A High Field ^{29}Si NMR Study of URu_2Si_2: A Novel Emergent Phase Out of the "Hidden-Order" State

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The so-called "hidden-order" (HO) state, which emerges in the URu_2Si_2 heavy-fermion superconductor below a zero field phase transition at T_O=17.5 K, has posed a long-standing mystery, particularly because of the inconclusive nature of its order parameter [1]. URu_2Si_2 is also a very attractive material displaying intriguing *T-H* phase diagrams (see Fig. 1(b)) determined by the electronic transport measurements such as resistivity and Hall effect. In particular, an anomaly in the Hall resistivity around H^*~22 T has been reported within the HO phase [2]. It is also reported that the critical field of the HO phase is about H_c=35 T by means of resistivity and magnetization measurements [3, 4]. Such electronic states in high fields are still unrevealed, let alone uncertain about the relationship with the HO state. Nuclear magnetic resonance (NMR) is a very suitable microscopic technique to investigate such anomalies in URu_2Si_2. Since we cannot reach the high magnetic field above 35 T for NMR purpose domestically at present, we have collaborated internationally with the research groups of National High Magnetic Field Laboratory and Los Alamos National Laboratory in USA. The single crystals studied in this project were enriched with 99.8% of ^{29}Si isotope, a very NMR sensitive nucleus, whose natural abundance is only 4.7%. High field NMR experiments in NHMFL were accepted to go up to 45 T, which is world record's static magnetic field. We have performed NMR experiments to take NMR shifts and spectrum just above H_c [5].

It is worth briefly mentioning few striking NMR results: the low-temperature ^{29}Si-NMR shift (*K*) anomaly around H^* and the critical increase of *K* towards H_c~35.6 T. Both observations must be related to crucial changes in the Fermi surface of URu_2Si_2. In general, *K* is proportional to static susceptibility χ via hyperfine coupling constant *A*, i.e., $K=A\chi$. By comparison with the magnetization $M=\chi H$ data [4], A=3.4 kOe/μ_B is obtained in the HO state, which is consistent with the reported values of 3.3-3.6 kOe/μ_B in the paramagnetic (PM) state. Consequently, this is also in agreement with the conclusion from recent low-field ^{29}Si-NMR measurements [6, 7] that there is no ordered magnetic moment in the HO state. From a magnetic point of view, the HO phase can thus be viewed as a Pauli-PM-like state. Therefore, now $K \propto \chi$ is proportional to the total density of states at Fermi level $D(E_F)$. Indeed, the change of *K* is qualitatively consistent with the change of $D(E_F)$ by external field, which has been calculated with internal collaboration of CCSE, JAEA. In the magnetic field scan procedure (A) in Fig. 1(b) at *T*=0.4 K, the transition between the HO and the magnetic II phases is also found to be of first order, since the ^{29}Si-NMR signal was gone suddenly just above H_c.

In the *T*-lowering (B) procedure in Fig. 1(b) at *H*~35.6 T, we have succeeded the first observation of the ^{29}Si NMR spectrum in the high field PM phase at 40 T as well as the complex ^{29}Si NMR spectrum in the magnetic II phase displayed in Fig. 1(a). This spectral shape for the II phase can be understood by a

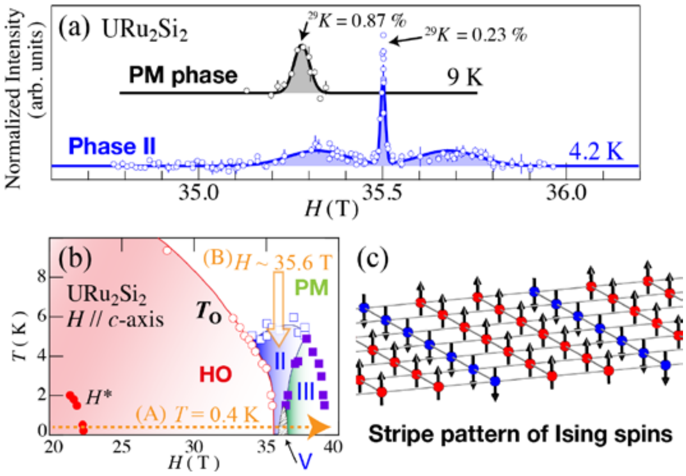

Fig. 1 (a) ^{29}Si NMR spectra in the paramagnetic (PM) and the magnetic II phases, (b) *T-H* phase diagram for URu_2Si_2, and (c) schematic spin structure in the II phase.

ferrimagnetic (or antiferromagnetic) ordering of Ising-type localized spin arrays, as illustrated in Fig. 1(c). Interestingly, the propagation vector is perpendicular to the symmetrical *c* axis (the external field direction). This magnetic structure violates the four-fold rotational symmetry in the crystal lattice. The estimate of Ising moments is uniformly about 0.6 μ_B per each uranium site.

Our high field NMR results provide the first microscopic evidence of the change in the magnetic susceptibility around H^*, which, in turn, could not be unveiled by previous transport measurements. The U localized moments with Ising character suddenly emerge in the II phase from non-magnetic itinerant HO state. This behaviour may find some connection with a recent proposed theoretical model [8]: the HO state might be an antiferro-ordered phase of extremely higher-order multipolar moments, i.e., dotriacontapole. Further investigations will clarify it more in the near future.

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