nomena, edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. 5B, 343.
[6]U. Schindewolf and M. Werner, Ber. Bunsenges. Phys. Chem. 81, 1008 (1977).
[7]P. Damay and P. Chieux, J. Chem. Phys. 84, 1203 (1980).
[8]M. Gitterman and V. Steinberg, Phys. Rev. A 20, 1236 (1979).
[9]M. H. Cohen and J. Jortner, Phys. Rev. A 10, 978 (1974).

Effect of High Magnetic Fields on the Electronic Specific Heat in the Strongly Pauli-Paramagnetic Compound $LuCo_2$

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The low-temperature (1.3 to 20.0 K) heat capacity of the strongly Pauli-paramagnetic $LuCo_2$ compound was measured in magnetic fields up to ~10 T. The measured results show that the electronic-specific-heat constant decreases with increasing magnetic field (11% at 10 T), while the Debye temperature does not change. This is probably due to the depression of spin fluctuation enhancement of the heat capacity by moments induced on the cobalt atoms by the high magnetic fields. These results appear to be in accord with recent theoretical predictions.

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The anomalous enhancement of the electronic contribution to the heat capacity due to spin fluctuations in strongly Pauli-paramagnetic metals has been of considerable interest theoretically for about a decade.[1] Apparently only Brinkman and Engelsberg[2] have treated the problem of the effect of magnetic field on the enhanced electronic specific heat, and they suggested enormous magnetic fields are required to reduce the spin fluctuation enhancement to the specific heat. As far as we are aware no experimental evidence has been reported showing that an applied field reduced the enhancement due to spin fluctuations. Recently, however, we have found that the application of a magnetic field of ~10 T lowers the electronic-specific-heat constant of $LuCo_2$ by about 10%. The results of low-temperature (1.3 to 20 K) heat-capacity measurements as a function of magnetic field and the impact of these results on the theoretical models is described below.

For a nearly ferromagnetic metal the low-temperature heat capacity is given by

$$C=\gamma T+\beta T^3+cT^3\ln(T/T_s), \quad (1)$$

where γT and βT^3 are, respectively, the usual electronic and lattice contributions to the heat capacity, and the third term on the right is the increase in the heat capacity due to spin fluctuations (paramagnon interactions).[1-4] The constant c includes the term $1/T_s^2$ where T_s is the characteristic spin fluctuation temperature. As shown by Lederer and Mills[5] and by Engelsberg, Brinkman, and Doniach,[6] Eq. (1) is valid only for the case of a uniform enhancement throughout the lattice, but if one takes into account local enhancement on one atom (the magnetically dilute solute atom) then the $T^3\ln T$ term is negligible except for $T<T_s/100$ and the next leading term is small and has a T^3 dependence. Subsequently many modifications and improvements have been made on the original models,[3-6] and these have been recently reviewed by Moriya.[1] However, most authors did not treat this problem as a function of magnetic field.

It was first pointed out by Brinkman and Engelsberg[2] and more recently by Hertel, Appel, and Fay[7] that the application of high magnetic fields offer at least one way of testing the spin fluctuation theory. Brinkman and Engelsberg explained that high magnetic fields of the order of characteristic spin fluctuation temperature, T_s, are required to quench the spin fluctuation enhancements. Hertel, Appel, and Fay,[7] who have made a more detailed mathematical analysis, suggest that if the Stoner enhancement and the mass enhancement due to spin fluctuations are large (~4 and ~1.5, respectively) and if the spin fluctuation temperature T_s is small (~15 K) a depression of the heat capacity of about 1% might be expected at 10 T. They have suggested an f-band metal such as UAl_2 might have the three parameters with the

appropriate magnitude to see such an effect.

$LuCo_2$ was chosen because it reportedly[8] has a large Stoner enhancement factor ~10 and because the isomorphic closely related compound YCo_2 has an experimental density of states much larger than the theoretical value.[9] However T_s is quite large, 630 K.[8] Heat capacity measurements of this strongly Pauli-paramagnetic compound[10] were made from 1.3 to 20 K in magnetic fields from 0 to 10 T.

