

Neutron scattering study of crystal fields in CeRhIn₅

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Neutron scattering results for the tetragonal compound CeRhIn₅ give evidence for two crystal field (CF) excitations at 6.9 and 23.6 meV. The scattering can be fit assuming a set of CF parameters $B_2^0 = -1.03$ meV, $B_4^0 = 0.044$ meV and $B_4^4 = 0.122$ meV. To compare our results to previous work, we calculate the susceptibility and specific heat for this CF scheme, including a molecular field term $\lambda = 35$ mol/emu to account for the Kondo effect. We also include a calculation based on these CF parameters that uses the non-crossing approximation to the Anderson model to estimate the effect of Kondo physics on the susceptibility, specific heat and neutron linewidths.

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CeRhIn₅ crystallizes in the same tetragonal HoCoGa₅ structure as the heavy fermion superconductors CeIrIn₅ and CeCoIn₅^{1,2,3}. At ambient pressure CeRhIn₅ undergoes a transition to an antiferromagnetic (AF) state at $T_N = 3.8$ K^{1,4}. With application of hydrostatic pressure the Néel temperature remains essentially constant until antiferromagnetism disappears and superconductivity appears at pressures above 15 kbar¹. Recently, Pagliuso *et al.*⁵ have suggested the importance of CF splitting to the ground state properties of the CeMIn₅ family of heavy fermion superconductors, underscoring the fact that the ultimate ground state achieved by a particular member of the family must grow out of the ground state crystal field doublet. Thus a careful determination of both the CF splitting and wavefunctions is important. To that end we have begun to directly probe the CF energy level splitting in the CeMIn₅ family using inelastic (IE) neutron scattering. The first step in our investigations has been determining the crystal field level scheme in CeRhIn₅.

In CeRhIn₅, as in the other members of CeMIn₅ family, the crystal field Hamiltonian in tetragonal symmetry can be written

$$H_{CF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4$$

where O_l^m and B_l^m are the Stevens operators and CF parameters respectively. The Ce³⁺ $J = 5/2$ wavefunction splits into three doublets, $\Gamma_7^{(1)} = \{\alpha|\pm 5/2\rangle + \beta|\mp 3/2\rangle\}$, $\Gamma_7^{(2)} = \{\beta|\pm 5/2\rangle - \alpha|\mp 3/2\rangle\}$ and $\Gamma_6 = |\pm 1/2\rangle$ ⁵. An analysis of susceptibility and thermal expansion results⁶ suggested crystal field levels $\Gamma_7^{(2)}$, $\Gamma_7^{(1)}$ and Γ_6 at $E=0$, 5.86 meV (68 K) and 28.43 meV (300 K) respectively, with $\beta = 0.969$ (yielding a nearly pure $|\pm 5/2\rangle$ ground state). A subsequent study⁵ based on an analysis of the susceptibility and specific heat suggested a similar scheme, but with splittings 6 and 12 meV (70 and 140 K). In this paper we report the results of an analysis of

neutron scattering data for CeRhIn₅ which indicate that these initial estimates are nearly correct; our results have somewhat different values for the splittings and a smaller value for the mixing parameter β , i.e., a greater admixture of $|\mp 3/2\rangle$ into the $|\pm 5/2\rangle$ ground state. To assist in comparison of our results to those of Pagliuso *et al.*⁵ and Takeuchi *et al.*⁶, we report calculations of the specific heat and magnetic susceptibility based on our CF parameters which include the Kondo effect in an ad hoc manner similar to those of refs. 5 and 6. We also present more sophisticated calculations that employ the non-crossing approximation (NCA)^{7,8} to the Anderson model in order to estimate the effect of Kondo spin fluctuations on the susceptibility, specific heat and IE neutron spectra.

