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Observation of two low-energy responses in UPd_2Al_3—interaction of magnetism and superconductivity?

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Abstract

We have performed neutron inelastic scattering measurements for the heavy fermion superconductor UPd_2Al_3. From comparison of the results with calculation, we suggest that the spin fluctuation determines the superconducting transition temperature T_c, and that the strong coupling between the heavy electrons and spin waves may lead to the observation of two low-energy responses. We also suggest, within the model investigated, that the spin wave excitation energy may be reduced below T_c. © 1998 Elsevier Science S.A.

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1. Introduction

UPd_2Al_3 crystallizes in the hexagonal $PrNi_2Al_3$ structure (space group P6/mmm) with the lattice constants of $a=5.350$ Å and $c=4.185$ Å at $T=300$ K, and shows coexistence of antiferromagnetism ($T_N=14.3$ K) and superconductivity below $T_c=2$ K [1]. Magnetic susceptibility [2] and neutron scattering measurements [3–5] have revealed that the moments are coupled ferromagnetically in the basal plane, but are coupled antiferromagnetically along the *c*-axis with an ordering wavevector $\mathbf{Q}_0=(0,0,1/2)$. Excellent agreement between de Haas-van Alphen effect measurements and band-structure calculations shows that 5f electrons in UPd_2Al_3 are itinerant *in the ground state* [6,7]. On the other hand, *finite temperature* (*energy*) *phenomena* such as heat capacity [8], μSR [9] and photoemission [10] experiments suggest the existence of the localized state as well as the itinerant state. This is hereafter referred to as the duality model, in which one assumes that the magnetism is carried by the localized-part of the 5f electron and superconductivity by the itinerant-part.

It was reported by Petersen et al. [11] that there were no changes in the spin-wave excitations around the antiferromagnetic zone center $\mathbf{Q}_0$ above and below T_c, but we observed for the first time the influence of onset of superconductivity on the spin fluctuation [12]. We have found that there are two contributions to the dynamical response in UPd_2Al_3. The first is a heavily-damped spin-wave, which has been observed previously by Petersen et al. It shows no appreciable change on warming through T_c, but softens and becomes overdamped as T approaches T_N. The second is a quasielastic-like component that exists in the antiferromagnetically ordered state and is strongly localized around the ordered wavevector. As a function of temperature this component exhibits a minimum at T_c, and increases strongly below T_c.

It was suggested previously [12] that the observation of the two components is consistent with the duality (i.e. two subsystem) model that there are two separate responses from 5f electrons, but the correlation between the two contributions remains unclear. Furthermore, the origin of the increase of the intensity below T_c is not understood at all. In the present paper, therefore, we will discuss the origin of the existence of the two components and their correlation. We will further discuss the effect of superconductivity on the spectrum.

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2. Experimental

The sample used in the present experiments was prepared by the Czochralski pulling method with a nominal composition of $UPd_{2.02}Al_{3.03}$ [13,14]. Neutron-scattering experiments were performed with the IN14 triple-axis spectrometer situated on the cold source at ILL, Grenoble. The spectrometer was operated in a constant k_f mode, with 1.3 $Å^{-1}$ (final energy 3.5 meV). The elastic energy resolution was about 0.12 meV (full width at half maximum) at zero energy transfer, and the q-resolution for $\mathbf{Q}=(0,0,\frac{1}{2}+q)$ was $\Delta q=0.0018$ r.l.u. (FWHM) at the Bragg peak. The q-resolution for an inelastic process is greater. A detailed description of the experiments is given elsewhere [12].

3. Results and discussion

3.1. What factor determines the superconducting transition temperature?

We performed the constant-**Q** scans along the c^*-axis ($\mathbf{Q}=(0,0,\frac{1}{2}+q)$) at several temperatures, and an example measured at $T=10$ K is plotted in Fig. 1. We found that the spectrum at this temperature is reproduced well in energy by a single Lorentzian function, Eq. (1), and that the line width $\Gamma(q,T)$ is proportional to q^2 for $q/q_B<0.2$ with a zone-boundary wavevector of $q_B=0.25$ r.l.u. (along the c^*-axis),

$$\frac{\mathrm{Im}\chi(q,\omega,T)}{\omega}=\chi(q,T)\frac{1}{\pi}\frac{\Gamma(q,T)}{\omega^2+\Gamma(q,T)^2}, \quad (1)$$