A $LuCo_2$ sample containing 35.0 at.% Lu was prepared by arc melting. The starting materials were 99.9 at.% pure Lu, which was prepared at this laboratory, and 99.99 at.% pure Co, which was purchased from Johnson Matthey, Ltd. The major magnetic impurities in the starting lutetium and cobalt metals were 34 and 3 ppm Fe, and 10 and 15 at. ppm Ni, respectively, and thus the resultant $LuCo_2$ compound contained 14 at. ppm Fe and 13 at. ppm Ni. The ingot was furnace-cooled after annealing in a sealed tantalum capsule at 1000 °C for a week. X-ray powder photographs and metallographic analyses have shown that the $LuCo_2$ sample is single phase. Moreover, the magnetization measurements have shown that the sample has no magnetic contamination.

The low-temperature calorimeter used in this work is of the usual isolation heat-pulse type with a mechanical heat switch. The temperature was measured by using a germanium resistance thermometer which had been calibrated at magnetic fields of 0, 2.50, 5.39, 7.62, and 9.98 T.[11] The heat capacity of the 1965 Calorimetry Conference standard[12] copper sample was measured to serve as a check of the apparatus and experimental technique. Accordingly, the low-temperature heat capacity results obtained on our copper standard sample were in agreement within ±1% at 0 T and ±2% at the four nonzero magnetic fields with the previous zero-field data.[12]

Figure 1 shows the results of the heat capacity measurements at low temperatures for $LuCo_2$ at various magnetic fields. As can be seen, all C/T vs T^2 curves are linear and there is no evidence for a $T^3 \ln T$ term in our data, except possibly for the slight upturn noted in 5.39-, 7.62-, and 9.98-T-field data. The absence of an upturn at $H=0$ and 2.50 T would tend to rule out the existence of a $T^3 \ln T$ contribution to the heat capacity, unless T_s was changing with increasing field. This is not too likely. A more reasonable explanation is that this increase is due to the superparamagnetic behavior of magnetic impurities such as iron in the $LuCo_2$. Moreover, one can distinctly

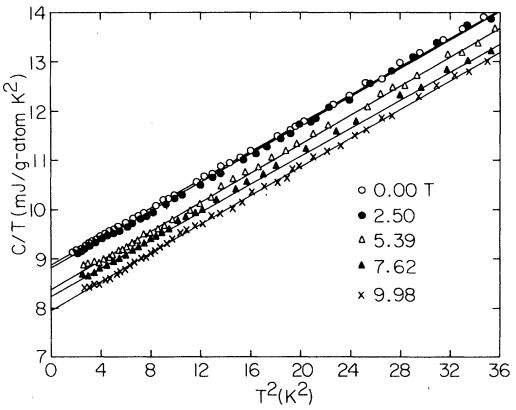

FIG. 1. The heat capacity data for $LuCo_2$ in the magnetic fields of 0, 2.50, 5.39, 7.62, and 9.98 T. The solid lines are the results of a least-squares fitting of the data for each of the five magnetic fields to the equation $C/T = \gamma + \beta T^2$.

see that the curves are parallel to one another and that they fall with increasing magnetic fields, especially for $H > 2.5$ T. The electronic-specific-heat constant γ and the "Debye temperature Θ_D" were calculated from a least-squares fit of the data for each field between 1.3 and 6.0 K to the equation $C/T = \gamma + \beta T^2$. The quotation marks are used here because if the paramagon contribution to the heat capacity has a T^3 dependence, then the resultant value extracted from the coefficient of the T^3 term, β, is not the true Debye temperature. The analytical results for the γ and "Θ_D" values of $LuCo_2$ in each field are shown in Fig. 2, where $\gamma = 8.9$ mJ/g-atom·K² and "Θ_D" = 238 K at zero field. As shown in Fig. 2, γ decreases slowly for $H < 2.5$ T and then more rapidly for $H > 2.5$ T with increasing magnetic fields and reaches ~89% of the zero-field result at 9.98 T, whereas "Θ_D" is independent of the magnetic field within the accuracy of measurements (the maximum difference in "Θ_D" is 0.9%). The field dependence of γ follows quite closely to that predicted by Hertel, Appel, and Fay.[7]

A polarized neutron scattering study of $LuCo_2$ in a field of 5.72 T at 100 K indicates that the magnetic field induces a moment on the Co atoms.[13] Thus, it appears that the induced moments on the cobalt atoms depress the spin fluctuation enhancement to the heat capacity. High-field magnetization studies up to 38 T show that the susceptibility of $LuCo_2$ at 4.2 K is constant up to ~22 T,[14] i.e., no change in the magnetic susceptibility is observed over the range of magnetic

