Enhancement of Magnetic Fluctuations on Passing below T_c in the Heavy Fermion Superconductor UPd_2Al_3

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The heavy fermion superconductor UPd_2Al_3 exhibits the unusual combination of an antiferromagnetic phase transition, at $T_N = 14.3$ K, followed by a superconducting phase transition below 2 K without destruction of the ordered magnetic moment. Polarized inelastic neutron scattering reveals the presence of two coupled modes, both transverse to the sublattice magnetization. On passing into the superconducting phase an abrupt change is observed in the magnetic inelastic response. We show that it is reasonable to consider the superconducting state as arising out of interactions between quasiparticles which are strongly renormalized by the low-frequency exchange field. [S0031-9007(98)07531-0]

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The discovery of the heavy fermion superconductor UPd_2Al_3 [1] which exhibits both an antiferromagnetic phase transition, $T_N = 14.3$ K, and a superconducting phase transition below $T_c = 2$ K has aroused great interest. The compound crystallizes in the hexagonal $PrNi_2Al_3$ structure (space group $P6/mmm$) with lattice constants $a = 5.350$ Å and $c = 4.185$ Å at room temperature. Neutron and x-ray scattering measurements have revealed unusually large low temperature ordered moments of $0.85\mu_B$ which are coupled in ferromagnetic sheets in the basal plane; these ferromagnetic planes are then stacked along the c axis with a wave vector $\mathbf{Q}_0 = (0\,0\,0.5)$ [2–4]. The measured discontinuity in the heat capacity at T_c is large, $\Delta C = 1.2\gamma T_c$ ($\gamma = 140$ mJ/mole K^2) [1,5] and suggests that the superconducting ground state evolves out of interactions between quasiparticles located in strongly renormalized states in a low energy shell around the Fermi surface.

In the hope of finding a connection between antiferromagnetism and superconductivity there have been many investigations in this compound since its discovery. Changes in antiferromagnetic Bragg peak intensities on passing through T_c were reported [6]; however, these results are questioned by independent measurements using both neutron diffraction and resonant magnetic x-ray scattering [3,7]. Pioneering inelastic neutron scattering experiments under relatively coarse energy resolution [8] revealed no changes in the spectrum around the antiferromagnetic zone center on cooling through T_c. An additional low energy component to the spectral response was first reported by Sato *et al.* [9], and Metoki *et al.* [10] have reported a gap opening below T_c and the disappearance of the low energy response when a magnetic field is applied.

In this Letter we report on new, high resolution, inelastic neutron scattering experiments carried out on the IN14 spectrometer at the Institut Laue-Langevin, Grenoble. With full neutron polarization analysis we have been able to identify the *transverse* nature of the dominant fluctuations at all temperatures below T_N and the opening of a gap in the energy spectrum of magnetic fluctuations at the lowest temperatures (150 mK) in the superconducting state. The spectra consist of a damped spin wave coupled to a low energy mode. In the normal state we develop a phenomenological description of the scattering and show that a major portion of the low temperature linear coefficient to the electronic heat capacity γ, characterizing the heavy fermion normal state out of which superconductivity evolves, may be accounted for by the low energy antiferromagnetic fluctuations [11–13]. In the paramagnetic phase the model is used to estimate the value of T_N.

The sample, which was pulled from a carefully homogenized melt of high purity elements by the Czochralski method with a nominal composition of $UPd_{2.02}Al_{3.03}$ [14,15], has a superconducting transition temperature of 1.90 ± 0.07 K. It was mounted with the c^*-a^* axes in the scattering plane. The spectrometer was optimized for low energy scattering at about $\mathbf{Q}_0$ with a full width at half maximum resolution of 90 μeV in energy and 0.05 Å^{-1} in the wave vector. Uncertainties in calibration, made with a standard vanadium sample, lead to errors in the estimation of the absolute values of the parameters used to characterize the spectra of the order of 20%.

