Magnetic Ground State of $CePb_3$

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The occurrence of magnetic order in the heavy-fermion system $CePb_3$ has been demonstrated by means of neutron scattering. The magnetic structure is antiferromagnetic with a moment modulation in the cubic (001) plane. The magnetic moments are aligned along the [001] axis, parallel to the antiferromagnetism direction. The local character of $4f$ electrons is evidenced by the observation of crystal-field transitions. The characteristics of $CePb_3$ are discussed in terms of competition between usual $4f$ properties and Kondo-type couplings.

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$CePb_3$ has been characterized as an anomalous Kondo lattice on the basis of the large linear term in the specific heat ($\gamma = 225$ mJ mole^{-1} K^{-2}) measured above 6 K.[1] At high temperature ($T > 2.5$ K) the cerium atoms behave as independent Kondo centers as evidenced by the logarithmic increase of the resistivity on cooling[2] and by the huge value of the thermoelectric power.[1] The observation of superconducting effects at high magnetic fields[2] has roused further interest for $CePb_3$. The specific heat is constant over a large range of temperature above the specific-heat anomaly,[1,2] but the possible occurrence of superconductivity at zero magnetic field was ruled out by resistivity measurements.[2] Magnetic ordering was conjectured but no experimental information was available either on the nature of the ordering or on the existence of crystal-field splitting. Renewed interest is now focused on the understanding of the existence in $4f$ or $5f$ compounds of a magnetic ground state in a relation to the f-level degeneracy, the crystal symmetry, and the electronic properties.[3–5] In particular, it has been realized that crude arguments based on a Kondo lattice with a doublet ground state[6] are not sufficient to account for nonmagnetic ground states quantitatively.[7] We have therefore used neutron-scattering techniques to investigate possible magnetic ordering as well as local properties of Ce ions.

$CePb_3$ crystallizes in the $AuCu_3$ structure with the O_h^1 space group. Several crystals were grown in a Bridgman furnace with typical dimensions of $13\times10\times3$ mm^3. The lattice constant at low temperature was $a = 4.849$ Å, in good agreement with a previous determination.[8] The low-temperature measurements were made with a dilution refrigerator,[9] allowing very quick reorientations of the sample. The temperature was determined from 70-Ω Matsushita and 100-Ω Allen-Bradley resistors.

Neutron-scattering experiments were carried out on triple-axis spectrometers at the high-flux reactor of the Laue-Langevin Institute in Grenoble. Pyrolitic graphite (002) crystals were used as monochromator and analyzer. Elastic measurements were performed at $k_i = 2.662$ Å^{-1} and $k_i = 1.5$ Å^{-1} with pyrolitic graphite and Be filters, respectively, in order to eliminate high-order contamination. Various collimations and constant-k_f values were used to perform inelastic-scattering experiments with adequate resolution.

During the search for superstructure reflections, the sample was mounted with different orientations. Q scans were performed along all high-symmetry directions of the Brillouin zone at temperatures above and below the expected ordering temperature deduced from measurements of bulk properties.[2] At low temperature they revealed new peaks around X points $(0,0,\frac{1}{2})$ but centered at incommensurate positions. Intensities were detected when scanning along the S line and the Z line on the Brillouin zone surface (inset of Fig. 1). Actually, because of the vertical divergence of the incident neutron beam, only tails of Bragg peaks were observed. The actual propagation vector has the form $\mathbf{q} = (\mu_1, \mu_2, \frac{1}{2})$. By tilting the crystal and using tight vertical collimation at $k_i = 1.5$ Å^{-1}, we could observe eight satellites around X points, corresponding to four pairs $(\mathbf{q}, -\mathbf{q})$ of wave vectors $\mathbf{q}$ (Fig. 1), and determine $\mu_1 = 0.135 \pm 0.001$ and $\mu_2 = 0.058 \pm 0.001$ at $T = 30$ mK. No high-order harmonic corresponding to a departure from a sine-wave modulation was ob-

