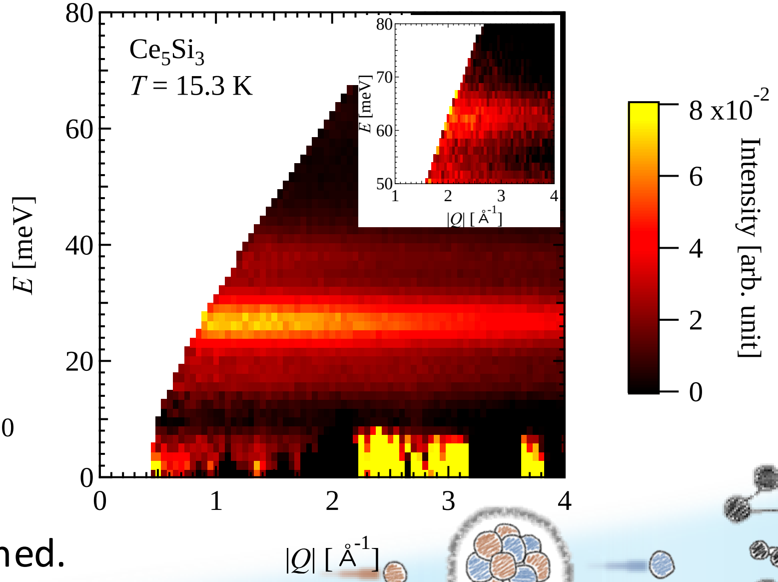


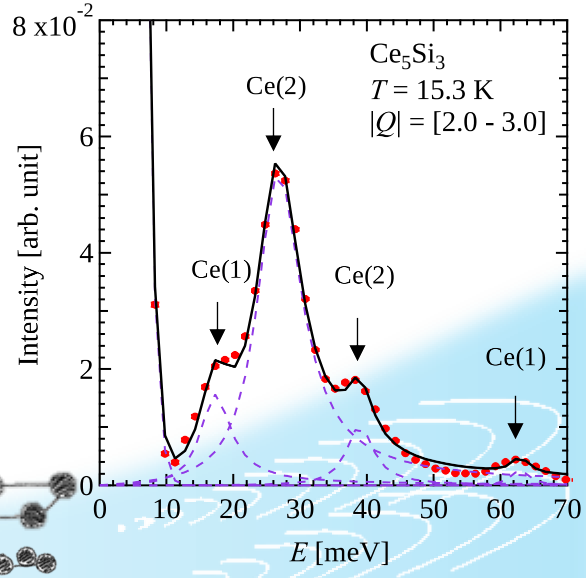












The behavior of the magnetic susceptibility is also well explained.

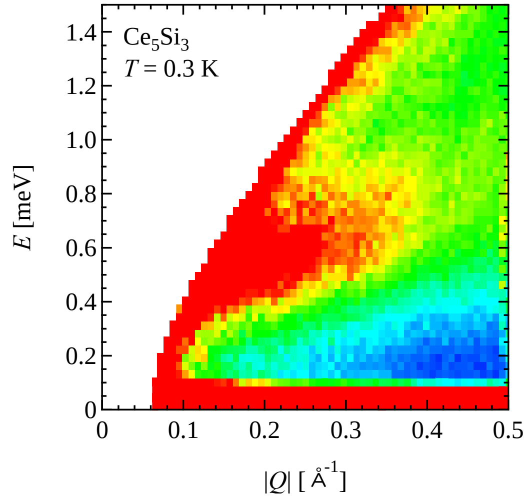
The crystalline electric field level schemes are determined.

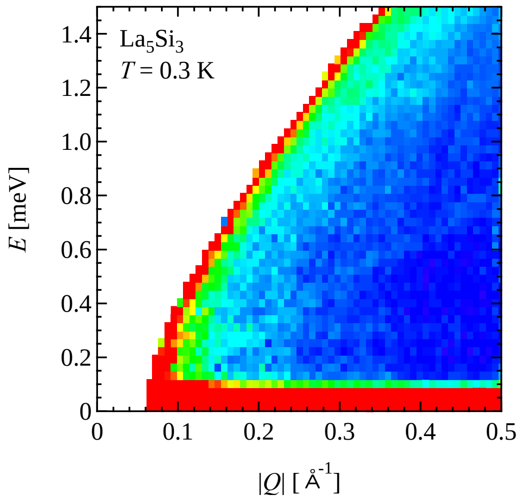
Low energy excitation due to spin dimer formation

