Wave-Vector– and Magnetic-Field–Dependent Spin Fluctuations in the Heavy-Fermion System $CeCu_6$

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We have performed neutron-scattering measurements on a single crystal of $CeCu_6$ in applied magnetic fields from 0 to 6.4 T. The observed momentum-dependent magnetic scattering implies the existence of short-range antiferromagnetic correlations. While it induces no resonances, the field broadens the spectra considerably at $T = 0.4$ K. An effective Hamiltonian which incorporates both single-impurity Kondo properties and antiferromagnetic Ruderman-Kittel-Kasuya-Yosida interactions describes our data.

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While heavy-fermion systems[1] share enhanced specific heats and Pauli susceptibilities with ordinary, dilute Kondo systems, they display qualitatively different transport properties. In particular, as the temperature T is reduced toward zero, the resistivities of heavy-fermion systems decrease rather than increase. Transport measurements performed in magnetic fields have been even more valuable as probes of the coherence effects which set heavy-fermion systems apart from dilute Kondo alloys.[2] In this paper, we present the first inelastic-neutron-scattering study of a heavy-fermion system, $CeCu_6$,[3] in magnetic fields[4] comparable to $kT_F/g\mu_B$, where T_F is the Fermi temperature deduced from bulk data. The inelastic spectra depend strongly on both wave vector and magnetic field, and cannot be accounted for in terms of either simple Fermi-liquid theory or the single-impurity Kondo model.

$CeCu_6$ is a heavy-fermion system, with an exceptionally large γ (≈ 1500 mJ/mole-K^2),[3] which displays neither magnetic order nor superconductivity to temperatures as low as 10 mK. The magnetic susceptibility, which is highest when measured along the c axis of this monoclinic[5] (below 220 K) material, saturates at a correspondingly high value of 0.1 emu/mole of Ce for $T \lesssim 4$ K. Inelastic neutron scattering[6] indicates that the powder-averaged spin-fluctuation spectrum, dominated by a quasielastic feature with a characteristic energy of order 1 meV, also ceases to evolve for $T < 4$ K. Nonetheless, specific-heat and resistance measurements carried out for applied magnetic fields[2,7] show new behavior at temperatures considerably below 4 K. In particular, the magnetoresistance[2] changes sign at ≈ 0.5 K; on cooling it becomes positive, as for ordinary metals, instead of negative, as for dilute Kondo systems. Furthermore, at $T = 0.5$ K, a 5.5-T field applied parallel to the c axis suppresses the specific heat by 50%, while for $T > 2$ K, such a field has no effect.[7]

We performed our experiments using a triple-axis spectrometer, with a quasielastic energy resolution of 0.3 meV, full width at half maximum, at a thermal beam of the Brookhaven National Laboratory high-flux reactor. The resistance ratio $\rho(T = 1.5\ \text{K})/\rho(T = 15\ \text{K})$ for a similarly prepared crystal is 0.44.[8] The $CeCu_6$ crystal was grown by the Czochralski method by use of pure copper and cerium as starting materials. Its shape is roughly conical, with a length of ~ 5.5 cm and a base of radius ~ 2 cm. We mounted the crystal in a ^{3}He Dewar equipped with a superconducting magnet. The orthorhombic c axis was parallel to the field direction, and perpendicular to the horizontal scattering plane of the spectrometer. In this paper, we label scattering vectors in terms of reduced lattice units for the room temperature, orthorhombic structure of $CeCu_6$, with $a^* = 0.776$ Å^{-1}.

Figure 1 shows inelastic spectra obtained for two different momentum transfers at 0.4 K. In addition to the resolution-limited nuclear incoherent scattering at $\hbar\omega = 0$, clearly visible inelastic scattering exists for 0.3 meV $< \hbar\omega <$ 1.5 meV. The latter is considerably narrower and more intense near (100) than (200). A field of 6.4 T largely suppresses the inelastic scattering, which confirms its magnetic origin. In contrast to what occurs for ordinary local moment systems, the field does not induce a resonance at $\hbar\omega = g\mu_{\text{eff}}H$

