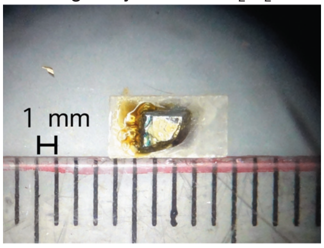
3–3 Novel Magnetism in Uranium Compound Revealed by World's Strongest Magnet

— High-Field Magnetic Structure in URu_2Si_2 Observed via Nuclear Magnetic Resonance —

(a) 45 T hybrid magnet in the National High Magnetic Field Laboratory at Tallahassee, Florida, USA

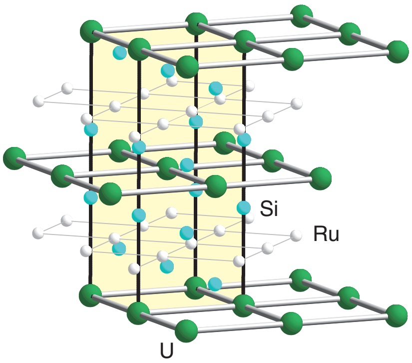
(b) ^{29}Si enriched single crystal of URu_2Si_2



Provided by Los Alamos National Laboratory (USA)

Fig.3-5 Photos of the hybrid magnet and single crystal of URu_2Si_2 used in this study
(a) This hybrid magnet can produce the world's strongest static field at 45 T, and is composed of a superconductive magnet of 11.5 T and a water-cooling resistive magnet of 33.5 T. A large tank filled with liquid helium and cold-water pipes are seen in this photo. This magnet consumes a huge amount of energy, namely 33 MW. (b) A photo of the isotopically enriched more than 99%–^{29}Si single crystal of URu_2Si_2.

(c) Unit cell of URu_2Si_2



(d) The magnetic structure is schematically illustrated when the external field above 35 T is applied. The arrows represent the magnetic moments on the U sites.

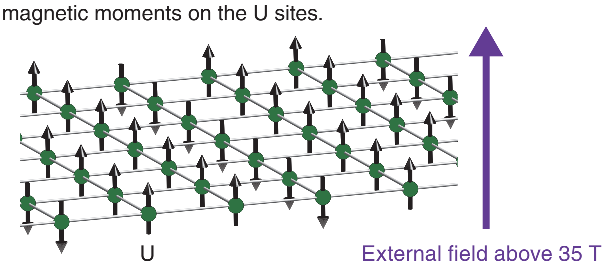


Fig.3-6 Crystal structure of URu_2Si_2 and the peculiar magnetic structure in the high field illustrated by the ^{29}Si NMR technique
(c) The unit cell is shaded yellow in color. U atoms form a square lattice. No magnetic moments on the U sites are generated even if a magnetic field of 35 T is applied. (d) Just above 35 T, a new magnetic state appears. The up and down arrows on the U sites represent the directions of the magnetic moments.

The uranium (U) material URu_2Si_2 exhibits a certain transition at 17.5 K accompanied by a large, specific heat jump, and it interestingly shows superconductivity at the low temperature of 1.5 K. This phase transition at 17.5 K is not an ordinary magnetic transition, and its origin is still unidentified after 30 years. Even now, many researchers are challenging to theoretically and experimentally identify this transition. If an external field is applied to this material, this transition temperature of 17.5 K gradually becomes lower. If the field reaches 35 T, the transition temperature becomes nearly 0 K and a new magnetic state emerges, the magnetic structure of which has not yet been determined. We have proposed a microscopic experiment to investigate the magnetic state via nuclear magnetic resonance (NMR), which is widely applied in medical research as magnetic resonance imaging. Consequently, our proposal to use the world's strongest hybrid magnet in the National High Magnetic Field Laboratory in Tallahassee, Florida, USA, was approved. Thanks to a collaboration with the Los Alamos National Laboratory, ^{29}Si nuclei enriched single crystals of URu_2Si_2 were prepared for our NMR purpose. Photos of the hybrid magnet and single crystal are shown in Fig.3-5.

As a consequence of the experiment, we have succeeded in observing ^{29}Si NMR signals in the magnetic state of URu_2Si_2 above 35 T. From the NMR spectral analysis, in this magnetic state, each U atom has a uniform magnetic moment and the moments' directions are bounded to be up or down along the external field, as shown in Fig.3-6. The propagation of the moments' array would be –up–up–down–, as if the array made striped patterns horizontally. On the other hand, below 35 T, no magnetic moment was observed; that is to say that the system is non-magnetic up to 35 T. Valence electrons of the U atom, i.e., 5f electrons, permeate site-to-site as metallic band electrons in low fields; however, if 5f electrons are bounded to the U sites suddenly above 35 T, then the generated magnetic moments are vertically aligned. Interestingly, the moments' array forms horizontal stripes with regard to the direction of the external field. Such a peculiar directional preference in the high-field metastable state is a clue to understanding the original transition below 35 T.

This study demonstrates that the nature of 5f electrons in a U atom can be highly variable with external parameters of magnetic fields or temperatures. This contributes to the knowledge for making new functional U compounds.

Reference
Sakai, H. et al., Emergent Antiferromagnetism Out of the "Hidden-Order" State in URu_2Si_2: High Magnetic Field Nuclear Magnetic Resonance to 40 T, Physical Review Letters, vol.112, issue 23, 2014, p.236401-1-236401-5.