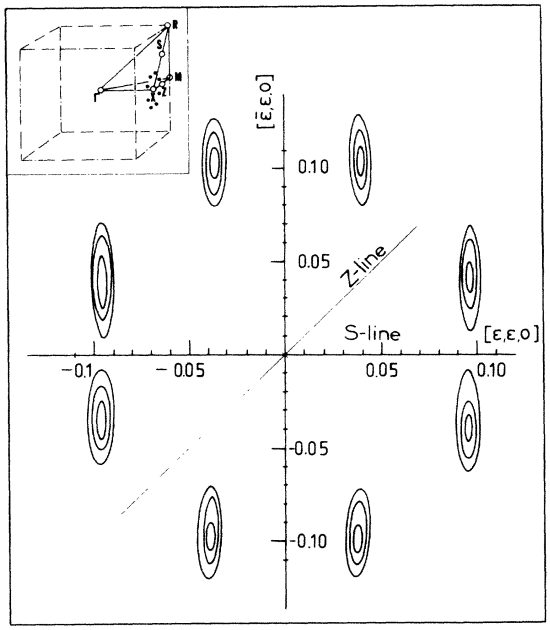

FIG. 1. Intensity-contour map around the X point (1,1,1.5) at $T=500$ mK. Contour lines correspond to 50, 100, and 200 counts per minute, respectively. The oblong shape of the observed satellites is due to the vertical resolution. Inset: symmetry lines on the Brillouin zone surface; the filled dots represent the satellites positions.

served. In the following we will assume a single-**q** structure although a coupling between different Fourier components of the order parameter cannot be ruled out. Since there are twelve pairs $(\mathbf{q}, -\mathbf{q})$, there will be twelve magnetic domains in this single-**q** model.

Satellites were observed in different Brillouin zones. However, no intensity was detected around the (0,0,1.5), (0,0,2.5), and (0,0,3.5) reciprocal-lattice points, which demonstrates that the Fourier component $\mathbf{m}_\mathbf{q}$ is parallel to a [001] direction. Integrated intensities were measured at $T=30$ mK in a two-axis configuration with an open detector. From these data we could estimate a Ce magnetic form factor in $CePb_3$ which is similar to that observed in other Ce compounds.[10] This confirms the magnetic character of the satellites. On the assumption of an equipartition of all the twelve magnetic domains, the normalization of magnetic intensities to weak nuclear reflections leads to a modulation amplitude $m_0=(0.55\pm0.10)\mu_B$ at $T=30$ mK.

The temperature dependence of the magnetic intensities is shown in Fig. 2. The Néel temperature is $T_N=1.16\pm0.05$ K, in good agreement with specific-heat data.[2] We approximated the temperature dependence of the order parameter below T_N by a power law:

$$m_q(T)/m_0=D(1-T/T_N)^\beta. \quad (1)$$

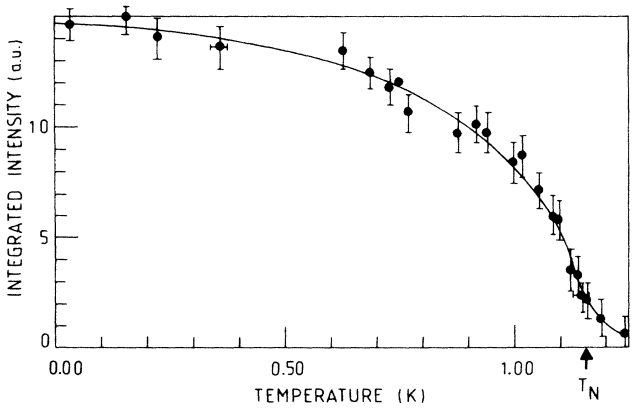

FIG. 2. Temperature dependence of the integrated magnetic intensity at the $(1+\mu_1, 1+\mu_2, 0.5)$ reciprocal-lattice position. The value of T_N was obtained by a fitting of the variation of the order parameter by Eq. (1).

