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Correlation between superconductivity and magnetism in uranium heavy fermion compounds

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Abstract
We review experimental results on typical uranium-based heavy fermion superconductors, UPd_2Al_3, UNi_2Al_3, URu_2Si_2 and UGe_2, which exhibit the coexistence of unconventional superconductivity (SC) with a magnetic long range order. On the basis of these results together with a possible model analysis, we argue the correlation between SC and magnetism.

1. Introduction

Correlation of superconductivity (SC) and magnetism has been one of the central issues in the field of strongly correlated electron systems. In particular, one may find interesting materials showing the coexistence of SC with ferromagnetic (FM) or antiferromagnetic (AF) order in uranium-based heavy fermion intermetallic compounds. These materials contain a periodic array of uranium ions [1]. Owing to both strong Coulomb correlation within the 5f shell and mixing of localized 5f-electron and itinerant electron wavefunctions, a heavy Fermi liquid state is formed at low temperatures, in which the electronic specific heat coefficient can be enhanced by several orders of magnitude over that of ordinary transition metals. When these materials are cooled from high temperatures, localized moments seem to progressively reduce due to local spin fluctuations. When a characteristic energy associated with the spin fluctuations is dominated by the RKKY type interaction between incompletely compensated atomic moments on different sites, the materials may undergo a magnetic phase transition at a finite temperature. In some materials, the heavy quasiparticles condense into the SC state at very low temperatures due to residual interactions among them.

It is well known that the series of rare-earth rhodium borides exhibits the coexistence of SC and magnetism [2]. It is particularly interesting to note that $ErRh_4B_4$ shows coexistence between SC and FM, although the SC finally disappears at very low temperatures and thus the coexistence is restricted to a narrow temperature range. Thorough investigation on the systems revealed that the magnetism is carried by 4f electrons well localized on rare-earth atom sites and the SC by 4d electrons of Rh atoms, which implies that SC and magnetism

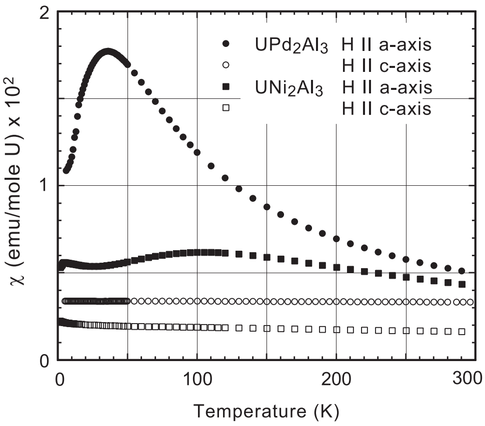

Figure 1. Temperature dependence of the magnetic susceptibility of UPd_2Al_3 and UNi_2Al_3. The external magnetic field is applied parallel and perpendicular to the hexagonal basal plane. Data were taken from [7].

are separated in real space. However, uranium heavy fermion compounds investigated here show a quite different kind of coexistence; both magnetism and SC are carried by 5f electrons. Typical examples are UPd_2Al_3, UNi_2Al_3, URu_2Si_2 and UGe_2. In the present paper, we review experimental results on these novel compounds, and discuss the correlation of these two long range orders.

2. UPd_2Al_3 and UNi_2Al_3

It is evident that both AF and SC in UPd_2Al_3 are carried by 5f electrons [3]. Polarized neutron scattering experiments revealed that the localized magnetic moments are sitting on uranium ion sites [4], whereas a specific heat jump at the SC transition temperature $T_{SC} \sim 2$ K is comparable to a value predicted from the BCS theory [3], the latter fact indicating that heavy quasiparticles originating from 5f electrons carry superconducting current. The system undergoes the phase transition into the AF order at $T_N = 14.3$ K; the magnetic moments are ferromagnetically aligned on a hexagonal basal plane, which is stacked antiferromagnetically along the c-axis with a propagation wavevector $\boldsymbol{Q}_0 = (0, 0, 1/2)$.

