Magnetic Ground State of a Heavy-Electron System: U_2Zn_{17}

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Measurements of the magnetic susceptibility, specific heat, and electrical resistivity of U_2Zn_{17} reveal a magnetic phase transition of this compound at 9.70 K. Details of the specific heat indicate that the ordering occurs among electrons whose energy spectrum shows an anomalously large density of states at the Fermi energy. This result is of interest with regard to the occurrence of superconductivity in similar systems.

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The recent discovery of superconductivity in UBe_{13} involving electrons of extremely heavy effective masses[1] has renewed the interest in the ground-state properties of such systems, where electron-electron interactions appear to dominate the behavior of physical properties at low temperatures. Characteristic representatives of these materials were almost exclusively found before among Ce intermetallic compounds such as $CeAl_3$ which remains a normal conductor down to a few millikelvins[2] and $CeCu_2Si_2$ which is a superconductor below about 0.65 K.[3] In both $CeCu_2Si_2$ and UBe_{13} superconducitivity occurs in an electronic system with extraordinary properties like very large densities of electronic states at the Fermi energy E_F and, as mentioned above, abnormally heavy effective electron masses.

In UBe_{13}, various experimental facts suggest that superconductivity is not primarily due to the conventional electron-phonon interaction. Above the superconducting transition T_c, the coefficient of the linear term in the temperature dependence of the specific heat, usually denoted as the electronic specific heat, increases rapidly with decreasing temperature from about 0.18 J/K^2 mole above 7 K to about 1 J/K^2 mole around 1 K,[4] a behavior that is also observed in $CeAl_3$,[4] and, particularly, in liquid ^3He in the normal state below its effective Fermi temperature T_F^*.[5] In the latter case it is known that strong spin fluctuations stabilize the superfluid state observed at very low temperatures.[6] Another indication that non-electron-phonon interactions are very important in this temperature range is obtained from the temperature and magnetic field dependence of the electrical resistivity around and below 1 K.[7] If indeed some kind of magnetic interactions are responsible for the low-temperature properties of UBe_{13}, it is conceivable that other materials exist where the balance of these interactions trigger a magnetically ordered ground state of the heavy-mass electron system. In this Letter we demonstrate that such a case has been found in U_2Zn_{17}, another uranium intermetallic compound.

Structural and magnetic properties of U_2Zn_{17} have been studied before. The crystal structure of U_2Zn_{17} was first reported by Makarov and Vinogradov,[8] in comparison with the neighboring compound Th_2Zn_{17}. While these authors claimed different crystal structures for the two compounds, later work by Vold and Peterson[9] and by Johnson, Smith, and Wood[10] questioned this statement and claimed the same structure for U_2Zn_{17} and Th_2Zn_{17}. Magnetic measurements by Misiuk, Mulak, and Czopnik[11] indicated a temperature-independent magnetic susceptibility below about 10 K and hence a nonmagnetic ground state of U ions was proposed by these authors. Our experimental data shown below indicate a magnetic phase transition occurring in a system of electrons with rather large effective mass.

Samples of U_2Zn_{17} were obtained in polycrystalline form by melting the proper amounts of U and Zn in our outgassed BeO crucible enclosed in a quartz tube at about 1000 °C and subsequent slow cooling down to room temperature. A careful x-ray inspection revealed the proper Th_2Zn_{17} structure with space group $R\bar{3}m$ and lattice parameters $a = 8.9859(1)$ Å and $c = 13.155(3)$ Å.[12] For reasons of comparison a polycrystalline sample of Th_2Zn_{17} was prepared in the same way as mentioned above.

