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IN THE HEAVY-FERMION COMPOUND PRINAG2

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Nonmagnetic Crystal-Electric-Field Ground State in the Heavy-Fermion Compound PrInAg_2

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Abstract: We have performed inelastic neutron scattering measurements that confirm that the crystal-electric-field split ground state in the heavy-fermion compound PrInAg_2 is a nonmagnetic, non-Kramers doublet. This implies that a quadrupolar Kondo interaction is responsible for the enhanced thermodynamic properties observed at low temperatures. We also observe anomalous broadening of the inelastic peaks and suggest two possible causes for this broadening.

Keywords: quadrupolar Kondo interaction, PrInAg_2 , inelastic neutron scattering, crystal electric field levels.

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The compound PrInAg_2 has been found to have a paramagnetic ground state with enhanced thermodynamic and transport properties at low temperatures [1]. Such renormalized behavior results when the conduction electrons interact with localized electrons having some internal degree of freedom. Usually this is a magnetic degree of freedom (as in the conventional Kondo interaction); however, previous experiments have indicated that the Pr^{3+} $4f$ ground state in the presence of a cubic crystal-electric-field (CEF) is a nonmagnetic, non-Kramers doublet. If this is correct, then the renormalized behavior at low temperatures could be due to a quadrupolar Kondo interaction [2], in which the internal degree of freedom is the quadrupolar moment of the $4f$ electron.

The correct CEF assignment is crucial in establishing the existence of this novel interaction. Furthermore, discrepancies exist between the predictions of the quadrupolar Kondo model and the measured thermodynamic and transport properties of PrInAg_2 . Presently, the calculations are based on a single impurity model, so the contradiction may arise from the periodicity of this compound. In addition to determining the CEF level scheme, we want to characterize the dynamic magnetic response of this new type of ground state which is unlike that observed in other correlated-electron compounds. In this paper, results from inelastic neutron scattering experiments will be presented which firmly establish a nonmagnetic ground state in PrInAg_2 . We also observe that the inelastic peaks are anomalously broadened and suggest two possible explanations for this.

Inelastic neutron scattering experiments were performed on the PHAROS chopper spectrometer at the Manuel Lujan, Jr. Neutron Scattering Center at Los Alamos National Laboratory. The sample consisted of 58 g of crushed, polycrystalline PrInAg_2 ; the preparation and characterization of this sample was described elsewhere [1].

The sample was mounted on a three-stage displacer refrigerator, and data were collected at two temperatures, 6.6 K and 77 K. At both temperatures, an incident neutron energy of 25.3 meV was used. The spectrometer resolution was measured with a vanadium plate where the elastic peak was very nearly gaussian with $\Delta E/E_i \sim 4\%$ (FWHM). Data from the planar array of linear, position-sensitive detectors were summed over the available Q-range (0.07 to 0.47 \AA^{-1} at the elastic line) and

histogrammed into constant energy bins. Since the transition matrix elements for some CEF level combinations are zero, a measurement was performed with the sample at 77 K where transitions between thermally populated states would be possible. After subtracting the data at 6.6 K, this result is shown in Fig. 1b, and two new peaks emerge at approximately 2 meV and 9 meV in the high temperature data.

Because the characteristic temperature of this system is so low ($T_K \sim 1$ K), higher-resolution data were collected at 4.4 K with the QENS spectrometer at the Intense Pulsed Neutron Source, Argonne National Laboratory. An inverse-geometry spectrometer, QENS uses a fixed final energy of $E_f = 3.65$ meV, and it has a resolution of ~ 0.150 meV at the elastic line. The same sample was used, this time mounted in a helium-4 cryostat. Data from each of the three detector banks on QENS were summed over the available range of Q and histogrammed into constant-energy bins. Only data from the high-angle bank ($Q = 2.2$ to 2.5 \AA^{-1} at the elastic line) are presented here in Fig. 2 because sample absorption strongly attenuated the signal in the lower angle banks. Unfortunately, spurious scattering from the cryostat is more problematic at high scattering angles, and the features from this effect are indicated with an arrow between ~ 0.15 and 0.35 meV in Fig. 2.

PrInAg_2 forms a fcc lattice with $a = 7.075 \text{ \AA}$. The CEF manifold of a $J = 4$ ion at a cubic symmetry site has four states: a Γ_1 singlet, Γ_4 triplet, Γ_5 triplet, and a Γ_3 nonmagnetic, non-Kramers doublet. Lea, Leask, and Wolf [3] determined the energies for these levels as a function of x , the ratio of the fourth to sixth order terms in the CEF Hamiltonian, and W , a parameter which sets the overall energy scale. When neutrons induce transitions between the various states in the manifold, the intensities of the resulting inelastic peaks are determined by matrix elements calculated by Birgeneau [4].