Large high quality single crystals of CeRhIn₅ and LaRhIn₅ were obtained using the flux-growth method¹. For the magnetic susceptibility and specific heat measurements, single crystals were carefully prepared which were free of residual In flux; in the case of the neutron scattering measurements, ~ 50 g of single crystals for both CeRhIn₅ and LaRhIn₅ were powdered. The neutron scattering experiments were performed in time-of-flight mode using LRMECS at IPNS (Argonne National Laboratory) with experimental conditions that were similar to that of an earlier report⁹. A key problem in our investigations was the high neutron absorption of both In and Rh. In initial experiments the standard LRMECS sample holder was used; however, in subsequent experiments a new sample holder was employed which was designed to maintain a more uniform sample thickness than the standard holder, thus allowing for a more accurate absorption correction. Neutron scattering spectra were collected for several different incident energies (E_i) and temperatures between 8 and 140 K with counting times ranging from 24 to 48 hours. To improve statistics, we were able to take advantage of the nondispersive nature of the CF scattering and group detectors into three bins with mean scattering angle 20° (low Q), 60° and 100° (high Q). A

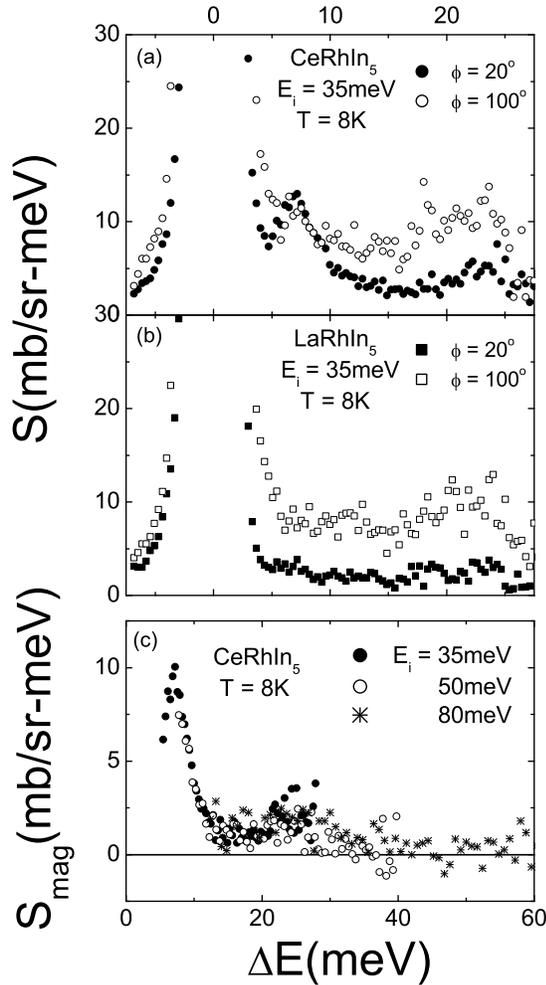


FIG. 1: Neutron energy spectra of (a) CeRhIn₅ and (b) LaRhIn₅ at an initial energy $E_i = 35$ meV, at 8 K and for two mean scattering angles, 20° and 100° . The data have been corrected for neutron absorption and the scattering from the sample holder has been subtracted from the data. (c) The $Q=0$ magnetic scattering, determined as described in the text, in CeRhIn₅ at 8K and for three incident energies E_i .

Vanadium standard was utilized to put the scattering on an absolute scale.

Data for CeRhIn₅ and LaRhIn₅ (measured to help identify the nonmagnetic scattering in CeRhIn₅) at 8 K and $E_i = 35$ meV for low and high Q are shown in Fig. 1. The data were corrected for absorption assuming a uniformly thick flat-plate sample and the scattering due to the empty sample holder was then subtracted from the data. Direct comparison of low angle scattering (where magnetic scattering is strongest) for CeRhIn₅ (Fig. 1a) and LaRhIn₅ (Fig. 1b) shows two additional peaks near 7 and 23 meV. In particular, we determine the nonmagnetic scattering in CeRhIn₅ in two ways: 1) By using