In Fig. 1 we show the temperature dependence of the inverse magnetic susceptibility χ^{-1} of U_2Zn_{17}

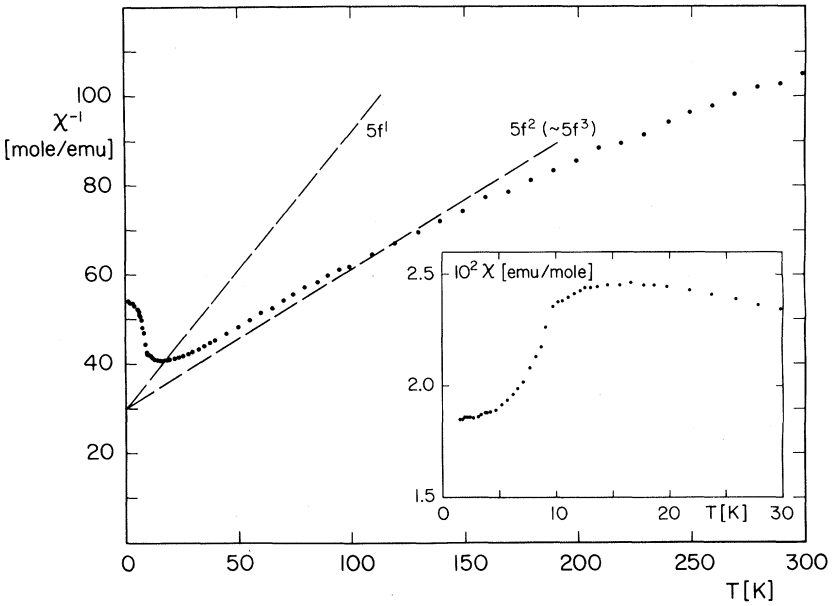

FIG. 1. Temperature dependence of the inverse magnetic susceptibility χ^{-1} of U_2Zn_{17} between 1.5 and 300 K.

between 1.5 K and room temperature. Also indicated are the expected slopes of $\chi^{-1}(T)$ for different integral occupations of the $5f$ shells of the U ions. Obviously the possible slopes never fit the experimental results, a fact which may have various reasons but is not of primary interest here. Also shown, as an inset in Fig. 1, is $\chi(T)$ between 1.5 and 30 K. It reveals a broad maximum around 16 K and an accentuated drop below 9.8 K. We note that $\chi(T)$ per U ion is very similar to that of UBe_{13}, at least between room temperature and 10 K. The data shown in Fig. 1 suggest a magnetic phase transition just below 10 K but they are certainly not sufficiently conclusive.

Clear evidence, however, for a distinct second-order phase transition is obtained from the temperature dependence of the specific heat $c_P(T)$ in this temperature range as shown in the upper part of Fig. 2. The discontinuity in $c_P(T)$ fixes the transition temperature at 9.70 K. Also shown in Fig. 2 is the specific heat of Th_2Zn_{17} which allows us to determine the lattice contribution to c_P, assumed to be equal in U_2Zn_{17} and Th_2Zn_{17}. Subtraction of this lattice contribution leads to the result shown in the lower part of Fig. 2, where

$$c_p^{\mathrm{el}}(U_2Zn_{17})/T = [c_p^{\mathrm{tot}}(U_2Zn_{17}) - c_p^{\mathrm{latt}}(Th_2Zn_{17})]/T \quad (1)$$

is plotted versus T^2. From its temperature dependence, c_p^{el} may be identified as an electronic contribution. Above the transition temperature, c_p^{el} clearly varies linearly with temperature and the resulting γ parameter, denoting the electronic specific heat in the paramagnetic region, is anomalously large, namely $\gamma_p = 1.07$ J/K^{2}mole. Since one formula unit contains two U atoms, this γ value has to be considered as about half as large as that of UBe_{13}. Below the magnetic phase transition which, according to $\chi(T)$, must be of antiferromagnetic nature, the contribution to the specific heat which varies linearly with temperature is obviously reduced to $\gamma_0 = 0.395$ J/K^{2}mole but is still considerably larger than in any normal metal. In addition we find, at the lowest temperatures, a residual contribution to c_p that varies as T^3.