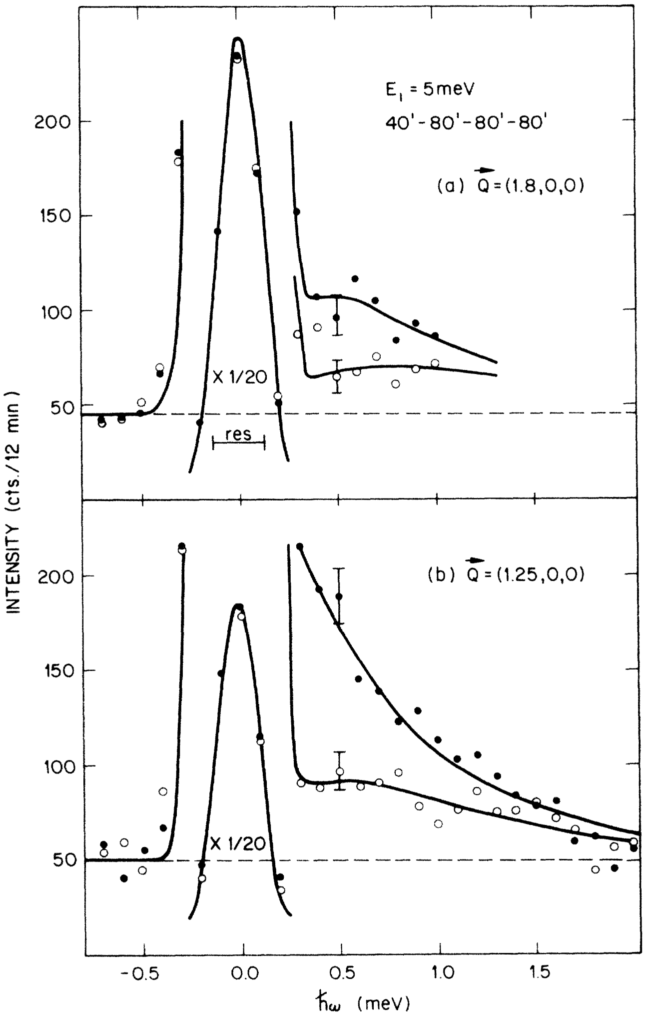

FIG. 1. Constant-Q energy spectra for $H=0$ (closed circles) and 6.4 T (open circles) at $T=0.4$ K. Background is indicated by dashed line; solid lines are derived from fits to theoretical forms described in text.

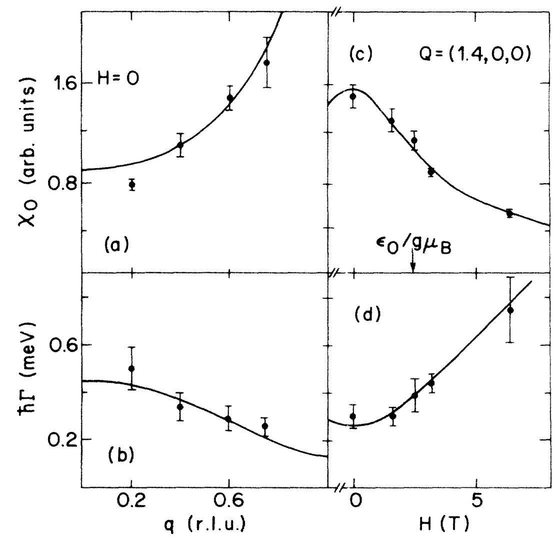

FIG. 2. Dependence of energy scale ($\hbar\Gamma$) and zero-frequency susceptibility (χ_0), obtained from fits to constant-Q spectra, (a) and (b) on reduced momentum $\mathbf{Q}=(2-q,0,0)$ for $T=0.4$ K and $H=0$, and (c) and (d) on H, for $T=0.4$ K and $\mathbf{Q}=(1.4,0,0)$. Solid lines are described in text.

($=0.74$ meV if $g=2$ and $\mu_{\rm eff}=1\mu_{\rm B}$); it simply broadens and reduces the magnetic response.

We parametrize our data using a simple single-pole approximation to the dynamical susceptibility,

$$\chi(q,\omega)=\chi_0\Gamma/(\Gamma+i\omega). \quad (1)$$

Note that $\hbar\Gamma$ is a q-dependent energy scale, while χ_0 is the q-dependent zero-frequency susceptibility. The corresponding neutron-scattering cross section is

$$\frac{\delta^2\sigma}{\delta\Omega\,\delta\omega}=\gamma_0^2\frac{k_f}{k_i}\frac{1}{1-\exp-\beta\hbar\omega}\frac{\chi''(q,\omega)}{\pi}, \quad (2)$$

