Observation of Magnetic-Field-Induced Superconductivity in a Heavy-Fermion Antiferromagnet: $CePb_3$

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(Received 18 April 1985)

We report here an entirely new phenomenon, namely magnetic-field-induced superconductivity in $CePb_3$, a system which at zero field is a heavy-fermion antiferromagnet. This phenomenon is novel in several respects. It is the first reported heavy-fermion magnetic-field-induced superconductor. It is also the first reported magnetic-field-induced superconductor that is also an antiferromagnet. Moreover, it has the simplest crystal structure, Cu_3Au, of any known heavy-fermion or magnetic-field-induced superconductor.

PACS numbers: 75.20.Hr, 72.15.Gd, 75.40.–s

Recently a great deal of interest has centered on the phenomenon of heavy fermions in both cerium and uranium systems.[1] Although the behavior of such systems is far from fully established, that feature which characterizes all heavy-fermion systems is an extremely large low-temperature electronic coefficient of specific heat, γ, which exceeds that of typical metals by two orders of magnitude. Systems that satisfy this criterion include superconductors, normal nonmagnetic metallic compounds, and magnetically ordered compounds. Here we report on an entirely new behavior, namely, a heavy-fermion system which undergoes an antiferromagnetic transition and then at sufficiently low temperatures is induced into the superconducting state by the application of a large magnetic field.

The new system reported here is $CePb_3$. X-ray powder patterns show that the sample is single phase and has the cubic Cu_3Au structure with $a = 4.872$ Å. Samples were prepared in an inert arc furnace and annealed at 500 °C for seven days. Extra Pb was added to compensate for weight losses (about 2%) that occurred during meltings. The specific heat has been measured in zero field and at 11 T. A standard adiabatic-heat-pulse technique was employed for the $H = 0$ measurements down to 1.5 K. A thermal-time-constant method was used for the $H = 0$ measurements below 1.5 K and for the 11 T measurements. Results obtained by the two methods were in good agreement in their region of overlap, $1.5 < T < 4$ K at $H = 0$. The resistivity measurements from 1.5 to 300 K were made using a four-probe dc technique. The low-temperature magnetoresistance was measured with use of a four-probe ac technique.

Figure 1 shows the specific heat, C, versus temperature, T, from $T \sim 0.6$ to 4 K for $H = 0$ and $H = 11$ T. The $H = 0$ curve has a sharp peak near 1.1 K. It is nearly constant from $T = 4$ K down to $T \sim 1.3$ K at a value of 1.8 J/(mol Ce) · K, then rises to a peak value of ~ 3.8 J/(mol Ce) · K at 1.1 K. (Hereafter, we use abbreviated units, J/mol · K.) Below 1.1 K it decreases rapidly, falling to 0.95 J/mol · K at 0.6 K, the lowest temperature datum point. The $H = 11$ T curve, on the other hand, does not show a peak in this temperature interval. Clearly, the specific heat peak for $H = 0$ is associated with a phase transition which is destroyed by the application of an 11 T magnetic field. If this transition is antiferromagnetic in nature one should expect an entropy removal of $R \ln 2$. If, on the other hand, it is a superconducting transition, one should expect a specific heat jump $\Delta C_N \sim 1.4 C(T_c^+)$ at

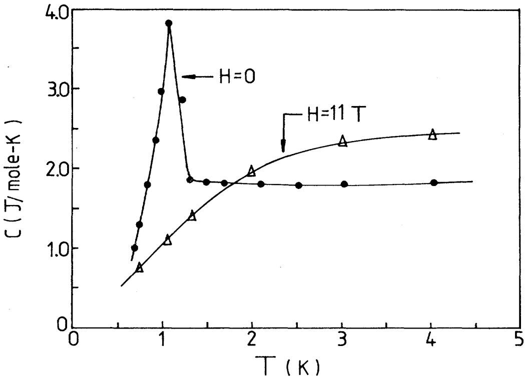

FIG. 1. Specific heat vs T at $H = 0$ and $H = 11$ T.

