High magnetic field thermal-expansion and elastic properties of CeRhIn$_5$

V. F. Correa,$^1$ W. E. Okraku,$^2$ J. B. Betts,$^1$ A. Migliori,$^1$ J. L. Sarrao,$^1$ and A. H. Lacerda$^1$

$^1$Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
$^2$Occidental College, Los Angeles, California 90041, USA

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We report high magnetic field thermal-expansion and magnetostriction results on CeRhIn$_5$ single crystals. Several transitions, both first and second order, are observed when the field is applied perpendicular to the crystallographic c-axis. The magnetic field dependence of the thermal-expansion coefficient above 15 K, where the magnetic correlations are negligible, can be explained supposing an almost pure $|\pm 5/2\rangle$ ground state doublet, in apparent contradiction with neutron scattering experiments. Although the spin-lattice interaction is relevant in this compound, the effect of the magnetic correlations on the elastic properties is relatively weak, as revealed by resonant ultrasound spectroscopy experiments.

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I. INTRODUCTION

The main characteristic of the heavy fermion (HF) systems is the strong interaction between the conduction electrons and the localized f-electrons resulting in large electronic effective masses. This hybridization also gives rise to a non-magnetic Kondo singlet state or long-range magnetic order depending on the relative importance of the Kondo effect and the RKKY interaction.

CeRhIn$_5$ is a member of the CeMIn$_5$ (M = Co, Rh, Ir) family. It crystallizes in the tetragonal HoCoGa$_5$ structure with alternating layers of magnetic CeIn$_3$ and non-magnetic Mn$_2$ along the c-axis. CeRhIn$_5$ shows pressure induced superconductivity ($T_c = 2.1$ K at $P = 16$ kbar) and is the only member of the family that exhibits ambient pressure antiferromagnetism ($T_N = 3.8$ K). The Ce magnetic moments, antiferromagnetically ordered, lie completely in the basal plane developing a helicoidal structure along the c-axis with a propagation vector $q = (1/2,1/2,0.297)$ that is incommensurate with the atomic lattice. The absence of any magnetic order in CeIrIn$_5$ and CeCoIn$_5$ suggests that Rh plays a crucial role in establishing the magnetic structure. In fact, the magnetic moments along the c-axis (via a superexchange mechanism) while the usual RKKY interaction accounts for the antiferromagnetic order within the basal plane. The enhanced hybridization between the Ce-4f electrons and the conduction electrons would explain the heavy fermion superconducting state in CeIrIn$_5$ and CeCoIn$_5$.

Another evidence of localized moment behavior is the crystal electric field (CEF) effect on the Ce-4f electronic levels. Different experiments in CeRhIn$_5$ can be quite well understood in terms of CEF effects, although there are some differences in the proposed schemes: the main difference arising from the degree of admixture of the $|\pm 5/2\rangle$ and $|\pm 3/2\rangle$ levels in the ground state doublet. While thermal-expansion, specific heat and magnetic susceptibility data are explained supposing an almost pure $|\pm 5/2\rangle$ ground state doublet, inelastic neutron scattering (INS) experiments clearly show a peak at 23 meV that would be forbidden in such case. Moreover, in a previous work in La doped Ce$_{0.6}$La$_{0.4}$RhIn$_5$ we showed that the magnetic field dependence of the thermal-expansion coefficient could only be explained by a pure $|\pm 5/2\rangle$ ground state. In this case, the effect was attributed to changes in hybridization strength and chemical pressure due to the doping.

On the other hand, an important Kondo compensation is also observed in CeRhIn$_5$ through neutron diffraction experiments and corroborated by specific heat and resistivity measurements, showing that there is a subtle competition between the de-localized HF and the localized magnetic behaviors.

Despite the layered structure, the anisotropy displays a varying role. While resistivity and susceptibility show little anisotropy, the magnetic and CEF contributions to the linear thermal-expansion present greater anisotropy. In the same way, the helicoidal magnetic structure suggests an important two-dimensional character. This is evident in specific heat and magnetization measurements where several field-induced transitions are observed only when the magnetic field is applied along the basal plane.

Being that the spin-lattice coupling is a relevant issue in these Ce-based compounds, in this work we investigate the magnetic effects on the volume and the elastic properties of CeRhIn$_5$ single crystals using high field linear thermal-expansion, magnetostriction and elastic constant measurements. Both first and second order transitions are observed in the magnetostriction and thermal-expansion coefficients giving rise to a field vs. temperature phase diagram similar to that obtained from specific heat experiments. However, the magnetic correlations seem to have a minor effect on the elastic constants. The magnetic field dependence of the CEF contribution to the thermal-expansion cannot be explained by the levels scheme obtained from inelastic neutron scattering experiments.
II. RESULTS

CeRhIn$_5$ single crystals were grown by the self flux technique. The thermal-expansion experiments were performed using a capacitance dilatometer. The elastic properties were studied using resonant ultrasound spectroscopy.