Finally we also show the temperature dependence of the electrical resistivity ρ between 1.2 K and room temperature in Fig. 3. $\rho(T)$ steadily increases with decreasing temperature below 300 K. It reaches a maximum between 17 and 18 K and subsequently decreases with increasing slope through the magnetic transition at $T_N = 9.7$ K. Finally ρ reaches a value of about $\rho_{\max}/80$ at 1.2 K. At the lowest temperatures attained here, ρ varies as $T^{5/2}$.

The same features as we report here have partially been observed before in other intermetallic compounds containing either rare-earth or actinide ions. The most striking similarity with our findings, especially for $c_p(T)$, was obtained in $NpSn_3$.[13] In the latter case, the electronic specific-heat coefficients above and below the phase transition were of the order of 250 and 90 mJ/K^{2}mole, respectivley. The relative discontinuity $\Delta c_p/c_p$ at the transition was of

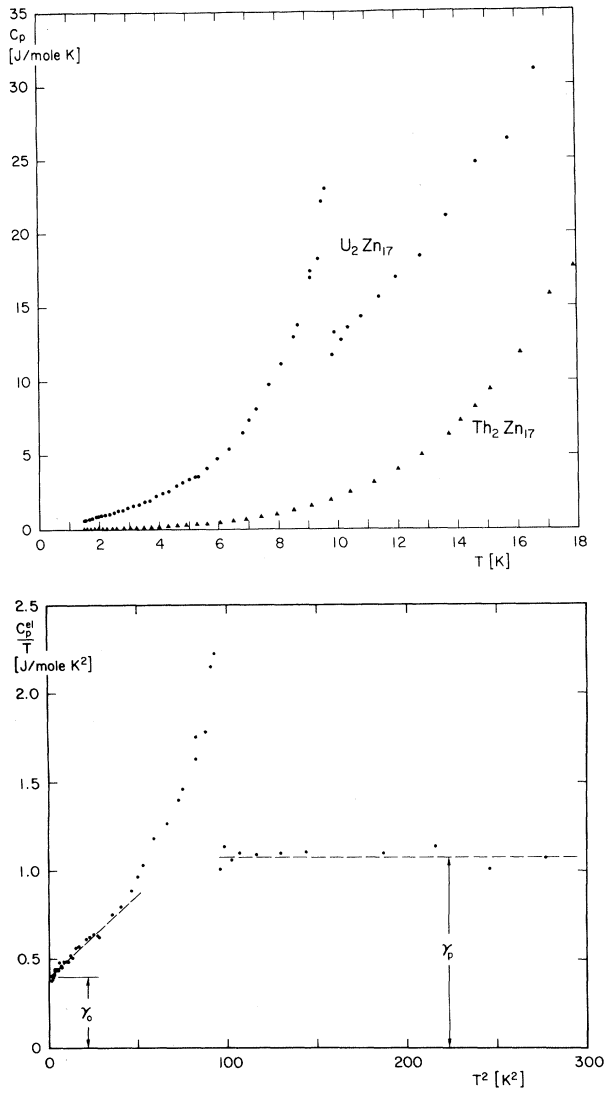

FIG. 2. Upper part, temperature dependence of the specific heats of U_2Zn_{17} and Th_2Zn_{17} between 1.5 and 18 K. Lower part, specific heat minus the lattice contribution of U_2Zn_{17} at low temperatures.

