Superconducting Energy Gap Observed in the Magnetic Excitation Spectra of a Heavy Fermion Superconductor UPd_2Al_3

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High resolution neutron scattering experiments have been carried out in order to study the interplay between magnetism and superconductivity in the heavy fermion superconductor UPd_2Al_3. We found direct evidence for a magnetic excitation gap associated with superconductivity. We observed 1% suppression of the antiferromagnetic Bragg intensities below the superconducting transition temperature $T_c = 1.9$ K. We also observed the increases of the spin wave excitation energy and its linewidth in the superconducting state. These results indicate a strong coupling between magnetism and superconductivity in this compound. [S0031-9007(98)06447-3]

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Anisotropic superconductivity in heavy fermion superconductors is one of the most exciting topics in the field of condensed matter physics. The most important issue for the heavy fermion superconductor is that quasiparticles with a heavy mass ($m^* \sim 10^2 m_0$) are of an f-electron character, condensing into Cooper pairs. When we compare the phonon-mediated attractive interaction to the strong repulsive interaction among the f electrons, it is theoretically difficult for the former interaction to overcome the latter one [1]. To avoid a large overlap of the wave functions of the paired particles, the heavy fermion system would rather choose an anisotropic channel, like a p-wave spin triplet or a d-wave spin singlet state, to form Cooper pairs. In fact, the heavy fermion superconductor exhibits antiferromagnetic (AFM) ordering. Therefore the interplay of superconductivity with the coexisting AFM ordering is a key concept for the ground state properties [2].

Very recently we have observed a magnetic excitation gap associated with superconductivity in UPd_2Al_3 [3]. UPd_2Al_3 is a typical heavy fermion superconductor with $T_c = 2$ K. It also exhibits antiferromagnetic ordering with a relatively large magnetic moment of $0.85\mu_B/U$ below the Néel temperature $T_N = 14.5$ K [4,5]. It is, however, reported that the neutron inelastic scattering profile could be explained by the coupling model which reproduces the quasielastic scattering due to strong damping of the spin wave excitation by the conduction electrons [6]. Namely, no trace of the magnetic excitation gap was found, which is inconsistent with our previous study.

We have continued studying neutron inelastic scattering experiments with much higher resolution and lower temperatures. The present paper indicates clear evidence for the superconducting energy gap appearing in the magnetic excitation spectra. We also present the neutron data to show the strong coupling of the magnetic and superconducting order parameters. Finally, we mention the influence of the superconductivity on the spin wave excitation.

Neutron scattering experiments were carried out using a cold neutron triple-axis spectrometer LTAS installed at C2-1 beam port of research reactor JRR-3M in Japan Atomic Energy Research Institute. The collimation was 26′-70′-72′-72′. The constant-Q profiles were measured with a fixed final energy $E_f = 3$ or 4 meV, which gave the energy resolution of 85 and 150 μeV, respectively, at the energy transfer $\Delta E = 0$ meV.

The samples were grown from the starting composition $UPd_{2.02}Al_{3.03}$ by the Czochralski pulling in a tetra-arc furnace [7]. We observed zero resistivity below 1.95 K, and a bulk superconducting transition at 1.85 K observed by the specific heat measurement. The residual resistivity ratio was 60. The samples were cooled down by a ^{3}He cryostat or ^{3}He-^{4}He dilution refrigerator.

Figure 1 shows the inelastic scattering profiles measured with $E_f = 4$ meV at the (0 0 0.5) antiferromagnetic Bragg point as a function of the sample temperature. At 4.2 K the profile can be described by a combination of a broad quasielastic (dotted line) and an inelastic Lorentzian line shape at $\Delta E = 1.5$ meV (dashed line), as well as a sharp Bragg peak (dashed line) and an incoherent scattering (dash dotted line) both centered at $\Delta E = 0.0$ meV. The broad peak at $\Delta E = 1.5$ meV is a spin wave excitation as reported before [8]. Below $T_c = 1.9$ K, the position of the quasielastic peak shifts to a higher energy. A clear peak with a maximum at $\Delta E = 0.4$ meV appears at 0.4 K. Figure 2 shows $\chi''(q,\omega)/\omega$, which was obtained from the spectra measured with much higher energy resolution ($E_f = 3$ meV). At 0.5 K $\chi''(q,\omega)/\omega$ exhibits a clear peak at $\Delta E = 0.36$ meV. This is evidence for the existence of a magnetic excitation gap associated with superconductivity. Any kind of quasielastic line shape disagrees with the experimental data. On the other hand, the data at 2 K show a typical quasielastic line shape which can be described by a Lorentzian centered at $\Delta E = 0$. Figures 1 and 2 show a continuous change of the magnetic excitation spectra from the quasielastic line shape to the inelastic one below T_c.