Figure 1 shows the zero field linear thermal-expansion coefficient $\alpha = 1/L(dL/dT)$ along the c-axis of the crystal and perpendicular to it (i.e., along the ab-basal plane), $\alpha_c$ and $\alpha_{ab}$ respectively. As stated in previous works, two features can be distinguished: the antiferromagnetic transition is detected as a huge and anisotropic peak at low temperature, and the negative contribution to $\alpha_c$ around 25 K associated with crystal electric field (CEF) effects is observed.

The main panel of Fig. 2 displays the low temperature $\alpha_c$ showing the magnetic correlations associated peaks, for zero field and $B = 18$ T applied along the ab-plane (main panel) and along the c-axis (inset).

Figure 3 shows the field dependence of $\alpha_c$ at $T_N$.

It is difficult to say something in CeRhIn$_5$ about the order of the phase transitions from thermal expansion measurements. A hint may be obtained from magnetostriction experiments. Typical results for fields in the ab-plane can be observed in the inset of Fig. 2 where the field dependence of the c-axis is shown. First and
second order phase transitions can be clearly distinguished in different temperature ranges. Putting all this information together results in the magnetic field versus temperature phase diagram shown in the main panel of Fig. 4. There is a remarkable good agreement between thermal-expansion (open symbols) and magnetostriction (solid symbols) data. This phase diagram also has strong similarities with that obtained from specific heat experiments. According to Cornelius et al.\(^{14}\) phase I and II correspond to magnetic structures incommensurate with the atomic lattice, while phase III corresponds to a commensurate magnetic structure. Transitions between phases I and II, and between phases II and III, are first order.

CEF effects are known to have relevant contributions to the specific heat, susceptibility and thermal-expansion of the 115 and the related 218 family.\(^9\),\(^10\),\(^11\),\(^12\),\(^13\),\(^14\),\(^15\),\(^16\) Figure 4 shows \(\alpha_c\) at zero field and 18 T along the \(c\)-axis in an extended temperature range. As previously observed\(^{12}\) in Ce\(_{0.5}\)La\(_{0.5}\)RhIn\(_5\), the CEF-related negative peak around 25 K moves towards higher \(T\) when the field is increased. This is an anomalous behavior if we consider the CEF scheme obtained from inelastic neutron scattering measurements.\(^{11}\) According to these data, the CEF induced splitting of the Ce\(^{3+}\) \(J = 5/2\) multiplet results in a \(J = 5/2\) \(\pm 5/2\) ground state doublet with a mixing parameter \(\eta = 0.6\). However, as shown in Ref. \(^{12}\), the field induced shift of the \(\alpha_c\) minimum can only be explained supposing an almost pure \(\pm 5/2\) ground state doublet (\(\eta \sim 0\)). In that case, the result was attributed to chemical pressure and hybridization changes due to La doping. But, what happens in the present case of non-doped CeRhIn\(_5\)? A peak in the INS spectra related to transitions from the ground state doublet to the second excited doublet (forbidden if \(\eta = 0\)), is unambiguously detected.\(^{11}\)

The other contribution that we are perhaps neglecting is the influence of the Kondo interaction on the CEF effects and vice versa. The Kondo scattering is reduced by the CEF splitting giving rise to an effective Kondo temperature \(T_K^{\text{eff}}\) for the ground doublet state that can be much lower than the Kondo temperature \(T_K\) for the whole \(J = 5/2\) multiplet.\(^{10,11}\) On the other hand, as a result of the hybridization between the conduction and the \(f\)-electrons, the CEF levels become dispersive bands whose width is \(T_K^{\text{eff}}\) (Ref. \(^{11}\)).

According to the CEF scheme from INS results, a magnetic field splits the three doublets and mixes together the ground state and the first excited doublet. For \(B = 18\) T, the separation between the two lowest singlet states is \(\Delta S^1(B = 18\) T\) \(\sim 21\) K. Hence, for the whole field range we can access (up to 18 T), \(\Delta S^1(B) < T_K^{\text{eff}} \sim 25\) K (Ref. \(^{11}\)), and both states overlap. Moreover, the expected value of the magnetic moment \(g\mu_B\langle J_z \rangle\) along \(B = 0.92\) \(\mu_B\) for zero field) for both the ground and first excited singlet states are amazingly different: 1.52 and 0.02 \(\mu_B\), respectively. As the Kondo effect is an interaction between the free electron and the localized magnetic moments, different hybridization can be expected for different magnitudes of the magnetic moment.

In fact, a good fit to the specific heat data needs not only a CEF contribution but also a Kondo contribution, too.\(^{10,11}\) So, it is reasonable to think that the same would be necessary for the thermal-expansion data. In this way, it must be stressed that fits to \(\alpha\) disregarding the Kondo effect\(^2\) lead to similar results as ours: an almost pure

\[\Delta S^1(B) \sim T_K^{\text{eff}}\]
Finally, we study the effect of the magnetic correlations on the elastic properties. Figure 6 shows the temperature dependence of the adiabatic bulk modulus $B_S$ at zero field. Only a very subtle effect can be observed across $T_N$. The same results are obtained for any elastic constant across any phase boundary displayed in Fig. 4. However, a small increase in $B_S$ is observed below 10 K, that is roughly the temperature up to which the antiferromagnetic fluctuations are detected. It leads us to the conclusion that although the magnetic correlations have an important effect on the lattice dimensions (a relevant spin-lattice coupling), as shown by thermal-expansion and magnetostriction measurements, they have a less important effect on the harmonic part of the interatomic potential energy, as evidenced by the elastic constants.