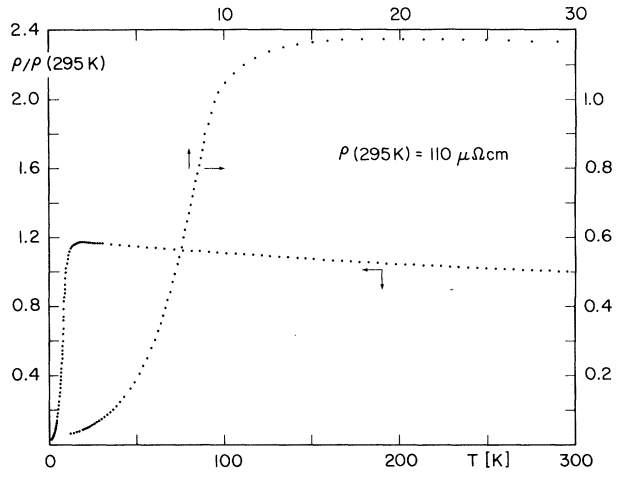

FIG. 3. Temperature dependence of the electrical resistivity of U_2Zn_{17} between 1.2 and 300 K. Note the different scales for different temperature intervals.

the same order of magnitude as ours, although smeared out over several kelvins. On the basis of these results, Trainor *et al.*[17] then claimed good agreement between experiment and a specific-heat anomaly as predicted by the BCS theory of superconductivity,[14] thus confirming the suggested itinerant-electron antiferromagnetism in $NpSn_3$. In our opinion, this conclusion was erroneous because the experimental data were not compared with the correct BCS prediction for the quantity c_p^{el}/T that was plotted in Fig. 3 of Ref. 11, but rather with the theoretical temperature dependence of c/c_n where $c_n = \gamma T_c$.[15] We agree, however, with the claim of magnetic order among electrons with a large density of states at E_F for which U_2Zn_{17} is an even more impressive example. Also in $NpSn_3$ and more recently in CePt,[16] nonlatttice T^3 contributions to c_p were found below the magnetic ordering temperature.

In an early theory of itinerant-electron antiferromagnetism, Fedders and Martin[17] demonstrated analogies between this phenomenon and superconductivity. For U_2Zn_{17}, these analogies, at least in part, do not seem to be valid. This may be seen from the fact that the entropy balance through the transition is not zero. The entropy associated with the electronic specific heat γ_0 below T_N amounts to 3.84 J/K mole while for the remaining part up to the ordering temperature we find 3.905 J/K mole. The sum of these two contributions is, however, considerably less than the entropy of 10.48 J/K mole associated with the electron system in the paramagnetic state which should be available between T_N and 0 K , if the transition did not occur. The magnetic entropy of the transition is also much less than $2R \ln 2 = 11.52$ J/K mole. Such small specific-heat anomalies were found before in induced-moment magnets with a singlet ground state of the localized f electrons.[18] Although this possibility might occur in U_2Zn_{17}, it is in fact the absence of any high-temperature tail in the specific-heat anomaly usually observed in these latter cases which leads us to discard it here. This obvious absence of fluctuations above the critical temperature, in contrast to the remarks made above, indeed emphasizes the analogy of this phase transition with the general features of supercon-

ducting transitions.

Another extraordinary feature is the large resistance drop below the transition. We recall that the other already mentioned similar materials like $CeAl_3$, $CeCu_2Si_2$, and UBe_{13} all also exhibit rather large resistivities of the order of 100 $\mu\Omega$ cm at room temperature and ρ still increases with decreasing temperature, as shown in Fig. 3. In all these examples $\rho(T)$ attains a maximum value and then decreases at even lower temperatures. Even without a cooperative phase transition, superconducting or magnetic, the ground state of these systems is characterized by a low electrical resistivity, as observed very clearly in $CeAl_3$.[2] A theoretical treatment of these systems will have to attempt to describe an almost localized behavior at high temperatures and a change to metallic behavior at very low temperatures. In addition it should allow for cooperative phase transitions of the electronic subsystem.

In conclusion we have shown that in metallic materials exhibiting enormous and strongly renormalized densities of electric states, in addition to the known paramagnetic normal conducting and the superconducting ground states also magnetically ordered ground states seem possible. This supports ideas that in such materials superconductivity and magnetism result from the same interactions and that it is only the ratio of the strength of these interactions that determines which ground state is finally adopted.

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