$T_c = 1.1$ K. It is difficult to estimate the entropy removal because C/T at the lowest temperature attained (~ 0.6 K) is still quite large. However, the apparent specific heat jump is ~ 2 J/mol·K, or about 80% of the $1.4 \times 1.8 = 2.52$ J/mol·K value that one should expect if the transition at 1.1 K is a superconducting transition. Since the data shown in Fig. 1 are taken at intervals of ~ 0.1 K near the transition it is entirely possible that the specific heat exceeds the maximum value measured in this experiment and that ΔC is in fact equal to ~ 2.52 J/mol·K. However, we have strong reasons to doubt that the peak at 1.1 K is due to a superconducting transition.

First, ac susceptibility of a $CePb_3$-powder sample failed to indicate superconductivity down to ~ 0.6 K. Second, low-field (1 T) resistivity measurements show a marked fall in resistivity from about 30 $\mu\Omega$ cm above 1.1 K to about 0.5 $\mu\Omega$ cm at 0.2 K, as one might expect for an antiferromagnetic metal which has strong spin-disorder scattering above T_N. However, given the sharpness and magnitude of the specific-heat peak it is difficult to believe that the finite value of resistivity at 0.2 K is due to an incomplete superconducting transition. Incomplete superconducting transitions measured resistively require the presence of enough normal-phase material to prevent the existence of even one completely superconducting current path in the sample. This seems highly unlikely, since one must conclude that at least 80% of the sample is superconducting if one interprets the specific-heat behavior as due to a 1.1-K superconducting transition. It should be noted, however, that the resistivity measurements had to be made in a field of 1 T in order to quench surface superconductivity associated with a thin layer of free Pb. It is, of course, possible that superconductivity at $H=0$ occurs at some temperature below 0.6 K. Third, the specific heat and susceptibility of $Ce(Pb_{1-x}In_x)_3$ samples have been measured.[2] In the range $x = 0.2$ to 0.5 specific-heat peaks are observed at temperatures above 2 K, allowing us to measure the susceptibility using a vibrating-sample magnetometer. Susceptibility measurements on these samples clearly demonstrate that the specific-heat peaks are due to antiferromagnetism, not superconductivity. A plot of T_N vs In concentration smoothly extrapolates to 1.1 K as $x \rightarrow 0$, predicting that $CePb_3$, in fact, should have an antiferromagnetic transition near 1.1 K. We are currently preparing to measure the dc susceptibility and perform neutron-scattering experiments on $CePb_3$ down to ^3He temperatures; however, it seems clear that the preponderance of evidence suggests that the transition at 1.1 K is, in fact, antiferromagnetic, not superconducting.

Another important issue concerning the nature of $CePb_3$ is whether or not it is a heavy-fermion system. As stated earlier, heavy-fermion behavior is characterized by unusually large values of the electronic coefficient of specific heat. In a typical metal $C = \gamma T + \beta T^3$ at low temperatures and both γ and β are temperature independent if $T \ll T_F$ and $T \ll \Theta_D$, respectively. Hence, plots of C/T vs T^2 give straight lines of slope β and $T=0$ intercept, γ. In heavy-fermion systems, however, C/T-vs-T^2 plots often show upswings at temperatures below 5 K.[1] Shown in Fig. 2 is C/T vs T^2 from $T^2 = 0.36$ to 16 K^2 ($T = 0.6$ to 4 K), for $H=0$. Also shown in the inset is C/T vs T^2 from $T^2 = 2.25$ to 100 K^2 ($T = 1.5$ to 10 K) for the $H=0$ data. The data shown in the inset are very similar to that observed in other heavy-fermion systems. In such systems γ is taken to be the low-temperature value of C/T, not the much smaller intercept value obtained by extrapolating the linear portion of the curve to $T^2 \rightarrow 0$. However, in the case of $CePb_3$, a portion of the upswing in C/T may be due to the presence of the antiferromagnetic transition at 1.1 K. This complicates the problem of extracting a value for γ. However, several points should be observed. First, the value of γ obtained by extrapolating from the linear region ($T^2 = 25$ to 100 K to $T^2 \rightarrow 0$) should represent a lower limit for γ and yet gives a very high value of 200 mJ/mol·K^2. This value should be compared to $LaPb_3$, for which we measured $\gamma \sim 1$ mJ/mol·K^2. Hence, there is no doubt that the f electrons in $CePb_3$ have greatly enhanced masses. It should be noted that C/T vs T^2 for $LaPb_3$ was linear from $T^2 = 2.25$ to 100 K^2. Second, looking at Fig. 1 the constant value of C from 1.3 to 4 K and the abrupt rise in C from 1.3 to 1.1 K suggests that contributions to C coming from the antiferromagnetic transition may be negligible above ~ 1.3 K. If so, one should explain the constant (at $H=0$) C-vs-T behavior of $CePb_3$ from 1.3 to 4 K as due to a constant heavy-fermion contribution to C in this interval. This type of heavy-fermion behavior has been observed in other systems.[1] The essentially constant specific heat gives rise to an