The CEF spectrum can be completely determined from the PHAROS data. Fitting the two inelastic peaks in the 6.6 K data with gaussians (with widths fixed to the instrument resolution) gives a ratio $I_{5,9}/I_{8,3} = 2.13 \pm 0.03$. From [4], the ratio of the intensity for the $\Gamma_3 \rightarrow \Gamma_4$ transition to that of the $\Gamma_3 \rightarrow \Gamma_5$ as 2.33, which is closer to the measured value than other possible

combinations. With Γ_4 or Γ_5 as the ground state, the ratio is 2.67 or 1.14, respectively. If Γ_1 were the ground state, then only one inelastic peak would be observed because transitions to this level are only allowed from Γ_4 . Therefore Γ_3 is assigned as the ground state, Γ_4 as the first excited level at 5.9 meV (68 K), and Γ_5 at 8.3 meV (95 K). At 77 K, the Γ_4 level is thermally populated, allowing us to observe additional transitions originating from this level. It follows then that the two new transitions observed at higher temperature correspond to $\Gamma_4 \rightarrow \Gamma_5$ (2 meV) and $\Gamma_4 \rightarrow \Gamma_1$ (9 meV), which puts the Γ_1 level at 16 meV above the ground state. This level scheme is shown in the inset to Fig. 1a. The energies of the Γ_4 and Γ_5 levels are very close to values observed by Galera *et al.*[5]. The present level arrangement is also similar to that estimated from the CEF contribution to the specific heat, except that from the specific heat method the energy of the Γ_1 level was estimated to be at 20 meV[1].

In the QENS data, the elastic peak is resolution limited, but the 5.9 meV peak is significantly broader than the instrument resolution. The inelastic peak at 8.3 meV may also be broadened. We suggest two possible explanations for this broadening. The crystal field symmetry may be lower than cubic, thus splitting the Γ_4 and Γ_5 levels. Alternatively, spin fluctuations involving a conventional, magnetic Kondo interaction between the excited magnetic CEF states and the conduction electrons may lead to a finite lifetime for this state. We are proceeding with further analysis to determine which scenario is correct.

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Figure Captions

Fig. 1. Scattering intensity versus neutron energy loss from PHAROS for PrInAg₂. (a) The sample temperature is 6.6 K. Inset shows the CEF level scheme determined from these measurements. (b) The sample temperature is 77 K. Note the left axis is expanded by a factor of 8.

Fig. 2. Scattering intensity versus neutron energy loss from QENS. This high-resolution data was taken on PrInAg₂ at $T = 4.4$ K. The arrow points to spurious scattering from the cryostat.