which reduces to $\gamma_0^2\chi_0\omega\Gamma/(\Gamma^2+\omega^2)$ for $kT<<\hbar\omega$. For comparison with experiment, we convolve Eq. (2) with the instrumental resolution function and add a resolution-limited Gaussian peak to account for the incoherent scattering. The solid lines in Fig. 1 represent the resulting theoretical form, evaluated by use of the parameters Γ and χ_0 which yield the best fit to the data over the range ($\hbar\omega\geqslant 0.3$ meV) where the incoherent-scattering contribution is small. Figure 2 shows $\hbar\Gamma$ and χ_0 plotted against q [$\mathbf{Q}=(2-q,0,0)$] for $H=0$, and against H for $\mathbf{Q}=(1.4,0,0)$, respectively. As revealed by inspection of Fig. 1, χ_0 is largest for $q=1$ and smallest at $q=0$, while the converse is true for the energy scale $\hbar\Gamma$. Furthermore, the product $\hbar\Gamma\chi_0$ remains roughly constant (0.42 ± 0.04 in the units of Fig. 2), which suggests that at high frequencies ($\hbar\omega\gtrsim 1$ meV) $\chi''(q,\omega)$ is essentially q and H independent.

Fermi-liquid theory has been useful for relating various properties of heavy-fermion compounds to each other. In particular, the Fermi-liquid parameters obtained from fits of the temperature-dependent specific heat also account for quantities such as the width $\hbar\Gamma$ measured by inelastic neutron scattering from polycrystalline samples.[9] It is worth noting, however, that the Fermi-liquid description has been more successful for U-based than for Ce-based heavy-fermion systems. Fermi-liquid theory has several specific predictions[10] for $\chi''(q,\omega)$. Firstly, $\chi''(q,\omega)$ narrows as q decreases, indeed, if we were to use Eq. (1) to describe $\chi''(q,\omega)$, $\hbar\Gamma\sim q$ as $q\to 0$. Second, χ_0 depends weakly on q for $q\lesssim p_{\rm F}$, where $p_{\rm F}$ is the Fermi momentum; if ferromagnetic coupling is included as for Pd or ^{3}He, χ_0 will be enhanced for small q. This last result is to be distinguished from the fact that $\int_0^\infty \chi''(q,\omega)d\omega$ vanishes as $q\to 0$, which follows because the exclusion principle leads to antiferromagnetic correlations in Fermi liquids. Our data for $CeCu_6$ display none of the features associated with simple Fermi liquids. Specifically, χ_0 increases with q and $\hbar\Gamma$ decreases with q. Furthermore, given that $\hbar\Gamma\chi_0$ is roughly q independent, it is unlikely that $\int_0^\infty \chi''(q,\omega)d\omega$, which corresponds to the instantaneous spin fluctuations, vanishes as $q \rightarrow 0$.

The most common starting point for theoretical investigations of rare-earth and actinide compounds has been the Anderson Hamiltonian, which describes a partially filled (metallic) conduction band interacting with localized electrons subject to a Coulombic repulsion U. Recently, several groups[11] have used a variational wave function, similar to that of Gutzwiller[12] for the Hubbard model, to describe the ground state of the Anderson Hamiltonian. The principal conclusion is that the localized and delocalized bands are hybridized near the Fermi energy, with the result that $\chi''(q,\omega)$ consists of two parts: a Fermi-liquid contribution of the type described above, and an interband contribution. The interband contribution is derived from a crossing of the hybridization gap,[13] which will be largest for small reduced-momentum transfers q and smallest for q of order p_F. For our data to be consistent with the Gutzwiller picture, (1) the Fermi-liquid contribution to $\chi''(q,\omega)$ must be invisible and (2) $p_F \cong a^*$. While, if pressed, one may argue for both (1) and (2), it is important to note that the Gutzwiller approaches are mean-field theories where the configurations at different sites are correlated only in a Bloch-wave sense. An *effective* Hamiltonian[14] which incorporates both the single-impurity Kondo properties (also built into the Gutzwiller theories) and the correlations between sites, brought about by renormalized Ruderman-Kittel-Katsuya-Yosida interactions (not accounted for in the Gutzwiller approaches), is

$$H_{\mathrm{eff}} = \Delta \sum_i S_i s_i + \sum_{i \neq j} K_{ij} S_i S_j. \tag{3}$$