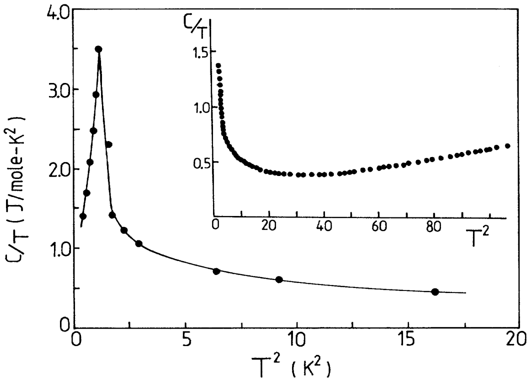

FIG. 2. Specific heat coefficient C/T vs T^2 at $H=0$. Data up to 10 K shown in the inset.

upswing in C/T as T is lowered. If this interpretation is correct, then γ of $CePb_3$ reaches a value as high as $C(T=1.3)/1.3\ K = 1400\ mJ/mol\cdot K^2$. On the other hand, if one wishes to attribute the entire upswing in C/T at low temperatures to the presence of an antiferromagnetic transition at 1.1 K rather than to a heavy-fermion contribution to the electronic specific-heat coefficient, then one must explain the constancy of the specific heat itself in the 1.3 to 4 K range in a highly implausible and artificial fashion. In particular, if the electronic specific heat coefficient, γ, is temperature independent, then the antiferromagnetic contribution to C would have to decrease linearly from 1.3 to 4 K so that the sum of this contribution, and the electronic contribution, γT, is constant in this range. Since magnetic contributions to the specific heat above T_N are not expected[3] to be substantial at $(T-T_N)/T_N \sim 3$, nor are they expected[3] to decrease linearly from $(T-T_N)/T_N \sim 0.2$ to 3, this explanation is highly unlikely.

Although a precise value for the electronic specific heat coefficient is difficult to determine for the reasons alluded to above, nonetheless, it is clear that $CePb_3$ has a γ value of at least $200\ mJ/mol\cdot K^2$ (a factor of 200 times larger than that of $LaPb_3$) and it might be as large as $1400\ mJ/mol\cdot K^2$. Moreover, $CePb_3$ becomes antiferromagnetic at 1.1 K in zero magnetic field. A field of 11 T depresses T_N to below 0.7 K and most likely to $T=0$. Hence, $CePb_3$ is a new heavy-fermion system which becomes antiferromagnetic at $H=0$.