An overview of the spectral response as a function of neutron momentum and energy transfer, $\mathbf{Q} = (0\,0\,Q)$ and $\hbar\omega$, is given in the top four panels of Fig. 1, and an overview of the temperature-energy transfer response is given in the bottom frame. The four panels give data for temperatures at about T_N, in the normal antiferromagnetic state, near T_c and in the superconducting antiferromagnetic phase, respectively. The solid lines are fits described

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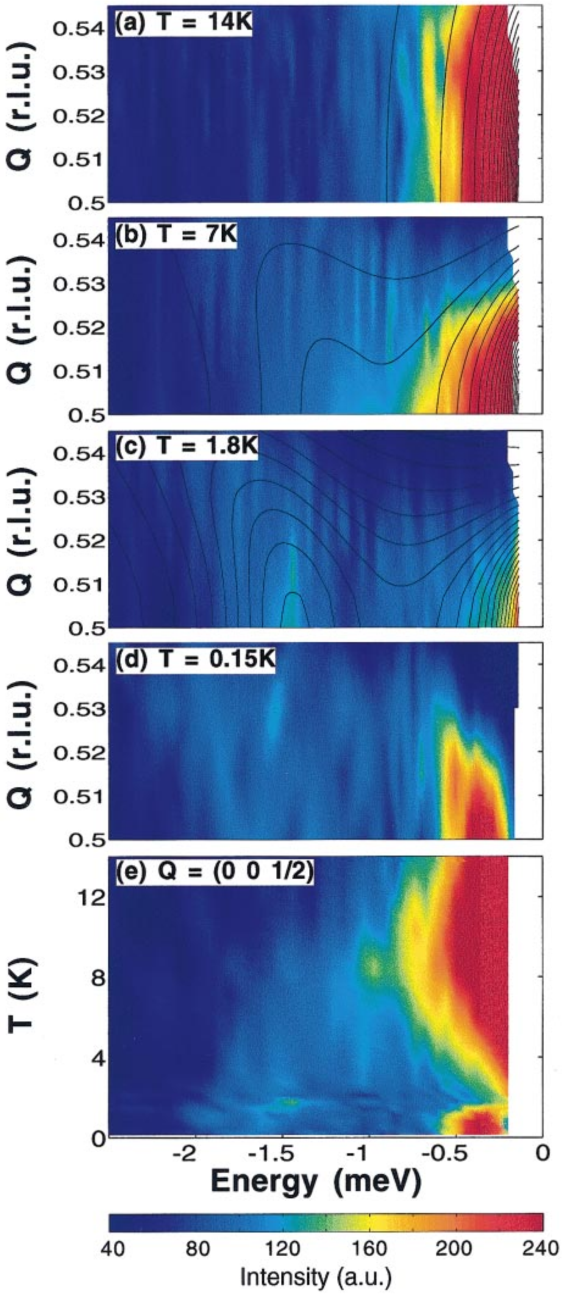

FIG. 1(color). Contour plots at four temperatures (as marked) of intensity as a function of Q, $\mathbf{Q} = (0\,0\,Q)$, and neutron energy loss. The section at the smallest energy transfers is inaccessible due to the incoherent elastic scattering, and, at $\mathbf{Q}_0$, due to the antiferromagnetic Bragg peak. The solid lines are calculated as described in the text. Since the form of the dynamical response below T_c is still in question, we show no calculated contours. Attention is drawn to (1) the development of two maxima in the spectral response, (2) the reduction in the q-space extent of the scattering at low energy on lowering the temperature, and (3) the appearance of a gap in the low energy response in the superconducting phase. Bottom panel: intensity at $\mathbf{Q}_0$ as a function of temperature and neutron energy loss. The strong renormalization of the spectrum is clearly seen below T_c.

later. At energy transfers of 1 meV or greater, in the antiferromagnetic state, one sees the inelastic (spin-wave) response [8]. The focus of this experiment is, however, on the considerable additional intensity present below the 1 meV energy transfer and its striking temperature dependence.