E_i = 3 meV, 100 Hz

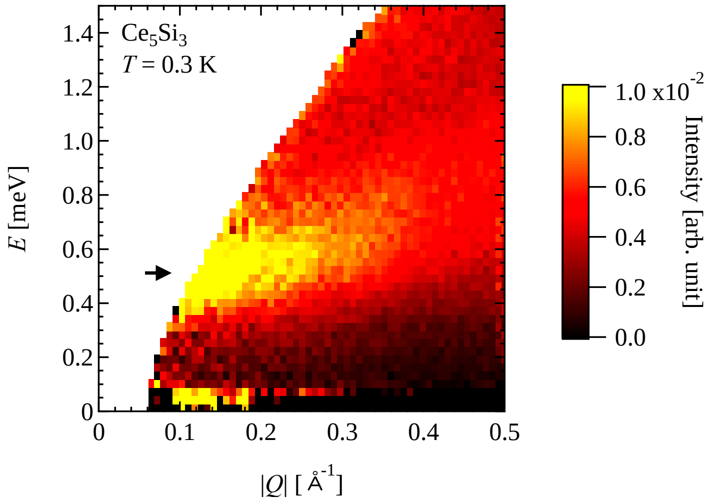
D. Ueta et al., PRB, 109 205127 (2024).

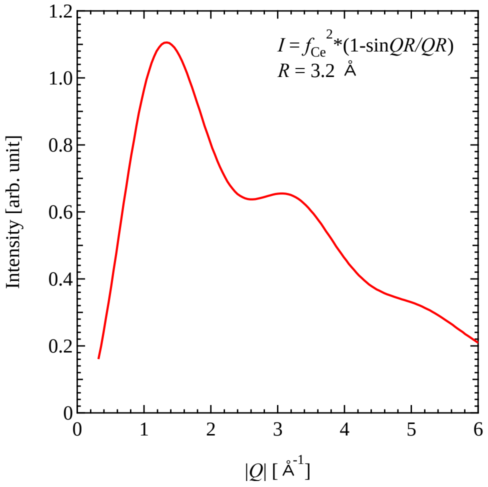
Single crystal





Ce_5Si_3 – La_5Si_3

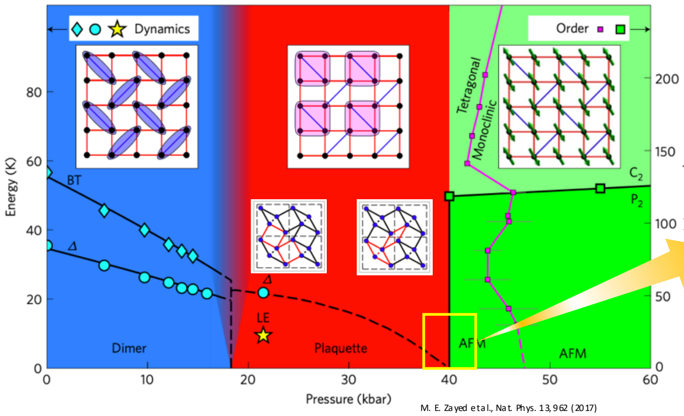




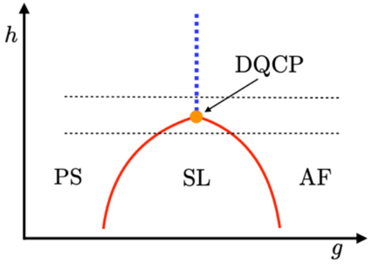
Magnetic excitation was observed around E = 0.5 meV, corresponding to the Schottky-type anomaly observed in the specific heat.
➔ Suggests splitting of the ground state.

Microscopic evidence of spin dimer formation is obtained.

Feature research plans



M. E. Zayed et al., Nat. Phys. 13, 962 (2017)



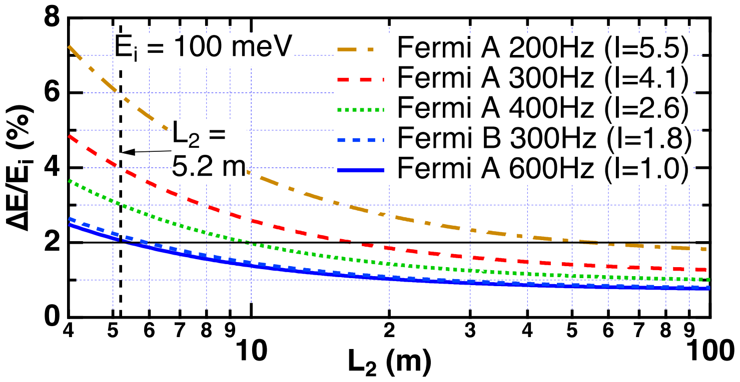
Spin Liquid phase

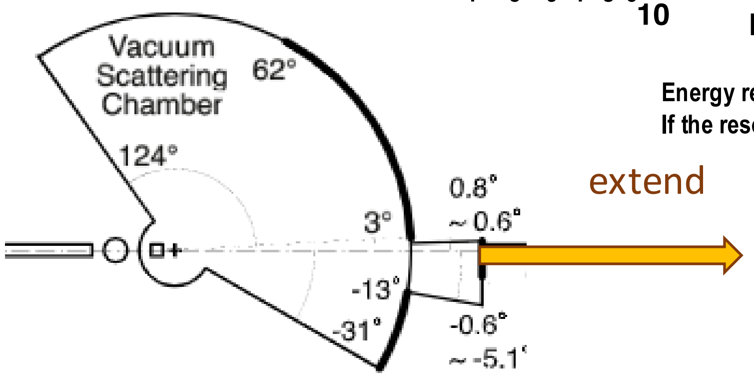
J. Yang et al., PRB. 105, L060409 (2022).