Another property of heavy-fermion systems which is of considerable interest is electrical resistivity. Figure 3 shows ρ_{mag} vs temperature from 1.5 to 300 K for $CePb_3$. Here ρ_{mag} is taken to be the total resistivity minus the phonon contribution. The latter is assumed to be identical to the temperature-dependent resistivity of $LaPb_3$. As noted above, the data of Fig. 3 were taken in a field of ~ 1 T in order to quench the surface superconductivity which invariably is present in Pb intermetallic compounds as a result of the presence of free Pb on the surface of the sample. The behavior shown in Fig. 3, namely two peaks in ρ_{mag}, one near 25 K, the other near 2.5 K, and a sharp falloff at the lowest temperatures, is very similar to that seen in the case of UBe_{13}, a heavy-fermion superconductor.[4] The low-temperature resistivity of $CePb_3$ (at 1 T), as shown in the inset of Fig. 3, falls substantially below 1 K, from $\sim 30\ \mu\Omega$ cm at 1 K to $\sim 0.5\ \mu\Omega$ cm at 0.2 K. As mentioned earlier, this drop is consistent with the freezing out of a large spin-disorder resistivity as the degree of antiferromagnetic ordering increases below T_N.

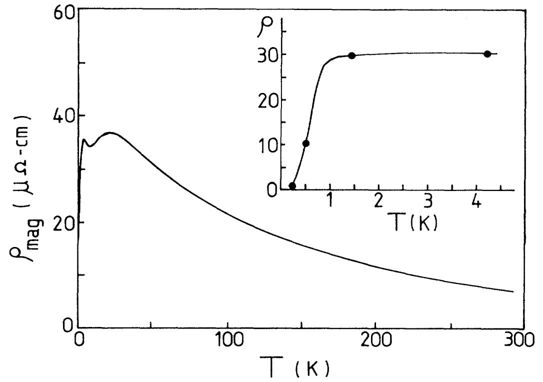

FIG. 3. The magnetic resistivity vs temperature at $H=1$ T for $CePb_3$. The inset shows ρ vs T at $H=0.93$ T for $T=0.2$ to 4 K.

Another way to alter spin-disorder scattering is to apply a uniform magnetic field. Above T_N one expects that a uniform magnetic field will reduce the spin-disorder scattering. Below T_N, however, one expects ρ to first increase until the field destroys the antiferromagnetic state and then to decrease at higher fields. Shown in Fig. 4 is ρ vs H for some temperatures both above and below T_N. Note that at $T=4.2$ and 1.42 K (i.e., $T > T_N = 1.1$ K) ρ monotonically decreases with increasing H while at $T=0.48$ and 0.20 K ρ first increases and then decreases. The resistivity is maximum at $H=4.5$ and 5.0 T at $T=0.48$ and 0.20 K, respectively. These fields are probably very close to the fields necessary to destroy antiferromagnetism at these temperatures. Note that at fields near 15 T the resistivity at both $T=0.48$ and 0.20 K is very small. Figure 5 shows ρ vs H at 0.20 K on a greatly expanded resistivity scale. At a field of 14 T we have determined that the resistivity is zero to within our resolution, i.e., $\rho < 0.005\ \mu\Omega$ cm, which corresponds to a mean free path $\lambda > 70\,000$ A, or a residual resistivity ratio (RRR) in excess of about 8000! Since these λ and RRR values are two to three orders of magnitude greater than those found for intermetallic compounds which are not superconducting, it seems safe to con-

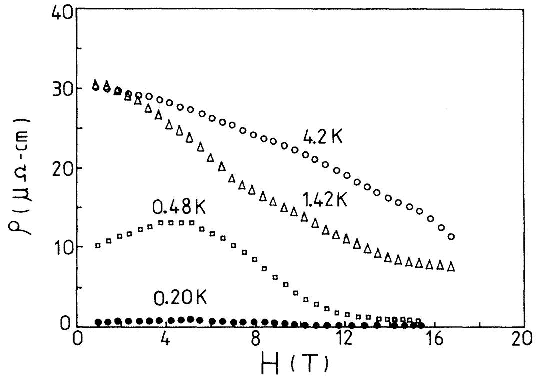

FIG. 4. Resistivity vs field at 4.2, 1.42, 0.48, and 0.20 K.