In order to determine the polarization of both the spin-wave and low energy scattering a full polarization analysis has been carried out. The sample was field cooled at 2.5 T into the antiferromagnetic phase and the field was lowered to 0.8 T at 8 K. Under these conditions, the (0 0 0.5) antiferromagnetic peak had an intensity 15 times lower than that observed in the similar configuration without polarization analysis. The flipping ratio at the antiferromagnetic peak was 2.5, in agreement with the magnetic phase diagram [3] in which the sublattice moments lie at 30° to the horizontal under the given conditions of temperature and applied field. The geometric constraints allow one to identify the fluctuations as being transverse to the sublattice magnetization and lying in the crystallographic $(a\text{-}b)$ plane. Data are shown at 10 K, with a background of 6 counts/mon = 1000 subtracted and corrected for the measured flipping ratio, in Fig. 2(a). In the superconducting state at 150 mK a similar experiment and analysis indicate that the polarization of the scattering does not change. This is shown in frame 2(b), where the strong low energy peak can be seen to join smoothly with the spin wave at higher energies. From the solid line, which is a scaled copy of data collected without polarization analysis (no applied field) in the same spectrometer configuration, e.g., frame 2(c), one sees that the field necessary for polarization analysis does not significantly distort the spectrum. Earlier work [8], unable to access the low energy response, has previously established the transverse nature of the spin wave. Having determined the polarization of the scattering, high resolution studies used to monitor the temperature evolution from 150 mK to 20 K were carried out with the full intensity of the unpolarized beam.

Above, and in the neighborhood of T_N, the response is quasielastic (Fig. 1) and may be characterized by a dynamic susceptibility of the form

$$\chi^{-1}(\overline{\mathbf{q}}, \omega) = \chi^{-1}(\overline{\mathbf{q}})\left[1 - \frac{i\omega}{\Gamma(\overline{\mathbf{q}})}\right], \qquad (1)$$

with $\overline{\mathbf{q}} = \mathbf{Q}_0 + \mathbf{q}$, $\chi^{-1}(\overline{\mathbf{q}}) = \chi^{-1}(\mathbf{Q}_0) + cq^2$, and $\Gamma(\overline{\mathbf{q}}) = u\chi^{-1}(\overline{\mathbf{q}})$. In the formulation, $\chi(\mathbf{Q}_0)$ is the static magnetic susceptibility at the antiferromagnetic wave vector, c is a torsional stiffness coefficient, q is the wave vector transfer as measured from the antiferromagnetic position, and u is a measure of the relaxation rate of the fluctuations. The lines (Fig. 1, top frame) have been calculated with the spectrometer resolution taken in the energy wave vector plane of the experiment and a dispersion surface based on the model for χ given in Eq. (1). Over the full experimental range (q, ω) the model calculation is in good agreement with the data; the parameters c, $\hbar u$, and $\chi(\mathbf{Q}_0)$ take the values 2.8 10^4 Å^2, 1.7 μeV, and 50, respectively.