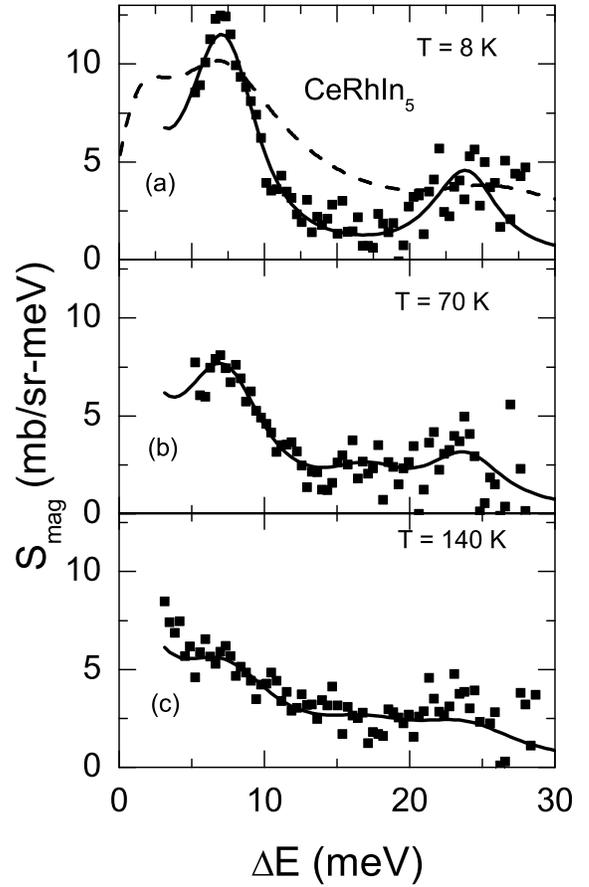


FIG. 2: Temperature dependence of the magnetic part of the IE neutron scattering response of CeRhIn₅ for $E_i = 35$ meV. The scattering dependence due to the Ce³⁺ form factor has been removed as in the previous figure. The data at all three temperatures ((a) 8 K, (b) 70 K, and (c) 140 K) have been fit simultaneously (solid lines) with a least squares fitting routine to determine the crystal field parameters. The results of the fitting parameters including the crystal field parameters are displayed in table I. We have included in (a) the results of the NCA calculation (dashed line).

the expression $S_{mag}(20^\circ) = S(Ce, 20^\circ) - fS(La, 20^\circ)$ where we choose the factor f as the ratio (0.75) of the total scattering cross-sections $\sigma(\text{CeRhIn}_5)/\sigma(\text{LaRhIn}_5)$. 2) By determining the ratio $R = S(La, 100^\circ)/S(La, 20^\circ)$ for scaling the high angle nonmagnetic scattering to low angle¹⁰. Excellent agreement with 1) is obtained using $S_{mag}(20^\circ) = S(Ce, 20^\circ) - FS(Ce, 100^\circ)/R$ with inclusion of an additional factor $F = 1.33$ to account for the difference in Q-scaling of the La and Ce compounds. The value of F is similar to the one used in a recent study of YbXCu₄⁸; it can be justified on the basis that for high angle scattering the data are predominantly single-phonon, proportional to σ , while the low angle scattering contains a significant contribution from multiple scattering (one elastic and one phonon) proportional to σ^2 , so that the

TABLE I: Crystal field parameters B_i^m , splittings and Lorentzian halfwidths Γ of the IE excitations at four temperatures for CeRhIn₅ and the wave function mixing parameter β . The units of all quantities (except for β , which is unitless) are meV. The reduced Chi-square for the fit was $\chi^2 = 0.69$

B_2^0	B_4^0	B_4^4
-1.03 ± 0.02	0.044 ± 0.001	0.122 ± 0.003
$E(\Gamma_7^1)$	$E(\Gamma_6)$	β
6.9 ± 0.3	23.6 ± 0.5	0.80 ± 0.02
$\Gamma(8K)$	$\Gamma(70K)$	$\Gamma(140K)$
2.3 ± 0.1	2.9 ± 0.2	4.2 ± 0.4

cross section does not cancel in the ratio. Results of this analysis for three different E_i are shown in Fig. 1c. The dependence of the scattering on the Ce³⁺ form factor has been removed in this plot, so the data represent the $Q = 0$ scattering with the assumption that the crystal fields are in fact purely local and uncoupled entities. The data have been truncated below $0.15E_i$ (where the elastic line dominates the scattering) and above $0.8E_i$, where statistics are small due to the k_f/k_i factor. Good agreement is evident for data taken at three different E_i , with all data sets displaying magnetic excitations at approximately 7 and 24 meV.