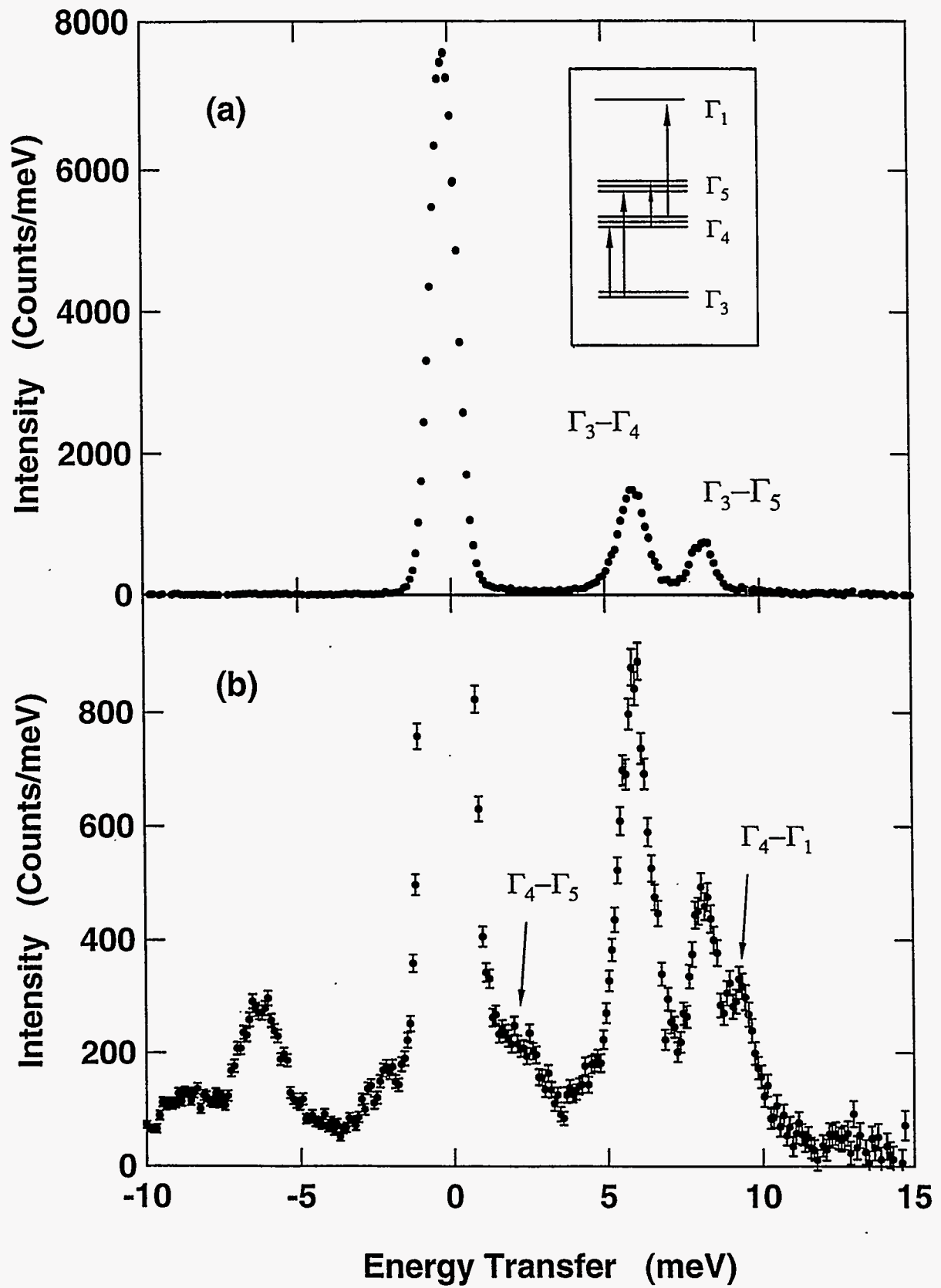


Fig. 1; Kelley *et al.*; SCES98

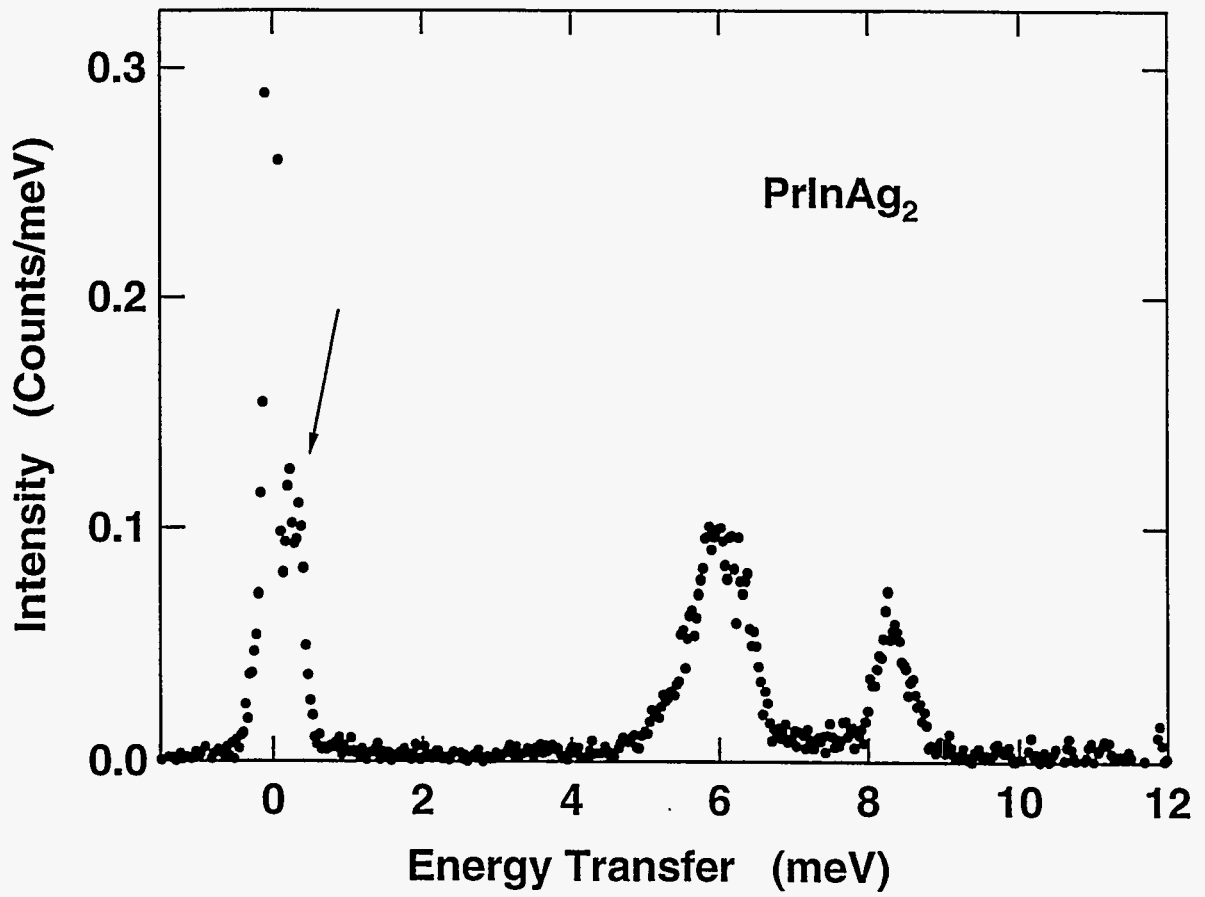


Fig. 2; Kelley *et al.*; SCES98