UNi_2Al_3 has the same crystal structure as UPd_2Al_3, and also exhibits the coexistence of the SC ($T_{SC} \sim 1$ K) and AF ($T_N \sim 4$ K) [5]. On the other hand, the magnetic ordering structure is an incommensurate magnetization density wave with an amplitude of staggered moment, $\sim$0.2 μ_B/uranium, along the a^*-axis [6], which is in remarkably contrast with the simple commensurate AF order in the Pd homologue having a larger staggered moment of 0.85 μ_B/uranium.

Figure 1 shows the temperature dependence of the magnetic susceptibility $\chi(T)$ of UPd_2Al_3 and UNi_2Al_3 [7]. The large anisotropic T-dependence implies that the magnetism is understood on the basis of the localized moment picture. Furthermore, the existence of a maximum in the $\chi(T)$ curves suggests that the crystalline electric field (CEF) ground state is a singlet for both systems [8, 9]. This means that the localized magnetic moment is carried by an even number of 5f electrons.

Figure 2 shows the inelastic neutron scattering spectra of (a) UPd_2Al_3 and (b) UNi_2Al_3. For UPd_2Al_3 we observe a two-peaked structure [10]. The lower energy peak sitting around 0.4 meV can be ascribed to the quasiparticle response, because the inelastic peak changes to a

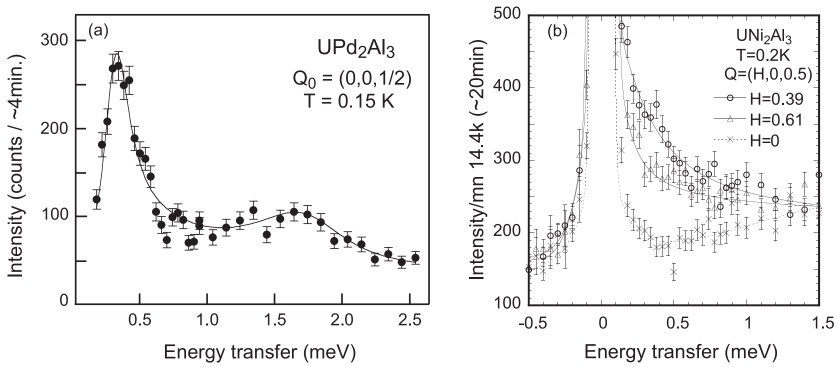

Figure 2. Inelastic neutron scattering spectra around the AF zone centre of UPd_2Al_3 (a) and UNi_2Al_3 (b). The measurements were made below the superconducting transition temperature $T_{SC} \sim 1.8$ and 1 K, respectively. See for details [10, 11, 14].

quasielastic feature with increasing temperature above T_{SC}, corresponding to closing of an SC energy gap. On the other hand, the higher energy broad peak around 1.6 meV can be attributed to a collective excitation of the localized moments, since the excitation energy increases on going away from the AF zone centre [10, 11]. This dispersive mode corresponds to magnetic exciton [10, 12], which is the collective mode of the (induced) localized magnetic moments propagating in the singlet CEF ground state system.

To explain the obtained spectra we proposed a duality model in which the coupling of the magnetic exciton to the quasiparticles leads to the heavy damping observed for the higher energy response [10]. This idea is based on the 'itinerant versus localized duality' model, which was originally applied to cerium compounds with a configuration of $(4f)^1$ [13]. In the present system, however, the total number of 5f electrons is not one but close to three. Therefore, instead of the original duality model for the cerium case, we assume that the existence of two types of 5f wavefunction, the well localized and less localized wavefunctions, leads to the dual nature in UPd_2Al_3. The former corresponds to the localized magnetic moments, whereas the latter leads to the itinerant heavy quasiparticles. Combining the above results deduced from the $\chi(T)$ curves, we describe the present situation as follows:

$$(5\text{f})^{\sim 3} \rightarrow (5\text{f localized component})^2 + (5\text{f itinerant component})^{\sim 1}.$$

Here, we note that only the 5f band of the itinerant component crosses the Fermi energy and thus does contribute to the Fermi surface volume. The above model to explain the inelastic neutron scattering spectra yields a strong coupling constant between the two components, which is consistent with the strong coupling feature observed in a tunnelling experiment. We wish to stress that, within the framework of the Eliashberg equation, both inelastic neutron scattering and tunnelling spectra are consistent with each other [10]. From all these results, we suggested that the magnetic excitons mediate superconducting pairing interaction between quasiparticles [10], although further theoretical investigations are required for a deeper understanding of the SC mechanism [12].