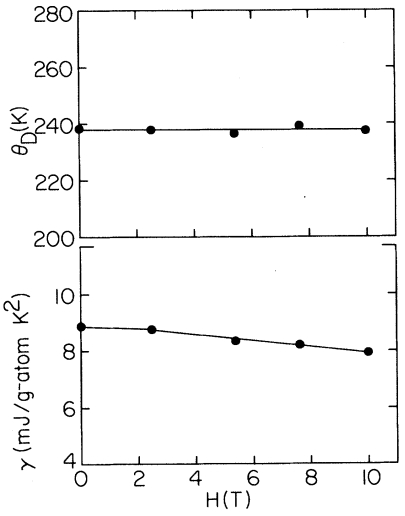

FIG. 2. The electronic-specific-heat parameter, γ, and the "Debye temperature, Θ_D", for $LuCo_2$ as a function of the magnetic fields to 9.98 T.

fields of our heat-capacity measurements.

The band structure calculations of Cyrot and Lavagna[9] for the RM_2 compounds, where R is a rare-earth element and M is Fe, Co, or Ni, show (1) that the $N(E)$ vs E curves for these compounds are nearly the same as the respective pure metal M and are independent of the rare-earth element R, and (2) that there is good agreement between the calculated and experimental γ values for YFe_2 and YNi_2 but not for YCo_2. Assuming that their density of states at the Fermi level value for YCo_2 is valid for $LuCo_2$ we have estimated the mass enhancement factor due to spin fluctuation as follows. The total enhancement factor, λ_{total}, is given by

$$1+\lambda_{\text{total}}=N(E_F)/N_0(E_F), \tag{2}$$

where $N(E_F)$ is the experimental and $N_0(E_F)$ is the calculated density of states at the Fermi level. By using Cyrot's and Lavagna's $N_0(E_F)$ value and the $N(E_F)$ value calculated from the zero-field γ value, one finds that $1+\lambda_{\text{total}}=5.1$. The total enhancement factor is given by

$$\lambda_{\text{total}}=\lambda_{ep}+\lambda_{\text{spin}}=4.1, \tag{3}$$

where λ_{ep} is the electron-phonon enhancement factor and λ_{spin} is the contribution due to spin fluctuations. The value of λ_{ep} for pure[15] Co is 0.2 and those reported for pure[16,17] Lu vary from 0.2 to 0.4; thus, an estimated value of 0.3 for $LuCo_2$ is not unreasonable, especially since $LuCo_2$ is not superconducting and is probably correct within a factor of 2. Substituting this value into Eq. (3) one obtains a value of $\lambda_{\text{spin}}=3.8$ which is quite large, but consistent with our observations (11% decrease in γ at 10 T) and the theory of Hertel, Appel, and Fay.[7] This large value of λ_{spin} calls for a more accurate band-structure calculation for $LuCo_2$ to verify the low value of $N_0(E_F)$ we have assumed for $LuCo_2$ based on the $N_0(E_F)$ value for YCo_2 calculated by Cyrot and Lavagna.[9] Even if the $N_0(E_F)$ value were wrong by a factor of 2, λ_{spin} would still be quite large, ~1.3, and consistent with the theory of Hertel, Appel, and Fay.[7] The large value of λ_{spin} estimated for $LuCo_2$, 3.8, makes it a prime candidate for a p-wave superconductor.[18]

The reported T_s value for $LuCo_2$ (630 K), however, is unreasonably large, by about two orders of magnitude. Based on our observations and the theory of Hertel, Appel, and Fay it is suggested that a careful low temperature, <20 K, magnetic susceptibility study be made to see if a peak in χ vs T is observed which corresponds to a T_s of ~10 K. The broad peak observed in the susceptibility at ~350 K for $LuCo_2$ (Ref. 10) is probably a band-structure effect and is not due to spin fluctuations.

In addition, our measurements up to 20.0 K show that the difference of the C/T vs T^2 curves at 0 and 9.98 T is a maximum around 12 K. This may mean that the enhanced electronic-specific-heat constant is predominant at low temperatures and the enhancement is suppressed because of the renormalization of spin fluctuation as the temperature increases.[19]

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[1]T. Moriya, J. Magn. Magn. Mater. 14, 1 (1979).

[2]W. F. Brinkman and S. Engelsberg, Phys. Rev. 169, 417 (1968).

[3]N. F. Berk and J. R. Schrieffer, Phys. Rev. Lett.