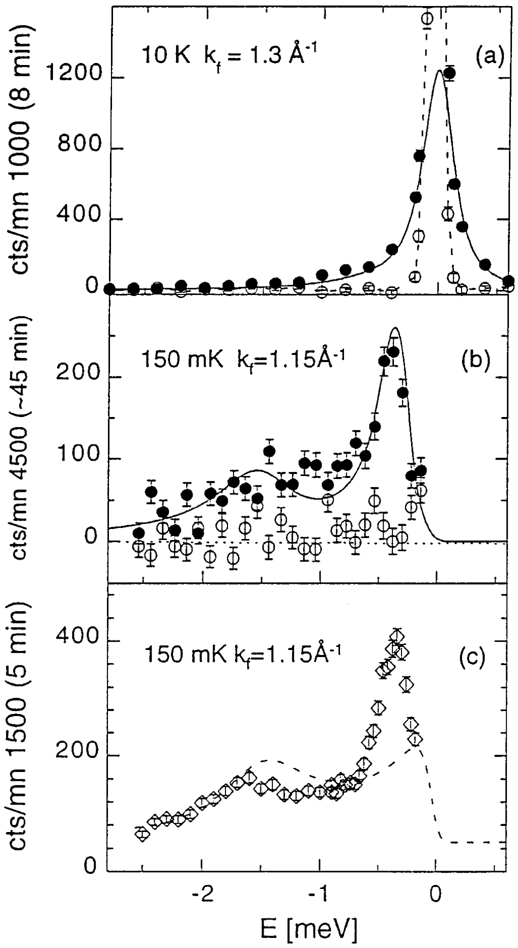

FIG. 2. Experimental data from UPd_2Al_3 at the antiferromagnetic wave vector $\mathbf{Q}_0$. The value of the fixed outgoing wave vector is given in each frame as is the temperature. (a) and (b) are taken with polarized neutrons and the transverse (longitudinal) response is shown as solid (open) circles. A background of 6 counts/mon = 1000 has been subtracted and the data corrected for the measured flipping ratio. In (b) the solid line is a scaled plot of the unpolarized data at 150 mK [frame (c)]. The dashed line is the zero level. The lower panel (open diamonds) is taken with an unpolarized incident beam. The dashed line is the calculated response in the normal antiferromagnetic state at this temperature.

Within the framework of Ginzburg-Landau mode coupling theory, in which a Gaussian approximation to the contribution from dynamic fluctuations in the first anharmonic term (quartic term) in the free energy density renormalizes the coefficient of the leading (quadratic) term, one has the following condition on T_N: $\sqrt{\langle m^2 \rangle} \approx M_0(Q_0)$. That is, the phase transition occurs when the root thermal variance of the local magnetization induced by fluctuations is of the order of the low temperature magnetization [11–13]. The fluctuations are integrated up to a thermal cutoff given by $k_B T_N = \hbar\Gamma(\overline{\mathbf{q}})$ which yields the estimated $T_N = 16$ K in reasonable accord with the experimental value.

For temperatures below T_N, as shown, for example, at 7 K in Fig. 1, the solid lines result from calculations in which the dynamical susceptibility is given by the sum of two modes, χ_1 and χ_2, in which all internal interactions have been included apart from their mutual coupling which is effected through a mean field parameter λ [16]

$$\chi = \frac{\chi_1 + \chi_2 + 2\lambda\chi_1\chi_2}{1 - \lambda^2\chi_1\chi_2}\,. \tag{2}$$

The low-frequency response is represented as a quasielastic excitation as in (1) above, together with a damped spin wave at pole $\omega_0(q)$ at high energies. This damped pole takes the form

$$\chi_2^{-1}(\overline{\mathbf{q}},\omega) = \chi_2^{-1}(\overline{\mathbf{q}}) \times \left[1 - \frac{\omega[\omega + i\Gamma_2(\overline{\mathbf{q}})]}{\omega^2(q) - i\Gamma_2(\overline{\mathbf{q}})[\omega + i\Gamma_2(\overline{\mathbf{q}})]}\right], \tag{3}$$

with $\hbar\omega(q) = \hbar\omega_0 + Dq^2$ and where $\chi_i^{-1}(\overline{\mathbf{q}})$ and $\Gamma_i(\overline{\mathbf{q}})$ have the same functional dependence as in (1). The spin-wave pole gap at the antiferromagnetic zone center is given by $\hbar\omega_0$ and D is the stiffness. This dispersion, local to the zone center, joins the previously established [8] dispersion at about $q = 0.06$ Å^{-1}.

In modeling the data over the range $T_c < T < T_N$, the evolution of linewidth and intensity over the measured energy and wave vector intervals may be consistently accounted for by a natural softening of the spin-wave pole and energy gap, together with a thermal renormalization of $\chi_{1,2}$. Near T_N the torsional stiffness of the quasielastic (c_1) and the spin-wave mode (c_2) take similar values and join smoothly with the value of c in the paramagnetic phase; however, below about 10 K the quasielastic stiffness c_1 increases by almost an order of magnitude [17]. The microscopic origin of such behavior remains to be understood. For $T < T_N/2$, the parameters become essentially temperature independent with the following values: $\hbar u_1 = \hbar u_2 = 1.4$ μeV, $c_1 = 1.510^5$ Å^2, $c_2 = 0.310^5$ Å^2, $D = 120$ meV Å^2, $\hbar\omega_0 = 1.4$ meV, $\chi_1^{-1} = 280$, and $\chi_2^{-1} = 230$. The coupling parameter $\lambda = 200$. This gives an enhancement at zero frequency and $\mathbf{Q}_0$ of $\sim$3.5 to the total susceptibility.

