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Spin Fluctuations in the Antiferromagnetic Heavy-Fermion System U_2Zn_{17}

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Inelastic neutron scattering from the antiferromagnetic ($T_N=9.7$ K) heavy-fermion system U_2Zn_{17} reveals magnetic fluctuations which are highly localized in reciprocal space and broad in frequency. At higher energies, the fluctuations persist almost unchanged up to temperatures well above T_N . Analysis of the neutron and bulk susceptibility data in terms of Kondo-type single-ion response functions modified by Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions between ions indicates that it is the temperature dependence—anticipated theoretically—of the effective RKKY coupling which drives the magnetic phase transition in U_2Zn_{17} .

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U_2Zn_{17} is an antiferromagnet with many remarkable properties.¹⁻⁴ For temperatures well below its Néel temperature ($T_N=9.7$ K), the magnetic specific heat consists of a large linear term, γT ($\gamma=200$ mJ/mole K^2),¹ generally associated with *paramagnetic* heavy-fermion systems, and a cubic contribution, αT^3 , characteristic of antiferromagnetic spin waves. The latter is unusual in that the typical spin-wave energy associated with α is 120 K, which exceeds T_N by more than an order of magnitude. Neutron diffraction² shows that the ordered moment ($0.8\mu_B$) is considerably less than the paramagnetic moment [(2.2–3.3) μ_B] deduced from high- T susceptibility data.^{1,3} Nonetheless, the magnetic structure is exceedingly simple: The magnetic unit cell, which contains two oppositely polarized U ions, is identical to the nuclear unit cell.

To observe the magnetic excitations which give rise to the anomalous properties of U_2Zn_{17} , we have performed an inelastic-neutron-scattering study. The principal result is that in the ordered state, the magnetic fluctuations, while very broad in energy, are sharply localized in reciprocal space. The fluctuations persist to temperatures above T_N , and finally disappear for $T>18$ K, where the maxima in the bulk susceptibility, resistivity, and Hall coefficient occur.^{1,3}

Our crystal of U_2Zn_{17} was found in a melt produced at 1050 °C from the appropriate amounts of U and Zn. Its

approximate dimensions are $5\times 7\times 8$ mm³. We attached the crystal to the cold finger of a pumped ⁴He cryostat, which in turn was mounted on the triple-axis spectrometer TAS-6 at Risø National Laboratory. The horizontal scattering plane of the spectrometer coincided with the ($h0l$) zone of the rhombohedral (space group⁵ $R\bar{3}m$) crystal; throughout this Letter, we use hexagonal indices, with $a^*=4\pi/\sqrt{3}a=0.811$ Å⁻¹ and $c^*=2\pi/c=0.479$ Å⁻¹, to label points in reciprocal space. Our sample displays macroscopic sixfold symmetry, which implies that it consists of twins related to each other by reflection through the (001) plane.

Pyrolytic graphite crystals, set for their (002) reflections, were the monochromating and analyzing elements of our spectrometer, operated with the final energy fixed at 7.5 meV. The energy resolution, as measured for incoherent elastic scattering, was 0.4 meV, full width at half maximum. Because the source of the neutrons was the Risø National Laboratory H₂ moderator, higher-order contamination of the signal was sufficiently small to make the use of a filter superfluous.⁶

We consider first the magnetic fluctuations in the ordered state. Figure 1 shows the scattered intensity, $S(Q, \omega)$, probed in a series of constant-energy scans for momentum transfers $Q=(h, 0, 3-h)$; note that there is a magnetic Bragg peak at $Q_0=(1, 0, 2)$, where $h=1$. For ordinary Heisenberg antiferromagnets (e.g., MnF₂),

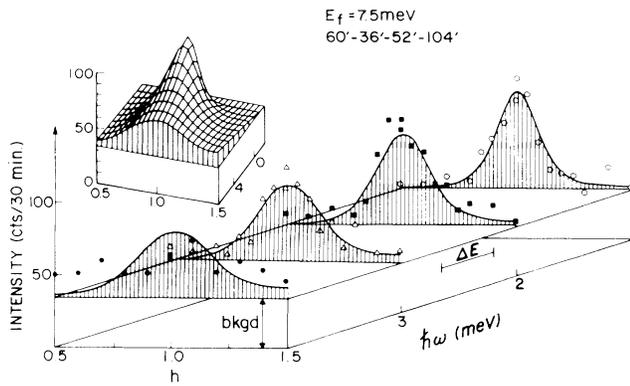


FIG. 1. Constant-energy scans along $(h, 0, 3-h)$ in the ordered phase at $T=2$ K. The solid lines are the result of the three-parameter fit described in the text. Inset: The corresponding model scattering function in a perspective view.