The quasielastic response was also found in UNi_2Al_3 around the AF zone centre $\boldsymbol{Q} = (0.5 \pm \delta, 0, 1/2)$ with $\delta \sim 0.11$, as seen in figure 2(b) [14]. Unfortunately, we have failed as yet to detect any change above and below T_{SC}. Further, we found no inelastic collective mode corresponding to the magnetic exciton. Therefore, we do not have any direct evidence for the correlation between magnetism and SC. This is due to experimental difficulties encountered in the preparation of single crystals showing SC [15], although even in the case of UPd_2Al_3 there was also an obstacle to obtaining a 'good' single crystal with higher T_{SC} [16]. Furthermore, the magnitude of the staggered moment of UNi_2Al_3 is one-quarter of that in UPd_2Al_3, and T_{SC} is only one-half. All of these factors prohibit us from unravelling the correlation between SC and AF in UNi_2Al_3.

The Knight shift deduced from NMR and muon spin relaxation (μSR) measurements gives useful information about the parity of SC: for UPd_2Al_3 both measurements consistently indicated a reduction of the Knight shift both parallel and perpendicular to the c-axis [17, 18], which indicates an even parity paring state, whereas for UNi_2Al_3 no reduction of the Knight shift was observed [19]. The latter fact strongly suggests the odd parity pairing, which is consistent with the results deduced from measurements of the SC upper critical magnetic field [15].

Finally, it remains an open question what is the origin of the difference in the magnetic and superconducting properties between UPd_2Al_3 and UNi_2Al_3, although we speculate that it may be related to the difference in the degree of itinerancy of 5f electrons.

3. URu_2Si_2

URu_2Si_2 undergoes two successive phase transitions at $T_0 \sim 17$ K and $T_{SC} \sim 1$ K [20]. The lower temperature transition corresponds to an onset of unconventional SC. On the other hand, it has been widely accepted that the higher temperature one is attributed to the AF ordering of extremely low moments of the order of 0.02–0.04 μ_B per uranium atom [21]. However, this transition seems to be incompatible with a large specific heat jump observed at T_0, $\Delta C/T \simeq 0.3$ J mol^{-1} K^{-2} [22]. This has led to many speculations that the true order parameter is not the weak magnetic dipole moment, but another of unknown symmetry such as quadrupoles.

Recent neutron scattering and NMR experiments under pressure unravelled the mysterious nature of this system. Neutron diffraction measurements indicated that the staggered moment increases almost linearly with pressure up to 0.25 μ_B/U at 10 kbar, and that a pressure-induced phase transition occurs at a critical pressure $P_c \sim 15$ kbar, above which the 3D-Ising type of AF phase appears [23]. NMR study using ^{29}Si nuclei gave the possible evidence for spatially inhomogeneous development of AF ordering below T_0 [24]; the AF volume fraction increases at the expense of the non-magnetic regions on cooling and/or applying pressure. These results seem to suggest that there is a first order phase transition between the phase-separated non-magnetic hidden and AF order, which is supported by μSR experiments performed under pressure [25]. Finally, these results strongly suggest that the weak Bragg peak intensity of the neutron diffraction experiments is not attributed to the extremely small moment, but to the small volume fraction of the AF region.