We have estimated the linear coefficient of the heat capacity, in the normal state as extrapolated to $T = 0$ K, contributed by the spectral weight of the antiferromagnetic fluctuations. Within the Ginzberg-Landau model used to estimate T_N, and assuming the spectra at the lowest energy in the normal state to be dominated by the quasielastic mode, we obtain $\gamma = 0.1$ J/mole K^2. Although it is difficult to have a precise figure, since it depends on a cutoff parameter of integration [11–13], it is close to, and certainly within an order of magnitude of, the experimental value. This strongly supports the idea that the dominant renormalization, yielding the massive quasiparticles which give rise to the superconducting ground state, may be considered as arising from the low-frequency exchange field.

On cooling below T_c, at the lowest temperatures, a reshaping of the spectrum occurs (see Fig. 1). This cannot be accounted for by the fall in the Bose population factor, as is illustrated in Fig. 2(c), where the dashed line is the projected scattering profile in the normal antiferromagnetic state at 150 mK. It is apparent that the low energy response has changed in a *qualitative* manner. This enhanced response in the superconducting state is seen around the antiferromagnetic reciprocal lattice vectors (r.l.v.) with a half width of 0.02 reciprocal lattice units (r.l.u.) (see Fig. 1).

To our knowledge, no forms of the dynamical magnetic susceptibility have been derived in the antiferromagnetic superconducting phase. However, if we assume that a general pairing mechanism survives in the antiferromagnetic superconducting state exhibiting (i) coherence of the ground state wave function and (ii) an energy gap for at least the majority of the low energy excitations, some qualitative comments may be made. First, the observed *enhanced* diffusion at about 0.4 meV ($\sim 2.4kT_c$) is in accord with the absence of an NMR coherence peak in the superconducting state, which has been interpreted as evidence for a sign change in the gap function [18,19]. Second, the formation of at least a partial gap in the magnetic excitation spectrum is in accord with the abrupt fall in heat capacity [1,5] and NMR relaxation rate [18] at low temperatures. Thus, the low temperature peaking of intensity at finite energy may naturally arise from the superconducting correlations in the ground state. Recalling the wave vector specificity of the scattering, the appearance of inelastic intensity around the antiferromagnetic r.l.v. raises the possibility that the sign change in the gap function [18] may be associated with the characteristic wave vector $\mathbf{Q}_0$. A model of superconducting order with antiferromagnetic periodicity has been advanced previously on the basis of general symmetry arguments in heavy electron systems [20]. Within such a scenario the width of the response around a r.l.v. gives an inverse measure of the spatial coherence of the superconducting order parameter. This is estimated to be about 100 Å (Fig. 1) and would be in agreement with indirect bulk measurements [1].

In summary, the main finding of this work is the observation, by means of polarized inelastic neutron scattering, of two, coupled, components in the low energy inelastic response of UPd_2Al_3 below T_N. They are *both* identified as being transverse fluctuations to the sublattice magnetization vector. One corresponds with the spin wave observed previously, while the second response appears at low energies with a characteristic frequency below that of the spin wave [21]. The low energy mode is strongly renormalized by the transition to the superconducting state, where a gap appears to develop in the spectrum of transverse excitations. It will be interesting to see if the twin threads, the identification of low energy antiferromagnetic fluctuations with the formation of the heavy quasiparticles that make the transition into the superconducting state, and the possible antiferromagnetic nature of the superconducting order parameter survive further experimental and theoretical analyses to provide us with spectroscopic evidence linking the two phenomena.

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