$$\Gamma(q,T)=\Gamma(\kappa^2+q^2)=2\pi T_0((\kappa/q_B)^2+(q/q_B)^2), \quad (2)$$

where κ^{-1} corresponds to a correlation length. According to Moriya and coworkers [15,16], the dynamical susceptibility around the antiferromagnetic instability in heavy electron systems takes the same form as in the self-consistent renormalization (SCR) theory for weak antiferromagnetic metals. We obtained from the least square fitting T_0 as 28.5±3 K, which characterizes the frequency spread of the spin fluctuation. A similar analysis for $T=20$ K data ($>T_N$), in which q_B is twice as large as that at $T=10$ K ($<T_N$), yielded $T_0=25.5\pm5.5$ K.

It may be possible to regard T_0 as the Debye temperature ω_D in a usual phonon-induced BCS superconductor. Assuming the degenerate temperature of the quasiparticle-band, T_F~70–100 K [17], we have the relatively large ratio of T_0/T_F~0.3. This suggests that UPd_2Al_3 is a strong-coupling superconductor. (For a typical BCS superconductor the magnitude of ω_D/T_F is less than 1%.)

It was suggested theoretically that there is a strong connection between T_0 and T_c [16]. Actually we observe the linearity-relation between them for UPd_2Al_3 and high-T_c cuprates in spite of quite different magnetism and dimensionality, as shown in the inset to Fig. 1. This result implies that there is common mechanism of superconductivity among those systems, i.e. the spin fluctuations [18].

3.2. What is the origin of the damping and the increase of intensity below T_c?

As the temperature is lowered below about 6 K, the spectrum exhibits two components [12], the inelastic component and the quasielastic-like one, as demonstrated in Fig. 2. The single Lorentzian function is not sufficient to describe the spectrum, and we require two terms, g_{QP} and g_{SW} (see Eq. (3)), each of which may correspond to the itinerant- and localized-part of the f-response, respectively, according to the duality model,

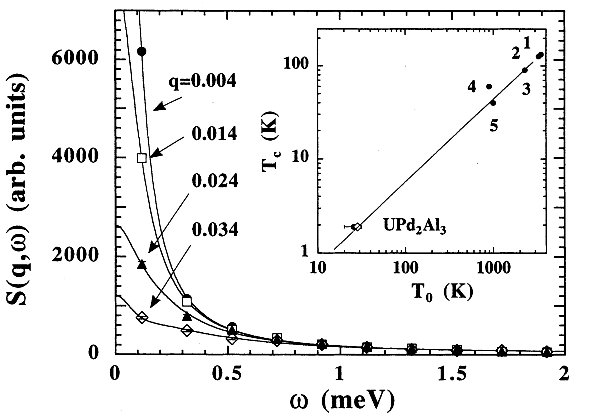

Fig. 1. Scattering function $S(q,\omega)$ for $\mathbf{Q}=(0,0,\frac{1}{2}+q)$ measured at $T=10$ K. Inset indicates the linearity-relation between T_0 and T_c for UPd_2Al_3 and high-T_c cuprates; 1, $HgBa_2Ca_2Cu_3O_{8+\delta}$; 2, $Tl_2Ba_2Ca_2Cu_3O_{10}$; 3, $YBa_2Cu_3O_{6+\delta}$; 4, $YBa_2Cu_3O_{6+\delta}$; 5, $La_{1.85}Sr_{0.15}CuO_4$. Closed circles (diamond) denote T_0 determined in the paramagnetic (antiferromagnetic ordered) phase.

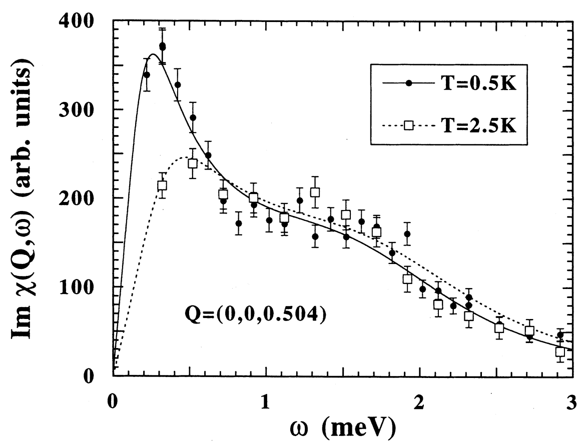

Fig. 2. Imaginary part of the dynamical susceptibility at $T=0.5$ K ($<T_c$) and 2.5 K ($>T_c$). Solid and broken lines are calculated curves (see text for details).