Figure 3 shows the temperature dependence of the energy of the magnetic excitation gap. The gap starts

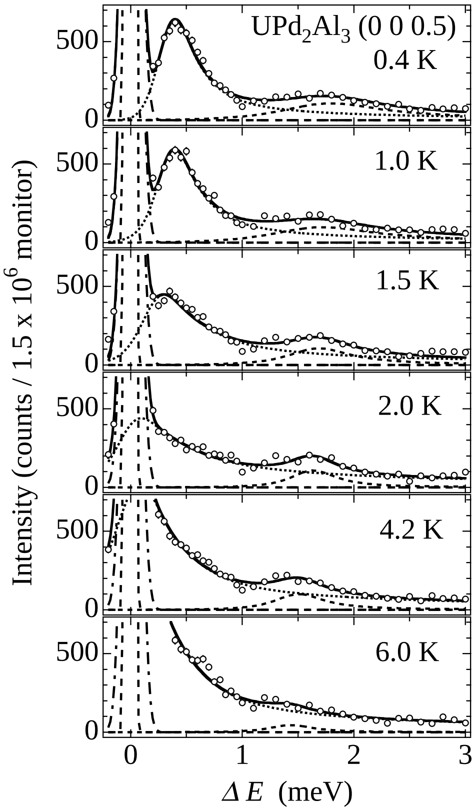

FIG. 1. Temperature dependence of the neutron inelastic scattering profile measured at $Q = (0\ 0\ 0.5)$ in UPd_2Al_3.

to open at $T_c = 1.9$ K and increases with decreasing the temperature. This temperature dependence is comparable to the one of the superconducting energy gap expected from the weak coupling BCS theory (dotted line), which is normalized by the maximum energy gap. The energy gap at the lowest temperature is 0.36 meV which corresponds

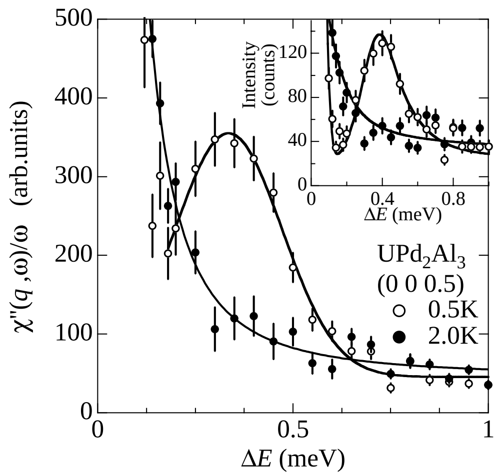

FIG. 2. Temperature dependence of the $\chi''(q,\omega)/\omega$ in UPd_2Al_3. The inset shows the neutron inelastic scattering profile measured at $Q = (0\ 0\ 0.5)$ with a fixed final energy of 3 meV.

to $2\Delta = 2.2k_BT_c$. It is in the same order compared with the weak-coupling BCS theory, $2\Delta = 3.5k_BT_c$. In addition we have confirmed that the gap disappeared with applying the magnetic field larger than the upper critical field H_{c2} [3].

From these results we conclude that the observed magnetic excitation gap corresponds to the superconducting gap. At present we don't know the microscopic mechanism why the magnetic excitation gap behaves very similar to the superconducting gap which is obviously a charge gap. It should be noted, however, that this phenomenon is due to the strong coupling between magnetism and superconductivity.

Since the existence of the anisotropic superconducting energy gap is clear from the power-law behavior of the specific heat [9] and NMR measurements [10–12] in UPd_2Al_3, it is of particular interest to measure the Q dependence of the energy gap in order to clarify the anisotropy of the superconducting gap. Figure 4 shows the neutron scattering intensity map measured as a function of the momentum transfer Q along the [0 0 l] direction. At 0.4 K the magnetic excitation gap exhibits a remarkable Q dependence. The energy gap shows a minimum about 0.36 meV ($\sim 2.2k_BT_c$) at the AFM Bragg point (0 0 0.5) which corresponds to the zone center in reciprocal space. It is noted that the gap increases with Q deviating from the zone center. We believe that this Q dependence includes a valuable piece of the information for the anisotropy of the energy gap. On the other hand, the quasielastic scattering centered at (0 0 0.5) is observed at 4.2 K which is above T_c.