such scans would each contain two maxima, at $h=1 \pm q$, derived from spin waves propagating in opposite directions. When q is small, $cq = \omega$ where c is the spin-wave velocity, typically of order $k_B T_N / (\sqrt{3}\hbar)$. Furthermore, $S(Q, \omega)$ essentially vanishes for $\hbar\omega \gtrsim k_B T_N$. From Fig. 1 it is apparent that U_2Zn_{17} is unusual. The surface above the $h-\omega$ plane defined by $S(Q, \omega)$ contains only a single ridge parallel to the ω axis, instead of two ridges—due to propagating spin waves—converging at the Bragg point, Q_0 , with $h=1$ and $\omega=0$. In addition to being sharply peaked at Q_0 for $\hbar\omega \lesssim 3$ meV, $S(Q, \omega)$ is appreciably above background for the highest energy (4 meV $\cong 4.8k_B T_N$) at which measurements were performed. Data similar to those in Fig. 1 have been obtained at much larger energy transfers ($\hbar\omega > 10$ meV) for both UN⁷ and Cr.⁸

As is known from studies of Cr and its Mn alloys,⁹ the finite instrumental resolution can merge the two spin-wave peaks (observed in constant $\hbar\omega$ scans) at $q = \pm \omega/c$ into a single peak at Q_0 . Nonetheless, attempts to fit the data in Fig. 1 by the resolution-corrected $S(Q, \omega)$ appropriate for undamped spin waves met with failure. Furthermore, in the absence of lifetime effects, the maximum resolvable spin-wave velocity for our spectrometer is roughly 30 meV \AA , three times larger than the spin-wave velocity inferred from the cubic term in the low-temperature heat capacity¹ of U_2Zn_{17} and eight times larger than $k_B T_N / \sqrt{3}$.

Having described the low- T properties of U_2Zn_{17} (note that for the data of Fig. 1, $\hbar\omega \gg k_B T$), we turn to the temperature-dependent behavior. Neutron diffraction from our single crystal shows that U_2Zn_{17} undergoes a quite ordinary antiferromagnetic transition at $T_N = 9.7$ K: the magnetic Bragg intensity measured at (102) is proportional to $(T - T_N)^{2\beta}$ where $\beta = 0.36 \pm 0.02$, close to the value expected for three-dimensional, short-range coupled x - y magnets. What is more interesting is that

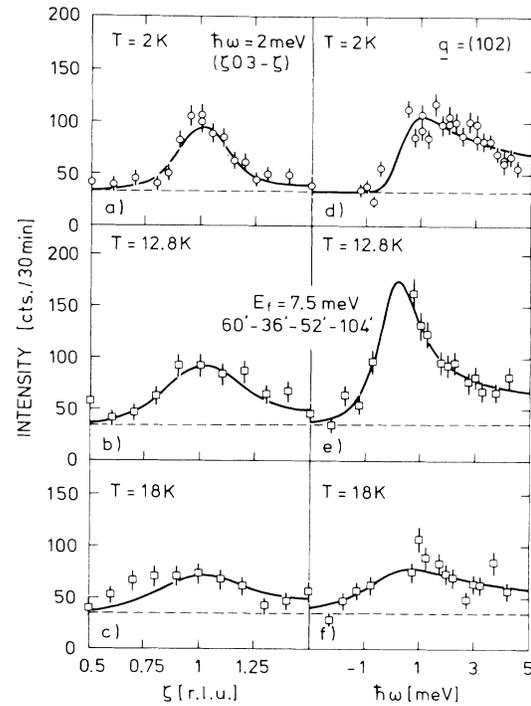


FIG. 2. Constant $\hbar\omega = 2$ meV and constant $Q = (1,0,2)$ scans at temperatures above and below T_N . The solid lines result from the model calculations described in text.

there is substantial critical scattering, even at energies comparable to $k_B T_N$. For example, constant- $Q = Q_0$ spectra, such as those in Figs. 2(d)–2(f), are considerably enhanced at $\hbar\omega \sim 1$ meV for $T \approx T_N$. At higher frequencies ($\hbar\omega \gtrsim 1.5$ meV), however, constant- $\hbar\omega$ scans show that antiferromagnetic fluctuations persist to temperatures as high as 14 K [see Figs. 2(a)–2(c)].