Spin dynamics in the low-Q range

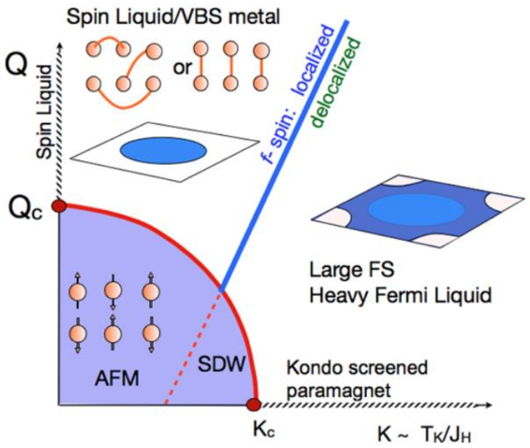
Increases resolution by extending the sample-detector distance.

E_i [meV]	ΔE [%]	ΔQ [$Å^{-1}$]
3	2	0.012
10	2	0.022
20	2	0.031
50	1	0.025
100	1	0.035
200	1	0.049
300	1	0.060

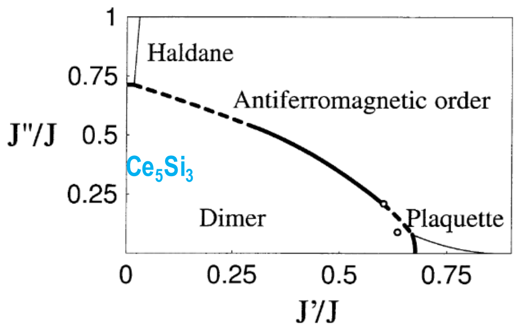




Energy resolution ➔ Q resolution.
If the resolution is the same, the intensity increases.



P. Coleman et al., J. Low Temp. Phys. 161, 182 (2010)



Exchange interaction
J : nearest neighbor, J' : next nearest neighbor, J" : inter-plane

A. Koga et al., JPSJ. 69, 3509 (2010)

In the d-electron system, the magnetic scattering intensity is halved at about 2.5 $Å^{-1}$ due to the magnetic form factor.
For samples that do not gain mass, it is useful to promote the study of magnetic excitations in the first Brillouin zone.

In the frustrated system, we aim to reach QCP and observe magnetic excitations due to spin liquid. Increase the small angle range performance (from 5° to 10°, higher resolution) and show the usefulness of the measurement in the 1st Brillouin zone.

Summary

HRC

- The fourth phase of HRC's project has begun.

➔ Focus on topological and frustration to promote research.

◆ The detector bank in the small-angle range is upgraded to improve the performance.

Ce_5Si_3 system

- Observed low-energy magnetic excitation with a gap of about 0.5 meV.

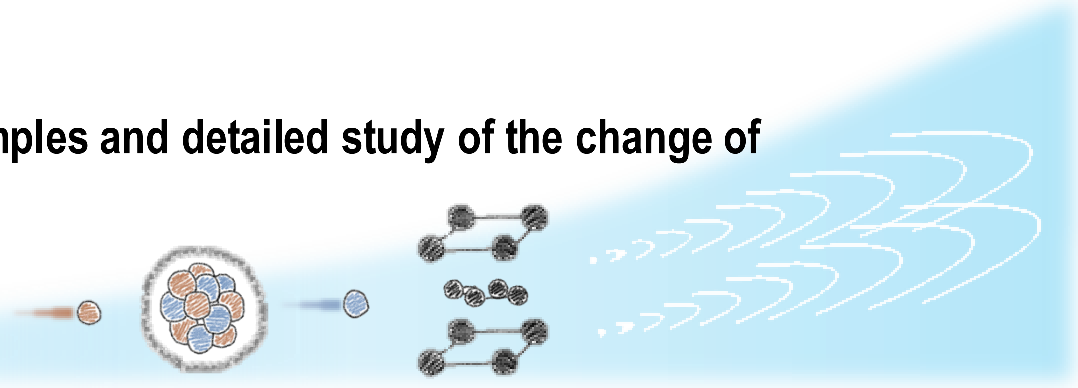
➔ **Microscopic evidence of spin dimer formation is obtained for the first time in the Ce system.**

➔ Magnetic excitations with large dispersion, which cannot be explained by SSL model.

➔ Suggests the influence of magnetically ordered Ce(1) sites and interdimer interactions in the interplanar direction.

◆ Comparison with Ce5Ga2Ge and Ce4LaSi3, in which the Ce(1) site is not magnetically ordered, reveals magnetic interactions unique to this system.

◆ Detailed observation of magnetic excitation using single crystal samples and detailed study of the change of dispersion under magnetic field.



Thank you for your kind attention