A clear superconducting gap at $2\Delta = 3.8k_BT_c$ has been observed in a recent study of tunneling spectroscopy of a thin UPd_2Al_3 film [13]. It is quite interesting that the temperature dependence of this gap, which is obviously a charge gap, is very similar to the one of the magnetic excitation gap observed in this study. Moreover, NMR studies reported the absence of the coherent peak and the T^3 dependence of the inverse relaxation time $1/T_1$ [10–12]. With the isotropic reduction of the Knight shift below T_c, it is concluded that d-wave pairing is realized

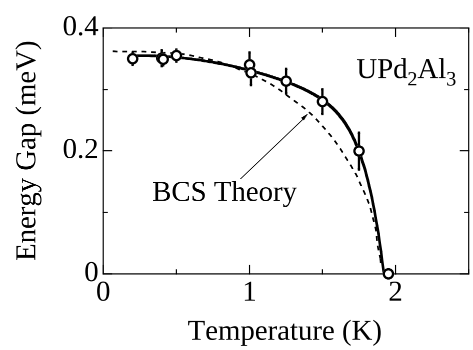

FIG. 3. Temperature dependence of the energy gap in UPd_2Al_3.

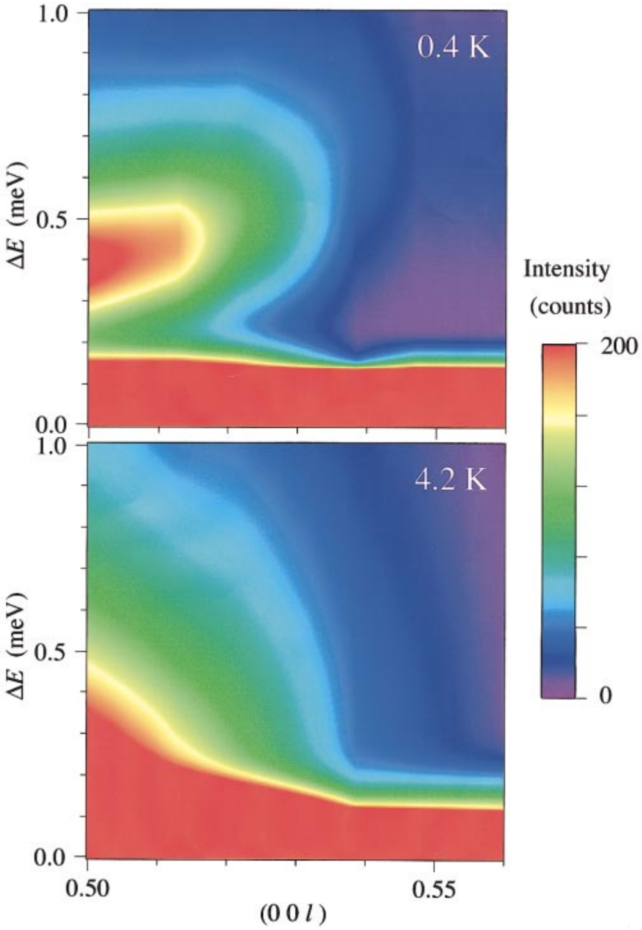

FIG. 4(color). Neutron scattering intensity map showing the excitation energy vs the momentum transfer Q along the $[00l]$ direction and the excitation energy in UPd_2Al_3.

in UPd_2Al_3 characterized by a line node of the energy gap with the gap $2\Delta = 5.5k_BT_c$. The present magnetic excitation gap of about $2\Delta = 2.2k_BT_c$ is smaller than the gap obtained from NMR study ($2\Delta = 5.5k_BT_c$) and the tunneling spectroscopy ($2\Delta = 3.8k_BT_c$). This might be due to the anisotropic gap.

Figure 5 shows the temperature dependence of the (0 0 0.5), (0 0 1.5), (1 0 0.5) magnetic peak intensities. The observed magnetic peak intensities increase continuously from the Néel temperature T_N down to T_c. Below T_c, the magnetic peak intensities turn to decrease with decreasing the sample temperature. On the contrary, the (001) nuclear peak intensity shows no change in the measured temperature range. No change of the nuclear peak intensity at T_c rules out the possibility that a small change of the magnetic peak intensity would be due to the slight change of the neutron absorption cross section and/or a small lattice distortion, associated with superconductivity. In addition, it was confirmed that the (0 0 0.5) peak intensity had a maximum at a lower temperature when magnetic field was applied, following the H-T phase diagram [3]. Therefore we conclude