Because of the absence of sharp magnetic excitations, conventional models involving crystal-field levels and exchange couplings cannot account for the spin dynamics of U_2Zn_{17} . Nevertheless, we have been able to identify a simple model which does describe our data. The essential ingredients of this model¹⁰ are (1) the excitation spectrum for a single f -like moment, because of its (Kondo) interaction with the conduction electrons, and (2) the Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling, also due to interactions with the conduction electrons, between f -like moments at different lattice sites. From extensive experimental and theoretical work,¹¹ it is known that the form $\chi_0(\omega) = \chi_0 \Gamma / (\Gamma - i\omega)$ gives a good description of the frequency-dependent susceptibility for a single Kondo impurity. Within the random-phase approximation, the generalized susceptibility for a lattice of coupled impurities is then

$$\chi(Q, \omega) = \sum_{\pm} \frac{\chi_0(\omega)(1 \pm \cos\phi_Q)}{1 - \chi_0(\omega)[J(Q) \pm |J'(Q)|]}, \quad (1)$$

where $J(Q)$ and $J'(Q) = |J'(Q)| \exp i\phi_Q$ are the Fourier-transformed intrasublattice and intersublattice RKKY interactions. The corresponding neutron-scattering cross section is

$$\partial^2 \sigma / \partial \Omega \partial \omega = \gamma_0^2 (k_f / k_i) |f(Q)|^2 [1 - \exp(-\hbar \omega / k_B T)]^{-1} [\chi''(Q, \omega) / \pi], \quad (2)$$

where k_i and k_f represent the wave vectors of the incident and scattered neutrons, $\gamma_0^2 = (0.073 \text{ b}) / \mu_B^2$, and $f(Q)$ is the form factor of the magnetic electrons. We assume that the only appreciable RKKY coupling, J'_0 , is isotropic and acts between nearest neighbors on different sublattices. The corresponding expression for the imaginary part of $\chi(Q, \omega)$ is then

$$\chi''(Q, \omega) = \sum_{\pm} \frac{\omega \chi_0 \Gamma_{Q_{\pm}} (1 \pm \cos \phi_Q)}{\Gamma_{Q_{\pm}}^2 + \omega^2}, \quad (3)$$

where $\Gamma_{Q_{\pm}} = \Gamma [1 \mp \chi_0 |J'(Q)|]$. We thus have a three-parameter form for the entire Q - and ω -dependent scattering function. The solid lines in Figs. 1 and 2 represent the theoretical cross section computed with use of the values for χ_0 , Γ , and J'_0 which yield the best description of the data. Although we cannot claim that we have determined the only possible model for $\partial^2 \sigma / \partial \Omega \partial \omega$, it is obvious that the exceedingly simple form chosen for $\chi(Q, \omega)$ accounts for the spectra both above and below T_N .

To study the temperature dependence of χ_0 , Γ , and J'_0 , we have performed two sets of fits, the first with the three parameters unconstrained and the second with the extrapolated $\chi(q=0, \omega=0)$ held equal to the temperature-dependent bulk susceptibility¹ scaled to match the neutron result at 2 K. The two procedures yield the same qualitative results. Figure 3 displays the temperature dependence of χ_0 , J'_0 , and $\hbar\Gamma$ obtained in the second series of fits. At all T , the system nearly satisfies the instability condition, $1 = \chi_0 |J'(Q_0)|$, which is associated with the onset of magnetic order. Not surprisingly, $\chi_0 |J'(Q_0)|$ is closest to unity at $10 \text{ K} \cong T_N$. Correspondingly, the order-parameter relaxation rate $\Gamma_0 = \Gamma [1 - \chi_0 |J'(Q_0)|]$, measured at $Q_0 = (1, 0, 2)$, approaches zero as T tends towards T_N from both above and below. Thus, U_2Zn_{17} displays critical slowing down, as does any other system undergoing a second-order phase transition. However, U_2Zn_{17} is highly unconventional in other respects. In particular, for ordinary magnets, a rising single-ion susceptibility χ_0 drives $|J'(Q_0)| \chi_0$ to unity and Γ_0 to zero. For U_2Zn_{17} , the rise in $|J'(Q_0)| \chi_0$ for $T < 1.5 T_N$ is due to an increase in J_0 rather than χ_0 . The remarkable result that the effective RKKY coupling grows with decreasing T is consistent with recent theoretical work¹² on the two-impurity Kondo problem.