In Fig. 2 we plot the $Q = 0$ (form factor removed) magnetic scattering (method 1), determined at $E_i = 35$ meV, for three different temperatures. We have performed a simultaneous least squares fit to four datasets (8 K, 70 K and 140 K at $E_i = 35$ meV and 8 K at $E_i = 80$ meV) to determine the CF parameters. The fit includes the effects of instrumental resolution. Variables of the fit include B_2^0, B_4^0, B_4^4 and an overall scale factor (which four parameters were constrained to the same values for all datasets) and the Lorentzian halfwidth Γ of the IE excitations which was allowed to vary with temperature. (We constrained the quasi-elastic (QE) halfwidth to $1/2 \Gamma$.) Results of the fit are shown in Table I and plotted in Fig. 2.

To compare our results to those of Pagliuso *et al.*⁵ and Takeuchi *et al.*⁶, we have calculated the susceptibility and specific heat (Fig. 3). The susceptibility includes a positive molecular field contribution $\lambda = 35$ mol/emu where λ represents contributions to $1/\chi$ from AF and Kondo fluctuations. At high T these contribute to $1/\chi$ as $(T_K + T_N)/C_J$; with $C_J = 0.807$ emu-K/mol for $J = 5/2$ and $T_N = 3.8$ K this gives $T_K \sim 25$ K. We note that this value of $k_B T_K$ is similar to the width of the 7 meV IE excitation at 8 K. The calculation for the specific heat contains both a Schottky term due to the excited levels and a Kondo doublet term¹¹ with $T_K = 25$ K for the ground state level, which puts the calculated specific heat in the range 20-50 K in better agreement with measured value – without this, the calculated value due only to the Schottky contribution is smaller by a factor of 0.8. We

have not attempted to fit for the effects on C_{mag} and χ of the AF transition at 3.8 K.

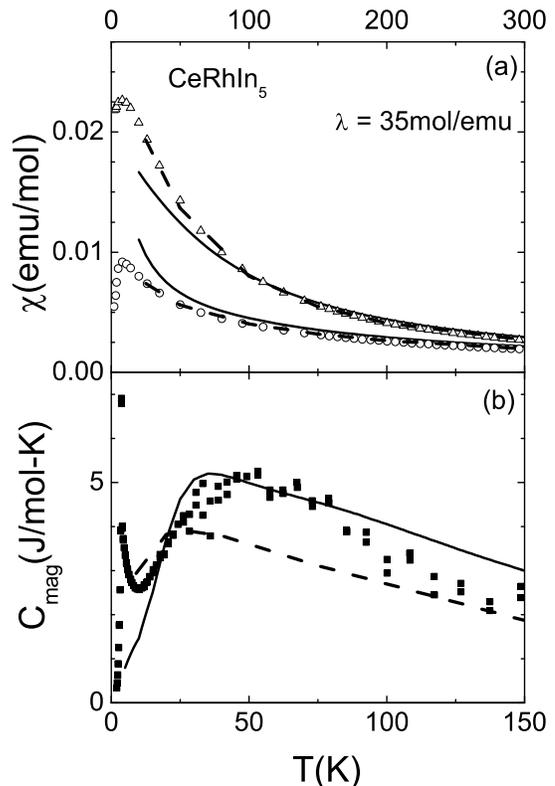


FIG. 3: (a) Measured anisotropic susceptibilities χ^{zz} (triangles) and χ^{xx} (circles) for CeRhIn₅ compared to the value calculated for the CF parameters of Table I with a molecular field contribution $\lambda = 35$ mol/emu (solid lines) and compared to the results of the NCA calculation (dashed lines). (b) Magnetic specific heat compared to the value calculated for a Schottky contribution from the excited levels and a Kondo contribution from the ground state doublet (solid line) and to the results of the NCA calculation (dashed line).