It seems thus important to (re)investigate thermodynamical properties under pressure. This is our motivation for dilatation measurements under pressure [26]. The T-dependence of the relative length change along the tetragonal a- and c-axes exhibited a kink around $T_0 \sim 17$ K at ambient pressure, corresponding to the phase transition between the paramagnetic (PM) and hidden order (HO) phase, consistent with the data in the literature. As pressure increases, T_0 shifts to higher temperatures accompanied by an attenuating of the anomaly, which finally could not be detected within the accuracy of our measurement above ~11 kbar. Surprisingly, we observed a new anomaly that evidently appears under pressure; for a sample it appeared around $T_p \sim 12$ K at 7 kbar [26]. As described above, this new anomaly is to be identified as the phase transition between HO and AF phases.

Plotting the temperature at which the anomaly appeared in the thermal expansion curves, we constructed the temperature (T) versus pressure (P) phase diagram for several samples

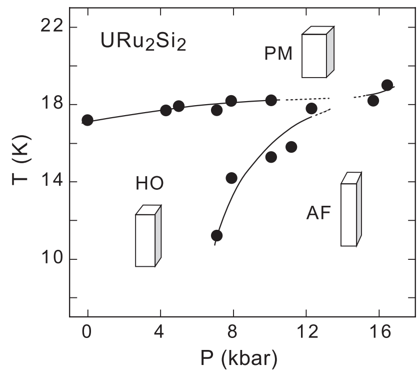

Figure 3. Temperature–pressure phase diagram of URu_2Si_2. PM, HO and AM denote the PM, HO and AF order. Each phase is characterized by the c/a-ratio, as schematically illustrated. Here a and c indicate the lattice constants along the a- and c-axes.

with different T_{SC} values [26, 27], a typical one being given in figure 3. Since it seems that the HO–AF phase boundary does not cross the vertical axis, we suggest that there is no AF region over the bulk of the sample at ambient pressure. (We thus speculate that the extremely small Bragg intensity observed at ambient pressure is due to the antiferromagnetism near the sample surface.) When lowering the temperature from the HO into the AF phase, we found that the sample length shrinks along the a-axis and elongates along the c-axis, with almost the same magnitude of the length change for both directions. Thus the c/a-ratio increases, where a and c denote the lattice constants along the a- and c-axes, respectively, and the volume evaluated from the relation $(\Delta V/V) = 2\Delta l_a/l_a + \Delta l_c/l_c$ reduces. Combining the results of the NMR and μSR experiments [24, 25], we consider that the new anomaly appearing around T_p is first order in nature. Furthermore, detailed examination of the thermal expansion data suggests that the first order line emanates from a bicritical point [27], which seems to be consistent with a recent theoretical prediction by Chandra *et al* [28].

4. UGe_2

UGe_2 is a ferromagnet in which SC appears in the pressure range of ∼10–16 kbar [29]. The Curie temperature T_c is about 53 K at ambient pressure and decreases with increasing pressure, and finally vanishes around ∼16 kbar. In the case of UPd_2Al_3, the period of the alternative stacking of $2c \sim 8$ Å is much smaller than an SC coherence length of the order of 100 Å; internal fields (due to the AF ordering), which SC Cooper paired electrons may observe, are probably cancelled out. In ferromagnetism such as in UGe_2, however, we expect that SC electrons detect a non-vanishing internal field. Thus, it is quite surprising that the FM with the local moment of the order of 1 μ_B/U coexists with the SC in UGe_2.

This interesting feature of UGe_2 raises a question concerning the nature of the coexistence of FM and SC. To resolve this question we made magnetization measurements in terms of ac and dc methods under external pressure [30–32].