Estimated values of the critical exponent β and amplitude D are $\beta=0.30\pm0.05$ and $D=1.4\pm0.2$. As shown in Fig. 3, the propagation wave vector varies with temperature and there is no indication for any lock-in to rational values for μ_1 and μ_2, although μ_1 remains constant over the whole range of temperature. It is not clear why only μ_2 shows some temperature dependence, but the apparent eightfold symmetry in Fig. 1 is purely accidental. The wave vector **q** rotates and changes its length. A temperature dependence of **q** was expected since the magnetic phase transition is found to be continuous.[11] At the phase transition, the order parameter, $\mathbf{m}_\mathbf{q}$, transforms according to one of two one-dimensional irreducible representations of the

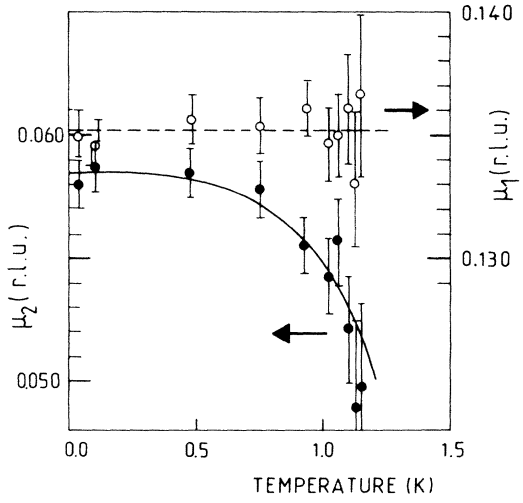

FIG. 3. Temperature dependence of the incommensurate components μ_1 (open circles) and μ_2 (filled circles) of the propagation vector in $CePb_3$. On the assumption of a constant value $\mu_1=0.135$, the accidental octogonal symmetry occurs for $\mu_2=0.056$.

group $G_{\mathbf{q}}$ (C_s) which leaves $\mathbf{q}$ invariant. Indeed, as mentioned above, $\mathbf{m}_{\mathbf{q}}$ has been found to be parallel to [001], which corresponds to the identical representation of C_s. Therefore in the single-$\mathbf{q}$ model, the magnetic moments are parallel to a [001] direction. The magnetic structure is approximately antiferromagnetic of type I in which ferromagnetic planes are stacked antiferromagnetically along [001]. For each [001] direction, the incommensurate components of the wave vector lead to an amplitude modulation of the moments in the (001) plane along four symmetry-related directions $[\mu_1\mu_2 0]$ which correspond to four domains. Since no high-order harmonics were observed, the magnetic-moment modulation can be represented by simple sine waves.

Crystalline-electrical-field (CEF) transitions between the Γ_7 and Γ_8 Ce multiplets have been observed at different points of the Brillouin zone below and above T_N. The measured mode is almost dispersionless and centered around 1.5 THz, although a weak softening can be detected near the satellite positions. The energy scans are much broader than the experimental resolution (Fig. 4). These observations are in agreement with previous time-of-flight experiments performed on polycrystalline samples.[12] Such anomalously wide inelastic transitions are to be contrasted with the absence of quasielastic scattering in our measurements at low temperature. The broadening appears to be constant in the Brillouin zone, and therefore cannot be related to a lifting of the Γ_8-multiplet degeneracy through the anisotropic exchange field, which should vanish at the Γ and R points.

From magnetic-susceptibility measurements,[13] it was deduced that the Ce ground state is the Γ_7 multiplet and that the CEF splitting is about 1.4 THz. This value, which is in agreement with our neutron study and with magnetoelasticity experiments,[14] fits very well in the variation of the CEF parameters across the $R\mathrm{Pb}_3$ series (R denotes rare earth).[13] Furthermore, antiferromagnetic order is a common feature among the $R\mathrm{In}_3$ and $R\mathrm{Pb}_3$ isomorphous compounds. The Néel temperature of the latter series is about 3 times smaller than in $R\mathrm{In}_3$, which corresponds to a weaker coupling of the $4f$ shell with the conduction band. However, there are different types of ordering which are characterized by commensurate wave vectors $(0,0,\frac{1}{2})$ and $(0,\frac{1}{2},\frac{1}{2})$ in the normal $R\mathrm{Pb}_3$ and $R\mathrm{In}_3$ series, respectively. The magnetic order in CePb_3, described by $(\mu_1,\mu_2,\frac{1}{2})$, is therefore reminiscent of what is observed in the normal $R\mathrm{Pb}_3$ compounds.[15] Nevertheless, the modulation amplitude m_0 is smaller than the expected value of the ordered moment for a pure Γ_7 CEF level ($0.71\mu_B$).