The S_i and s_i correspond to the spins of localized and conduction electrons, respectively, at the sites i. If the coupling K_{ij} vanishes, the ground state for each site is a singlet, separated by an energy Δ from a triplet excited state. The Ruderman-Kittel-Katsuya-Yosida coupling K_{ij} will mix the singlets with triplets at other sites. For Bravais lattices, the mixing will lead to a q-dependent singlet-triplet splitting[13] at $(T=0)$,

$$\Delta(q) = \Delta[1-(K/\Delta)\cos(4\pi q)]^{1/2}, \tag{4}$$

where q is in reciprocal lattice units and we have assumed that $K_{ij}=K$, with $|K|<\Delta$, couples only nearest neighbors. The effective-boson approximation[15] which yields (4) also gives

$$\chi_0(q) \sim 1/\Delta(q) \tag{5}$$

at $T=0$. The solid lines in Figs. 2(a) and 2(b) represent Eqs. (4) and (5) with $\Delta = 0.33$ meV and $K=0.28$ meV.[16] As is apparent from Fig. 2, H_{eff} provides the basis for a quite adequate, but by no means unique, parametrization of our data. We emphasize that $\Delta(q)$ does not correspond to a well-defined excitation as in the problem where Δ is generated by (fixed) crystal fields. Instead, $\Delta(q)$ is a characteristic energy scale for spin fluctuations with wave number q, in the same sense that for the single-impurity Kondo problem $(K=0)$, Δ is the energy scale, of order the Kondo temperature, for fluctuations of the impurity spin.

We turn now to the effects of the external magnetic field. The important result is that, as pointed out above, there is no well-resolved resonance line. For ordinary crystal-field doublets with finite spin-fluctuation times, we would find such resonance lines with widths comparable to the width of the quasielastic scattering for $H=0$. For a free Fermi liquid, a field creates separate up-spin and down-spin subbands, and so, for small q (i.e., $q \ll p_F$), $\chi''(q,\omega)$ will be dominated by a sharp peak due to transitions between the two subbands. To understand our field-dependent data, it is more useful to refer again to the effective Hamiltonian (3). If we add a Zeeman term involving only the local moments S_i, the singlet will be mixed with the triplet to produce four nondegenerate states at each site. $\chi''(q,\omega)$ will be governed by transitions among these states; at $T=0$, the dominant transition has energy

$$\Delta(H) = [\epsilon_0^2 + (g\mu_{\mathrm{eff}}H)^2]^{1/2}, \tag{6}$$

where for $K=0$, $\epsilon_0 = \Delta$. As can be seen in Figs. 2(c) and 2(d), Eqs. (6) and (5) (solid lines) with $g\mu_{\mathrm{eff}} = 2\mu_B$ and $\epsilon_0 = 0.26$ meV adequately fit the data. The specific heat as a function of the magnetic field[6] can also be described by use of Eq. (6). However, the specific-heat data, which yield only the ratio $\epsilon_0/g\mu_{\mathrm{eff}}$, indicate that $\epsilon_0 = 0.45$ meV when $g\mu_{\mathrm{eff}} = 2\mu_B$. In view of the q dependence of the magnetic fluctuations in $CeCu_6$, the disparity between the neutron results at a particular q and the thermodynamic (zone-averaged) data should come as no surprise.

In summary, we have shown that in the low-temperature, coherent, metallic regime of a paramagnetic heavy-fermion system, there are antiferromagnetic correlations of a kind not anticipated by simple Fermi-liquid theory. We have also demonstrated how magnetic fields quench the spin fluctuations in $CeCu_6$. Resonance lines, which exist for both local-moment systems and weakly interacting Fermi liquids, are absent for all momentum transfers and fields probed. Instead, the spectra remain quasielastic to fields as high as $H=6.4$ T, which corresponds to an energy larger than the spin-fluctuation energies (0.25–0.5 meV) measured at $H=0$.

An effective Hamiltonian H_{eff}, which accounts for the conduction electrons only in the sense that they result in an additional spin degree of freedom at each site of a lattice of local moments, yields an adequate parametrization of our results. Because H_{eff} contains two couplings, Δ and K, which compete to give characteristic energies as low as $\Delta(1-K/\Delta)^{1/2}$, it is also possible to understand anomalies, such as specific-heat maxima,[7] which occur well below the crossover to Pauli-type behavior in the bulk ($q=0$) susceptibility. We caution, however, that H_{eff} alone cannot be used to describe the physics of heavy-fermion systems. Indeed, K and Δ are merely parameters, yet to be determined from a theory which takes full account of the conduction electrons. Furthermore, the delocalized nature of the conduction electrons must be responsible for making the magnetic-fluctuation spectrum at each q a continuum rather than a well-defined resonance, as naive analysis of Eq. (3) would suggest. Finally, H_{eff} by itself does not explain the success of Fermi-liquid phenomenology for various bulk properties. Thus, a description of the heavy-fermion ground state must incorporate both Fermi-liquid aspects[17] and the physics accounted for by H_{eff}.

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