Figure 4(a) shows the temperature dependence of the ac magnetic susceptibility around T_{SC} [30]. We clearly observe that the SC appears above ∼10 kbar; however, the diamagnetic susceptibility at low temperatures does not show the perfect shielding effect. (We observed the diamagnetism even at 8 kbar, but we note that the value of $4\pi\chi$ is as small as -0.2 at ∼50 mK [31].) Although the ambiguity of a demagnetizing field coefficient leads to an error

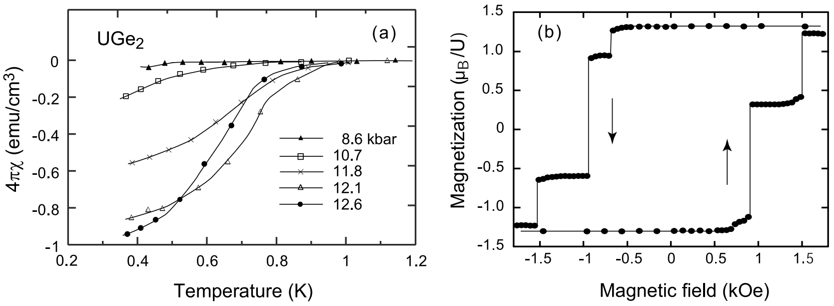

Figure 4. Temperature dependence of ac magnetic susceptibility around T_{SC} (a) and dc magnetization curve at ~0.5 K (b). The pressure dependence of the diamagnetic susceptibility indicates that the superconducting volume fraction grows with increasing pressure. The unusual steplike hysteresis curve may be due to the MQT effect. See for details [30] and [31].

in the absolute value, it is clear that the relative variation of the magnitude of the diamagnetism indicates that the SC volume fraction grows with increasing pressure and seems to reach almost 100% volume fraction around $P_{max} \sim 12$ kbar, where the SC (onset) temperature exhibits a maximum. On the other hand, we found that a peak in the ac magnetic susceptibility at the Curie temperature became broadened at pressures where the system shows the SC. From this observation, we conjectured that the FM loses the long range order nature at the high pressures. Finally, we suggested possible competitive coexistence of SC and FM in the investigated pressure region ($P < P_{max}$).

Furthermore, we discovered an interesting feature in the FM hysteresis curves at very low temperatures: the hysteresis curve shows a continuous behaviour at 4.2 K, as is usual, but at low temperatures below about 1 K the curve changes to a steplike feature (see figure 4(b)) [32]. (We note that very recently we observed a single-step hysteresis curve at ~0.5 K for a different sample with a smaller size (not shown here) [33].) The jump of the magnetization is very large, implying that a lot of magnetic moments change their direction almost simultaneously. Since the steps seemed to occur at regular intervals of magnetic field, we proposed a field-tuned resonant tunnelling model [32], which is a kind of macroscopic quantum tunnelling (MQT) phenomenon, and which led to the suggestion that the magnetic domain size is smaller than its SC coherence length. Indeed, there are different explanations from the above field-tuned resonant tunnelling model. The simplest one may be a domain wall effect: depinning of a magnetic domain wall from a pinning centre gives rise to many jumps in the hysteresis curve, which occur at irregular intervals of magnetic field for a usual ferromagnet. It is to be recalled that even in the domain wall motion process, there are two ways to overcome a potential barrier originating from the pinning of the wall, the classical and quantum tunnelling paths. It is not easy to give a crossover temperature that separates the classical and quantum regions. More detailed measurements are needed to reveal the origin of the novel magnetization behaviour.

5. Summary

In summary, we have shown the experimental results on the heavy fermion superconductors, UPd_2Al_3, UNi_2Al_3, URu_2Si_2 and UGe_2. For UPd_2Al_3, the duality model of 5f electrons seems to be plausible and allows us to suggest a new superconducting mechanism; the exchange of magnetic excitons (bosonic excitation) produces the SC pairing interaction between the heavy quasiparticles. It was also revealed that UPd_2Al_3 has the even parity paring whereas UNi_2Al_3 shows the odd parity. The origin of this difference remains an open question. For URu_2Si_2, we discovered a new anomaly in the thermal expansion that appears only under pressure. We constructed the temperature–pressure phase diagram, in which the first order line separating the hidden and AF orders emanates from the bicritical point. Finally, we suggested for UGe_2 that the SC coexists with the ferromagnetism in a competitive way in the investigated pressure range. Further, we reported the discovery of the steplike hysteresis curve at very low temperatures. To reveal the origin we need further measurements.

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