$$g_{QP}(\omega) = \chi_{st}\gamma/(i\omega - \gamma),\ g_{SW}(\omega) = \alpha^2/(\omega - \Delta). \quad (3)$$

Here, χ_{st} and γ represent the static susceptibility and a characteristic energy-width of the quasiparticle (QP) part, respectively, and the spin wave (SW) excitation is assumed to occur at Δ with spin matrix element α. Then we introduce the coupling between QP and SW, J. Finally we obtain the following expression of Eq. (4) for the f-response at $T=0$ K. This model was originally derived by Becker et al. for study of line width of crystal-field excitations in the well-localized systems [19]. Then it was applied to actinide systems to investigate the electronic damping of the broadened spectral-response by Buyers and Holden, as discussed in [20], in which they put the numerator of g_{QP} to be unity, $\chi_{st}\gamma = 1$.

$$\mathrm{Im}G_{ff}(q,\omega) = \left(\frac{-\alpha^4 J^2 \chi_{st}\gamma\omega}{\omega^2+\gamma^2}\right) \Big/ \left(\left(\omega - \omega_q + \frac{\alpha^2 J^2 \chi_{st}\gamma^2}{\omega^2+\gamma^2}\right)^2 + \left(\frac{\alpha^2 J^2 \chi_{st}\gamma\omega}{\omega^2+\gamma^2}\right)^2\right) + \left(\frac{-\alpha^4 J^2 \chi_{st}\gamma\omega}{\omega^2+\gamma^2}\right) \Big/ \left(\left(\omega + \omega_q - \frac{\alpha^2 J^2 \chi_{st}\gamma^2}{\omega^2+\gamma^2}\right)^2 + \left(\frac{\alpha^2 J^2 \chi_{st}\gamma\omega}{\omega^2+\gamma^2}\right)^2\right), \quad (4)$$

Using $\omega_q = \Delta + \alpha^2 J_{ff}$ (J_{ff} is a coupling constant between f-sites) and $\chi(q,\omega) = -g_J^2 \mu_B^2 G(q,\omega)$, we calculated Im $\chi(q,\omega)$, as denoted by the solid and broken lines in Fig. 2. If we assume $J=0$, then Eq. (4) leads to the delta-function with a sharp inelastic peak at $\omega=\omega_q$. It should be noted, however, that the response of g_{QP} is always situated around $\omega\sim 0$, independent of the magnitude of J. This means that as g_{QP} is measured through the window of G_{ff}, g_{QP} can be measured only when J is sufficiently large. The good agreement between experiment and theory (see below for details), suggests that the broadened spectrum may be ascribed to the non-vanishing coupling between QP and SW. This seems to be consistent with the above results, i.e. the strong coupling between superconductivity and spin fluctuations.

To investigate the origin of the increase of the intensity with decreasing temperature below T_c, we examined the above model by simulations, and we found some possibilities; the increase of the coupling constant J and/or the decrease of the excitation energy ω_q, the latter being simulated in Fig. 3. For a conventional BCS superconductor the magnitude of electron-phonon interaction may not be changed above and below T_c. Thus, assuming the same coupling constant for both temperatures, we obtain from the least square fitting the following set of parameters for $T=0.5$ K (2.5 K); $\omega_q = 1.82\pm0.02$ (2.02 ± 0.03) meV and $\gamma = 1.25\pm0.06$ (1.46 ± 0.10) meV. Here α and χ_{st} were also assumed not to be changed above and below T_c. The good agreement suggests that the increase of the intensity may be ascribed to the decrease of ω_q *within this model*, and the result may be interpreted as follows: the energy-gap is already induced above T_c by the antiferromagnetic ordering at the Fermi surface [21]. As temperature is lowered below T_c, superconductivity intends to open the energy gap. Hence the gap-formation due to superconductivity may compete with that due to antiferromagnetism and eventually overcome the latter, leading to the reduction of the magnitude of the gap formed by antiferromagnetism above T_c. This may correspond to the reduction of ω_q, as observed. (But this reduction may be small, because antiferromagnetism maintains superconductivity; in other words, small feedback effect.)