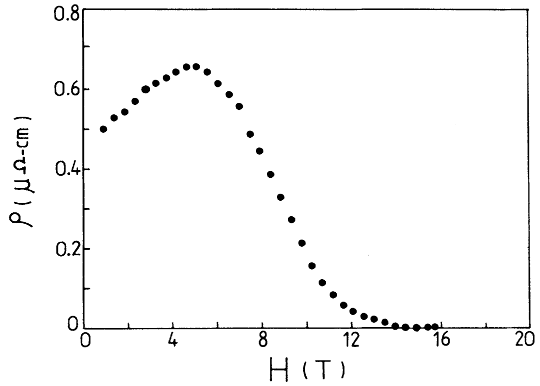

FIG. 5. Resistivity vs field at 0.20 K.

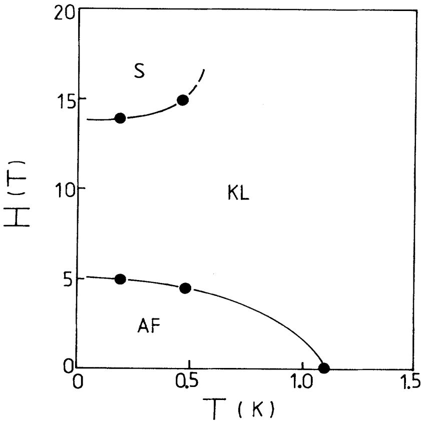

FIG. 6. Tentative phase diagram of $CePb_3$ in the magnetic-field–temperature plane. AF, S, and KL stand for antiferromagnetic, superconducting, and Kondo lattice.

clude that at 0.20 K a magnetic field of 14 T induces the system into the superconducting state. Similarly, at 0.48 K, 15 T drives the sample superconducting. The phenomenon of magnetic-field-induced superconductivity has been seen in other systems, particularly in Eu Chevrel phase systems.[5] This is, however, the first observation of the effect in a heavy-fermion system and also the first observation of magnetic-field-induced superconductivity in a simple cubic system.

If one takes the field at which ρ is maximum as determining the magnetic-field-dependent Néel temperature, $T_N(H)$, and the field at which ρ vanishes as giving the field-induced superconducting transition temperature, $T_c(H)$, then one obtains the H-T phase diagram shown in Fig. 6. Contrasting this diagram to that observed in the Chevrel phase magnetic-field-induced superconducting systems[5] the latter have a low-field superconducting phase rather than the antiferromagnetic phase observed here for $CePb_3$. It should be pointed out, however, that our ρ measurements were not done at zero field but rather at 1 T because of the problem of free-surface Pb. It is conceivable that at $H=0$ $CePb_3$ undergoes an antiferromagnetic transition at 1.1 K followed by a superconducting transition below 0.6 K, the lowest temperature at which the ac susceptibility was measured. The $T=0$ critical field of this superconducting phase might be less than 1 T.

Although a great deal of work needs to be done in order to establish the nature of the field-induced superconductivity of $CePb_3$, it seems reasonable to assume that it may arise from the Jaccarino-Peter[6] mechanism proposed for the Chevrel phase systems. This mechanism requires a negative conduction-electron exchange field which can then be compensated by an effective field arising from the ferromagnetic alignment of spins in a large applied field. In any event, both antiferromagnetism and field-induced superconductivity have now been observed in a heavy-fermion system.

This work was supported by the National Science Foundation through Grant No. DMR-8219782, by the National Magnet Laboratory, MIT, and by the U.S. Department of Energy.

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[1] G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984), and references therein.

[2] C. Lin, J. E. Crow, and T. Mihalisin, to be published.

[3] See H. Eugene Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford Univ. Press, New York, 1971).

[4] H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **43**, 1595 (1983).

[5] H. W. Meul, C. Rossel, M. Decroux, O. Fisher, G. Remenyi, and A. Briggs, Physica (Utrecht) **126B**, 44 (1984).

[6] V. Jaccarino and M. Peter, Phys. Rev. Lett. **9**, 290 (1962).