A more sophisticated way to include Kondo spin fluctuations is through calculation for the Anderson model. We present results obtained using the non-crossing approximation (NCA)^{7,8}. We have used a Gaussian background band with density of states $N(\varepsilon) = e^{-(\varepsilon/W)^2}/\sqrt{\pi}W$ with $W = 3$ eV and we set the $4f$ level position at $E_f = 2$ eV and the spin-orbit splitting at $E_{so} = 0.273$ eV, which are standard values for Ce. Since the Kondo physics renormalizes the CF levels upward by an amount approximately equal to the Kondo temperature the bare level energies were chosen to be $E_b = 5.3$ meV and $E_c = 23$ meV which are smaller than the measured level energies. The mixing parameter $\beta = 0.80$ was chosen to be similar to that obtained in Table 1. The hybridization was then varied until a good fit to the anisotropic susceptibility was obtained for $V = 0.4665$ eV. The results for S_{mag} , χ

and C_{mag} are given in Figs. 2a and 3.

We now turn to discussion of the effect of systematic errors on our conclusions. As mentioned previously, the neutron absorption of In and Rh is an important consideration. Comparison of the data for two different sample holders (which exhibited small differences in sample thickness and distribution) indicated similar results, augmenting our belief that the absorption correction employed is correct. If the nonmagnetic background subtraction is varied by varying f (method 1) or F/R (method 2), the scattering at 7 meV is relatively unaffected but the strength of the 24 meV scattering, and hence β , is affected somewhat. Given the good consistency between results at different E_i and T we think that our CF scheme is basically correct. We were unable to observe quasi-elastic (QE) scattering, due to the requirement that to obtain the resolution necessary a small E_i is required which causes the effects of neutron absorption, which varies as $1/\sqrt{E}$, to become large. In our fits we constrained the QE halfwidth to half the value of the IE width to prevent proliferation of fit parameters. Constraining to other values (e.g. $\Gamma_{QE} = \Gamma_{IE}$) leads only to minor variation in the final fits.

Our fits to χ using the CF parameters plus a molecular field term are not as good as those of Takeuchi *et al.*⁶ or Pagliuso *et al.*⁵. However, their fits use a value of β very close to unity, indicating essentially no $[\mp 3/2]$ admixture into $[\pm 5/2]$ ground state. In this case there would be no observable amplitude for the $\Delta m_z = 1$ transition to $[\pm 1/2]$ state at 24 meV. This cannot be correct as we clearly observe this transition in the neutron scattering data. A possible reason that our fits are not as good as those of refs. 5 and 6 is that we do not include the effect of exchange anisotropy, which should only be important below 20 K. Such anisotropy can be mimicked as in Pagliuso *et al.*⁵ through inclusion of an anisotropic mean field parameter, which we have chosen not to do

for simplicity.

On the other hand, the NCA calculations based on our CF scheme and a Kondo temperature of order 25 K does an excellent job reproducing χ . However, it overestimates the width of the 7 meV excitation as seen in Fig. 2a and underestimates the temperature of the peak in the specific heat (Fig. 3b). These deviations from the data may reflect the fact that we have neither included antiferromagnetic exchange, exchange anisotropy nor anisotropic hybridization (i.e. different hybridization to the different CF multiplets) in the NCA fits.

In summary, we find a more significant $[\mp 3/2]$ admixture into $[\pm 5/2]$ ($\beta = 0.80$) ground state than found earlier by Takeuchi *et al.* ($\beta = .969$)⁶ or Pagliuso *et al.* ($\beta \sim 1$)⁵. The resulting CF level parameters provide reasonable fits to both the magnetic susceptibility and specific heat with the inclusion of a mean field parameter and a Kondo doublet respectively. In addition, NCA fits the susceptibility remarkably well with some deficiencies in both the specific heat and neutron scattering linewidths. Taken together the NCA calculations and the fits to specific heat and susceptibility all indicate a $T_K \sim 25$ K. We note that the ordered moment $g\mu_B \langle J_z \rangle = 0.92\mu_B$ deduced for $\beta = 0.80$ is substantially larger than the value $0.36\mu_B$ needed to fit the diffraction pattern in the ordered state⁴. For our estimate of T_K the moment is reduced by the Kondo physics at temperatures $T > T_N$.

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