III. CONCLUSIONS

Thermal-expansion, magnetostriction and elastic constant measurements were performed in CeRhIn$_5$ single crystals. A rich magnetic field vs. temperature phase diagram is observed when the field is applied along the ab-plane, in good agreement with specific heat experiments. Both first and second order transitions can be clearly distinguished. The magnetic field dependence of the CEF contribution to the thermal-expansion cannot be explained by the levels scheme obtained from inelastic neutron scattering experiments, suggesting that perhaps the effect of the Kondo interaction on the thermal-expansion should be considered. The magnetic correlations show no major effect on the elastic properties, the most relevant effect being a small increase in the adiabatic bulk modulus below 10 K, presumably associated with the antiferromagnetic fluctuations.

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1. A. C. Hewson, in The Kondo Problem to Heavy Fermions, edited by D. Edwards and D. Melville (Cambridge University Press, United Kingdom, 1992).
2. H. Hegger, C. Petrovic, E. G. Moshopoulou, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. Lett. 84, 4986 (2000).
3. C. Petrovic, R. Movshovich, M. Jaime, P. G. Pagliuso, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Europhys. Lett. 53, 354 (2001).
4. C. Petrovic, P. G. Pagliuso, M. F. Hundley, R. Movshovich, J. L. Sarrao, J. D. Thompson, Z. Fisk, and P. Monthoux, J. Phys. Condens. Matter 13, L337 (2001).
5. R. A. Fisher, F. Bouquet, N. E. Phillips, M. F. Hundley, P. G. Pagliuso, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. B 65, 224509 (2002).
6. N. J. Curro, P. C. Hammel, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, and Z. Fisk, Phys. Rev. B 62, R6100 (2000).
7. W. Bao, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, Z. Fisk, J. W. Lynn, and R. W. Erwin, Phys. Rev. B 62, R14621 (2000); Phys. Rev. B 63, 219901(E) (2001); Phys. Rev. B 67, 099903(E) (2003).
8. A. Schenck, D. Andreica, F. N. Gygax, D. Aoki, and Y. Onuki, Phys. Rev. B 66, 144404 (2002).
9. T. Takeuchi, T. Inoue, K. Sugiyama, D. Aoki, Y. Tokiwa, Y. Haga, K. Kindo, and Y. Onuki, J. Phys. Soc. Jpn. 70, 877 (2001).
10. P. G. Pagliuso, N. J. Curro, N. O. Moreno, M. F. Hundley, J. D. Thompson, J. L. Sarrao, and Z. Fisk, Phys. B 320, 370 (2002).
11. A. D. Christianson, J. M. Lawrence, P. G. Pagliuso, N. O. Moreno, J. L. Sarrao, J. D. Thompson, P. S. Riseborough, S. Kern, E. A. Goremychkin, and A. H. Lacerda, Phys. Rev. B 66, 193102 (2002).
12. V. F. Correa, L. Tung, S. M. Hollen, P. G. Pagliuso, N. O. Moreno, J. C. Lashley, J. L. Sarrao, and A. H. Lacerda, Phys. Rev. B 69, 174424 (2004).
13. A. D. Christianson, A. H. Lacerda, M. F. Hundley, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. B 66, 054410 (2002).
14. A. L. Cornelius, P. G. Pagliuso, M. F. Hundley, and J. L. Sarrao, Phys. Rev. B 64, 144411 (2001).
15. A. L. Cornelius, A. J. Arko, J. L. Sarrao, M. F. Hundley, and Z. Fisk, Phys. Rev. B 62, 14181 (2000).
16. A. Malinowski, M. F. Hundley, N. O. Moreno, P. G. Pagliuso, J. L. Sarrao, and J. D. Thompson, Phys. Rev. B 68, 184419 (2003).
For a good description of the technique see A. Migliori and J. L. Sarrao, in *Resonant Ultrasound Spectroscopy* (John Wiley and Sons, Inc., USA, 1997).

A. D. Christianson, E. D. Bauer, J. M. Lawrence, P. S. Riseborough, N. O. Moreno, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, E. A. Goremychkin, F. R. Trouw, M. P. Hehlen, and R. J. McQueeney, Phys. Rev. B **70**, 134505 (2004).

H. Suzuki, H. Kitazawa, T. Naka, J. Tang, and G. Kido, Solid State Commun. **107**, 447 (1998).

W. Bao, G. Aeppli, J. W. Lynn, P. G. Pagliuso, J. L. Sarrao, M. F. Hundley, J. D. Thompson, and Z. Fisk, Phys. Rev. B **65**, 100505(R) (2002).