Another feature of Fig. 3 is that the characteristic energy $\hbar\Gamma$ for the single-ion fluctuations increases greatly as T passes through T_N . Thus, U_2Zn_{17} is not analogous to typical singlet ground-state systems, where the (crystal-field) parameters specifying single-ion dynamics

are temperature independent. However, the evolution of $\hbar\Gamma$ is not so surprising when we consider that for heavy-electron systems, $\hbar\Gamma$ is inversely proportional to the density $n(\epsilon_F)$ of quasiparticle states at the Fermi level,¹³ and that spin-density-wave transitions in metals generally reduce $n(\epsilon_F)$. Indeed, the specific-heat measurements¹ indicate that $n(\epsilon_F)$ is ~ 2.5 times smaller in the ordered phase of U_2Zn_{17} , in agreement with the increase in the parameter $\hbar\Gamma$ extracted from our data.

In summary, we have performed elastic- and inelastic-neutron-scattering measurements on the heavy-electron antiferromagnet U_2Zn_{17} . The order parameter develops in the manner expected for short-range-coupled x - y magnets. The magnetic fluctuations both above and

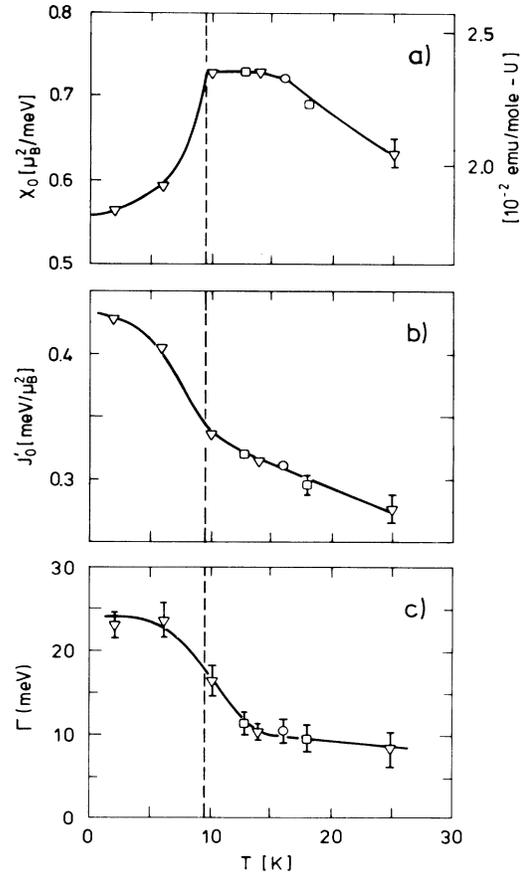


FIG. 3. Temperature dependence of the model parameters obtained from fits described in text. Different symbols correspond to different experimental runs. Where no error bars are shown, the errors are comparable to or smaller than the symbols. Solid lines are guides to the eye.

below T_N can be described in terms of a surprisingly simple model, where U moments with individual Kondo-impurity-type dynamics are coupled to their nearest neighbors via an effective RKKY interaction, J'_0 . The phase transition appears to be driven not by an increasing single-ion susceptibility, χ_0 , as for conventional singlet ground-state systems, but by a *rise* in J'_0 . Thus, U_2Zn_{17} is a system where the data can be described in terms of a model where the theoretically anticipated¹² renormalization of the RKKY interaction occurs. It will be interesting to establish whether the same result obtains for other systems with strong interactions between local moments and conduction electrons.

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