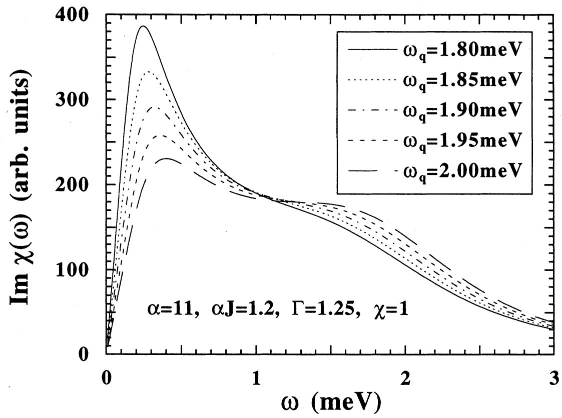

Fig. 3. Simulation on the basis of the phenomenological model given in the text. Note that the smaller value of ω_q increases the intensity of the low energy response.

The above model looks to explain the temperature dependence, but it fails to describe the strong **Q**-dependence, as may be seen in Fig. 4. In the fitting we assume the parameters γ and χ_{st} are independent of **Q**, because the

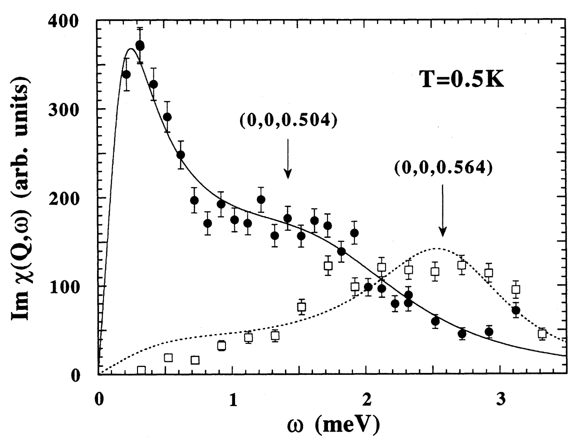

Fig. 4. Im $\chi(\mathbf{Q},\omega)$ measured at $T=0.5$ K ($<T_c$). Note the presence of two components at $\mathbf{Q}=(0,0,0.504)$ but only the damped spin-wave mode at $\mathbf{Q}=(0,0,0.564)$.

itinerant states in the above model have no wavevector dependence in the sense that the response will be at all **Q**-values. The fact that the quasielastic response is so sharply peaked around the antiferromagnetic ordering wavevector is a crucial discovery of these experiments, and it is one which is not reproduced by any theories to date.

In the present paper we have tried to describe the interaction of the two low-temperature responses (inelastic spin wave and quasielastic-like response) with the formalism developed originally by Becker et al., and applied to 5f systems by Buyers and Holden. We find that a qualitative fit to the data can be obtained with reasonable parameters. As temperature is lowered the increasing strength of the quasielastic response may be simulated by decreasing the excitation energy ω_q. Despite this parameterization of our results, a number of caveats need to be mentioned.

1. The theory developed by Becker et al. treats the response between itinerant and localized states. In the present model, we correspond these two states to the respective ones of the duality model. But for UPd_2Al_3 no crystal-field transition, which is the signature of the localized f-states, has been observed. The strong interaction between the two — at all temperatures — prevents the identification of well-defined crystal-field states.
2. No account has been taken in the above description of the data of the fact that there must be a gap-opening effect in the magnetic response below T_c [22]. This may happen on yet another energy scale, and the decrease of the width γ suggested above may reflect the reduction of the quasiparticle density of states. Given this caveat, it appears that the major influence of superconductivity on the spin fluctuations is the decrease of the excitation energy ω_q.

In conclusion, we obtain the characteristic temperature of the spin fluctuation, $T_0 \sim 27$ K (an average of 28.5 K and 25.5 K), and found that it determines the superconducting transition temperature T_c. We suggest that the strong coupling between the quasiparticles and spin waves, both of which originate from 5f electrons, seems to lead to the observation of two low-energy responses. The increase of the intensity of the quasielastic-like response below T_c may be attributed to the reduction of the excitation energy ω_q within the model investigated.

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