17, 433 (1966).
[4]S. Doniach and S. Engelsberg, Phys. Rev. Lett. 17, 750 (1966).
[5]P. Lederer and D. L. Mills, Phys. Rev. Lett. 20, 1036 (1968).
[6]S. Engelsberg, W. F. Brinkman, and S. Doniach, Phys. Rev. Lett. 20, 1040 (1968).
[7]P. Hertel, J. Appel, and D. Fay, Phys. Rev. B 22, 534 (1980).
[8]G. Barnea, J. Phys. F 7, 315 (1977).
[9]M. Cyrot and M. Lavagna, J. Phys. 40, 763 (1979).
[10]D. Block, F. Chaisse, F. Givord, J. Voiron, and E. Burzo, J. Phys. (Paris), Colloq. 32, C1-659 (1971).
[11]K. Ikeda, K. A. Gschneidner, Jr., B. J. Beaudry, and U. Atzmony, to be published.
[12]D. W. Osborne, H. E. Flotow, and F. Shreiner, Rev. Sci. Instrum. 38, 159 (1967); D. L. Martin, Can. J. Phys. 47, 1253 (1969).
[13]D. Gignoux, F. Givord, W. C. Koehler, and R. M. Moon, J. Magn. Magn. Mater. 5, 172 (1977).
[14]C. J. Schinkel, J. Phys. F 8, L87 (1978).
[15]D. A. Papaconstantopoulos, L. L. Boyer, B. M. Klein, A. R. Williams, V. L. Morruzzi, and J. F. Janak, Phys. Rev. B 15, 4221 (1977).
[16]K. H. Bennemann and J. W. Garland, in *Superconductivity in d- and f-Band Metals—1971*, edited by D. H. Douglas, AIP Conference Proceedings No. 4 (American Institute of Physics, New York, 1972), p. 103.
[17]J. J. Hopfield, Physica 55, 41 (1971).
[18]D. Fay and J. Appel, Phys. Rev. B 16, 2325 (1977).
[19]H. Hasegawa, J. Phys. Soc. Jpn. 38, 107 (1975).

Effect of Collective Oscillations on Low-Energy Thermal-Neutron Scattering and Transport in Liquid Hydrogen

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A thermal-neutron inelastic scattering kernel based on recently observed collective motion in liquid hydrogen, taken to be of quasicrystalline nature with an appropriate chemical binding energy, is found to explain the experimental pulsed neutron and steady-state spectra studies at 20 K. Thus the collective motion plays a very significant role in the transport of low-energy thermal neutrons in liquid hydrogen.

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In a recent study Carneiro, Nielsen, and McTague[1] have reported the presence of collective motion in liquid parahydrogen at 14.7 K observed by them using coherent neutron inelastic scattering. A distinguished feature of their measurement *vis à vis* other measurements[2,3] on monatomic coherent liquids is the presence of two peaks in the scattering law corresponding to the transverse and longitudinal modes similar to what one expects in a polycrystal. The significance of the presence of collective motions in liquid parahydrogen particularly for low-energy thermal-neutron inelastic scattering, and hence transport studies, is shown by the experiment itself. If the collective motions were absent, or were of minor importance, the scattering law would not have contained the low-energy dynamical structure. In another similar experiment on solid hydrogen performed at 5.4 K, Nielsen and Carneiro[4] have found that the phonon dispersion curves are similar to those of continuum model of a solid. Even the measured values of mean square displacements in C direction and the basal plane are almost equal, i.e., the mean square displacement of a hydrogen molecule is isotropic even in hydrogen crystal. Thus, it is evident that the collective motion is not only present in liquid hydrogen, but that it can be approximated rather well by the collective motion of a continuous medium. This conclusion is consistent with the inference of Carneiro, Nielsen, and McTague, based on their experimental results on liquid hydrogen, that in several respects the dynamics of liquid hydrogen is solidlike. We, therefore, suggest that the effect of collective motion on low-energy thermal-neutron scattering and hence transport from liquid hydrogen may be like that from a continuous or a quasicrystalline medium, with an appropriate chemical binding energy represented by a characteristic Debye temperature, Θ_D. Such a model for the dynamics of liquid hydrogen has, so far, not been used for thermal-neutron scattering and transport studies.

There have been several attempts[5-8] to explain the total neutron-scattering cross sections of thermal neutrons from liquid hydrogen. But all these attempts, though take into account the scattering of neutrons from energetic molecular