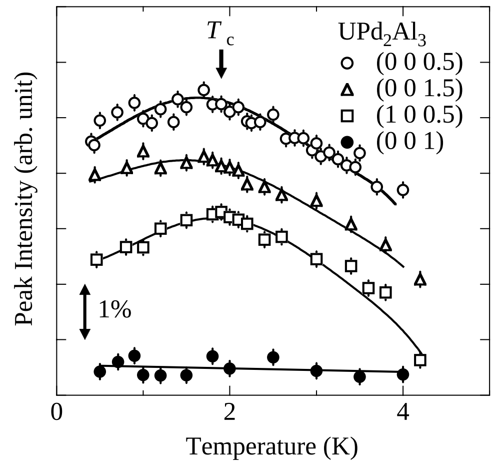

FIG. 5. Temperature dependence of the (0 0 0.5), (0 0 1.5), (1 0 0.5) magnetic peaks and the (0 0 1) nuclear peak intensities in UPd_2Al_3.

that the present suppressing of the magnetic intensities is due to superconductivity. This behavior might be understood in terms of the coupling between magnetic and superconducting order parameters. Here, we note that the accuracy of the previous studies (typically 1%) is not sufficient to observe this behavior [14,15]. From the similar observations in UPt_3 [16–18] and/or UNi_2Al_3 [19], and very recent observation in URu_2Si_2 [20], it is concluded that the coupling of the magnetic and superconducting order parameters would be a characteristic feature in the heavy fermion superconductor.

The data in Fig. 1 are plotted again in Fig. 6 to display the temperature dependence of the spin wave excitation which is observed as a broad peak at $\Delta E = 1.5$ meV. We found that the spin wave excitation energy and the

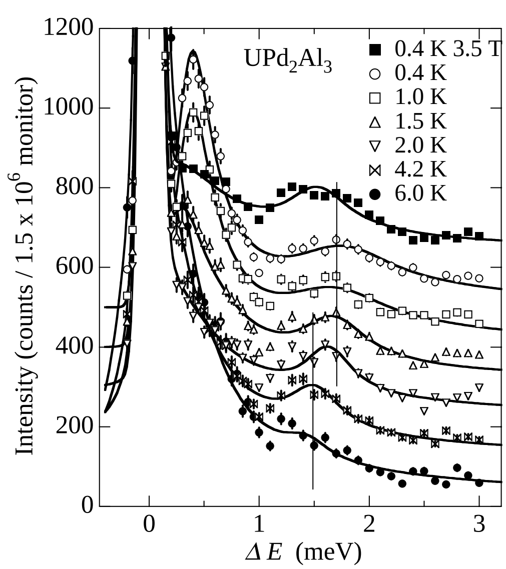

FIG. 6. Temperature dependence of the neutron inelastic scattering profile measured at $Q = (0\ 0\ 0.5)$ in UPd_2Al_3.

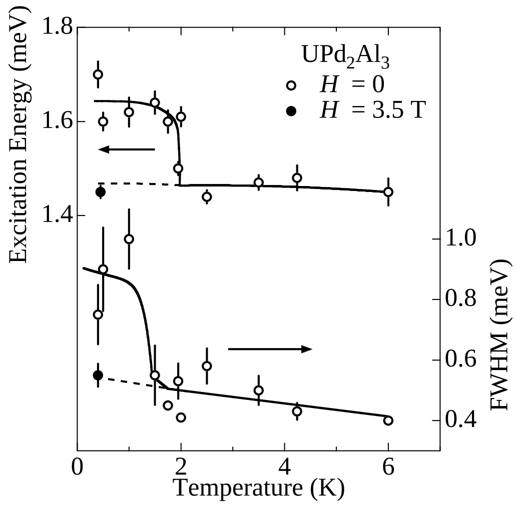

FIG. 7. Temperature dependence of the spin wave excitation energy and the linewidth of the spin wave excitation peak measured at $Q = (0\ 0\ 0.5)$ in UPd_2Al_3.

linewidth increase below T_c as shown in Fig. 7, respectively. This behavior is associated with the superconductivity. The spin wave excitation energy and the peak width are recovered to the value expected in the normal state as shown by closed circles in Fig. 7, when superconductivity is destroyed by applying the magnetic field of 3.5 T along the [110] direction. It should be noted that the spin wave excitation has relatively large excitation energy of about $10k_BT_c$. This result indicates that the strong coupling between magnetism and superconductivity has a large influence on the magnetic excitation with a fairly high energy compared to the superconducting energy gap.

In conclusion, we have observed a magnetic excitation gap associated with superconductivity. Suppression of the antiferromagnetic Bragg intensity and increases of the spin wave excitation energy and its linewidth indicate the strong coupling between magnetism and super conductivity.

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