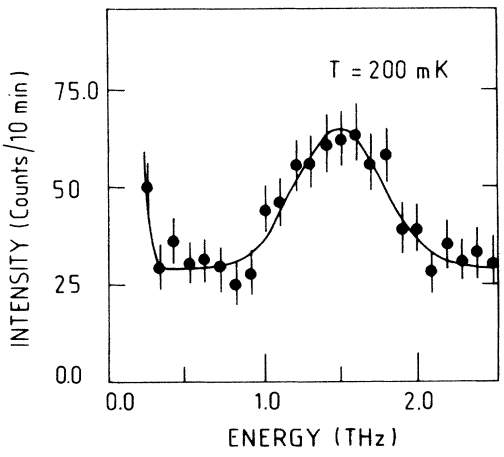

FIG. 4. Energy scan at $\mathbf{q}=(1,1,0.4)$ with constant $k_f=2.662$ Å^{-1}. The observed width (0.6 THz) is much larger than the experimental energy resolution (0.19 THz).

CePb_3 is a new illustration of modulated structure stable at zero temperature. As has been shown in another Kondo lattice CeAl_2,[16], the Kondo coupling leads to a singlet magnetic ground state and the anisotropy arising from the Γ_7-Γ_8 mixing provides the energy which is required to stabilize the amplitude-modulated structure rather than the spiral-like structure.[17] Another similarity with CeAl_2 lies in the temperature depedence of the resistivity which exhibits two maxima respectively around $T_1=2.5$ K and $T_2=25$ K for CePb_3,[2] and $T_1=4.6$ K and $T_2=65$ K for CeAl_2.[16] Scaling these values reproduces the differences in the Néel temperatures (1.16 and 3.8 K, respectively) and in the CEF splittings (1.5 and 2.9 THz[18]). The large broadening of the inelastic line is comparable to that observed in CeAl_2[19] and in CeIn_3.[20] Our experiment was not meant to measure narrow quasielastic scattering, but we can set an upper limit of $\Gamma \simeq 0.15$ THz for the width of such scattering at temperatures below 6 K.

Our neutron-scattering experiments have demonstrated that the ground state of CePb_3 at low temperatures is antiferromagnetic. The complicated modulation of the magnetic moments is related to electronic properties (Kondo effect above T_N) and the local character of the Ce^{3+} ions (CEF effects). The delicate link between electronic couplings and the microscopic nature of the magnetic structure can be studied continuously by the application of pressure. It would be interesting to verify if the amplitude-modulated magnetic structure of CePb_3 condenses into a simple antiferromagnetic structure when the CEF effects apparently collapse, under application of hydrostatic pressure, as observed in CeAl_2 at $P=20$ kbar,[21] or in CeIn_3 at normal pressure.[20,22] Furthermore, the proximity of a magnetic to nonmagentic transition can be related to the value of the critical exponent β.[23] The observed value $\beta \simeq 0.3$ in CePb_3, which corresponds to a Wilson-type exponent, suggests that CePb_3 is far

from such a transition. At high pressure an evolution of β toward a mean-field value would indicate the presence of spin fluctuations, as observed in $CeIn_3$.[20]

Our results confirm that the trivalent cubic Ce compounds ($CeAl_2$,[16] $CeIn_3$,[20,22] $CePb_3$) are magnetic. The absence of magnetic order in cubic Ce compounds[24] ($CeSn_3$, $CePd_3$, $CeBe_{13}$) corresponds to the occurrence of the intermediate-valence regime[3,4] as defined by the collapse of CEF effects, i.e., the recovery of all the $2J+1$ channels of the full angular momentum ($J=\frac{5}{2}$) of the Ce atoms. Our experiments confirm the important role played by the crystalline anisotropy in preventing magnetic ordering in heavy-fermion systems.[25] All the well-known nonmagnetic examples, $CeAl_3$, $CeCu_6$, $CeCu_2Si_2$, are noncubic materials. Physically, it seems that a strong anisotropy enhances electronic correlations and reduces magnetic couplings.

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