

Resistance study on the itinerant ferromagnet CrBe<sub>12</sub> under pressure

X. Y. Shao,\* T. H. Lin, P. H. Hor, M. K. Wu, and C. W. Chu

*Department of Physics and Energy Laboratory, University of Houston, Houston, Texas 77004*

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The electrical resistance of CrBe<sub>12</sub> has been measured between 1.2 and 300 K under hydrostatic pressure up to 20 kbar. The results demonstrate the existence of two kinds of phase transition from the paramagnetic state on cooling under pressure: one below  $\sim 11$  kbar and the other above, in agreement with previous high-pressure magnetic study.

The body-centered tetragonal intermetallic compound CrBe<sub>12</sub> has been found<sup>1</sup> to order ferromagnetically with a Curie temperature of 50 K at ambient pressure. Detailed magnetic studies<sup>2</sup> suggested that CrBe<sub>12</sub> might be an itinerant ferromagnet. Recent ac magnetic susceptibility measurements<sup>3</sup> showed that in addition to the paramagnetic  $\rightarrow$  ferromagnetic transition previously reported at  $T_{C1} \sim 50$  K, a pressure-induced transition also exists in CrBe<sub>12</sub> at lower temperature  $T_{C2}$ . A  $T$ - $P$  phase diagram of CrBe<sub>12</sub> was thus obtained. The close similarity of this CrBe<sub>12</sub> phase diagram to that<sup>4</sup> of the Cr-based alloys has led to the postulation<sup>3</sup> that interesting and complex magnetic structures might have occurred in CrBe<sub>12</sub>. It is known<sup>4</sup> that the Cr+0.2 at.% Ru alloy undergoes a sequence of I (paramagnetic)  $\rightarrow$  II (commensurate)  $\rightarrow$  III (incommensurate) antiferromagnetic transitions at low temperature and low pressure and a direct I  $\rightarrow$  III transition at low temperature and high pressure. The resistance anomalies associated with the transitions in the Cr alloys are also known<sup>4</sup> to be characteristically different, depending on the nature of the transition, i.e., I  $\rightarrow$  II or I  $\rightarrow$  III. In an attempt to gain insights to the nature of the transitions in CrBe<sub>12</sub>, we have measured the electrical resistance  $R$  of the CrBe<sub>12</sub> compound between 1.2 and 300 K under hydrostatic pressures up to 20 kbar. Only one  $R$  anomaly signaling a phase transition has been detected throughout our temperature and pressure ranges. However, the  $R$  anomaly below  $\sim 11$  kbar is characteristically different from that above  $\sim 11$  kbar. The results suggest that the phase transitions so detected in these two pressure regions are different in nature, consistent with the phase diagram previously determined magnetically.

The CrBe<sub>12</sub> samples examined in the present study were pieces broken off the same arc-melted ingot investigated previously.<sup>3</sup> A standard four-lead ac technique operating at 23 Hz was employed to measure the  $R$ . The four Pt electrodes were soldered ultrasonically with In onto the samples. The hydrostatic pressure environment was provided by a Be-Cu clamp,<sup>5</sup> using the 1:1 fluid mixture of *n*-pentane and isoamyl alcohol as the pressure medium. The pressure was determined with a Pb manometer situated next to the sample, and the temperature by a Chromel-Alumel thermocouple above 20 K and a Ge thermometer below.

We have measured the  $R$  of three CrBe<sub>12</sub> samples. The resistivity of these samples was estimated to be  $\sim 10^{-4}$   $\Omega$  cm at ambient temperature and pressure, with a  $\sim 100\%$  uncertainty due to the irregular shape of the samples, resulting from the extreme brittleness of the compound. Therefore, only  $R$  will be discussed in this investigation.

Typical temperature dependence of  $R$  normalized to that at 78.5 K is shown in Fig. 1 for some of the experimental runs under various pressures. At ambient pressure,  $R$  decreases almost linearly with temperature  $T$  from 300 K down to  $\sim 50$  K, but displays a drastic change in slope below  $\sim 50$  K. Such a drastic change in slope occurs at a temperature where a paramagnetic  $\rightarrow$  ferromagnetic transition has been detected<sup>1,3</sup> magnetically. We, consequently, associate this sudden slope change of the  $R$ - $T$  curve with the ferromagnetic ordering at ambient pressure. The inflection point temperature<sup>6</sup> is taken as the phase transition temperature  $T_0$ , which is  $45 \pm 1.5$  K at ambient pressure in contrast to 46 K, determined by the ac magnetic susceptibility anomaly. Under pressure,  $T_0$  is shifted reversibly toward lower temperature, as summarized in Fig. 2, but no other  $R$  anomaly

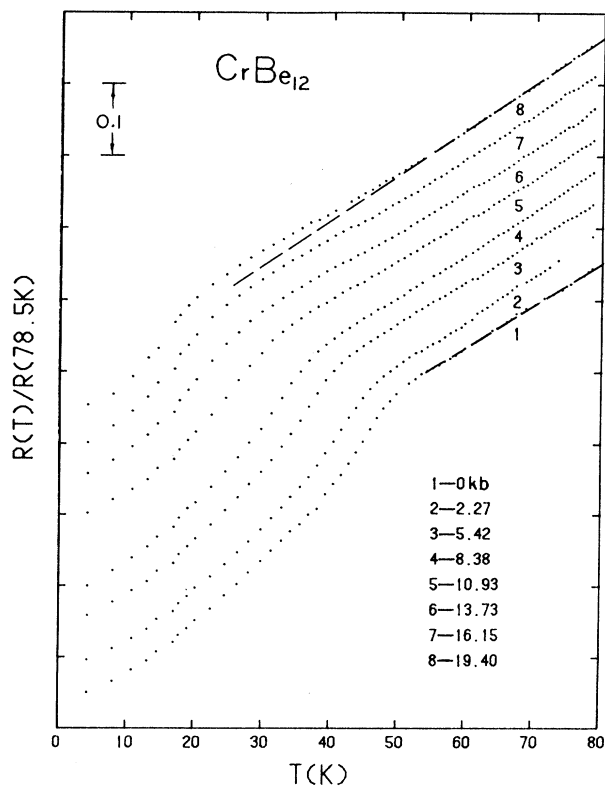


FIG. 1. The temperature dependence of resistance at different pressures. Note the difference in curvature near the transition below and above  $\sim 11$  kbar.

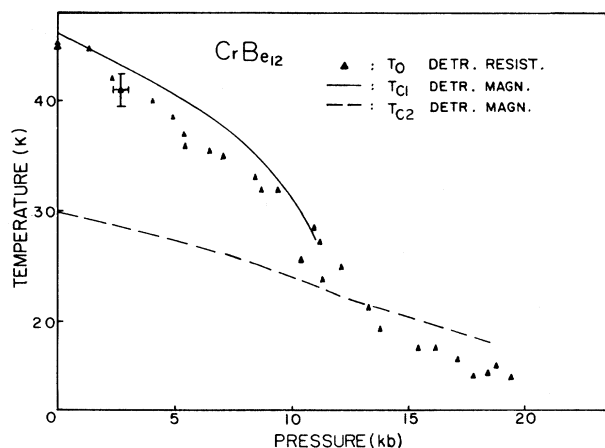


FIG. 2. The temperature-dependence diagram for  $\text{CrBe}_{12}$  determined resistively in the present study and magnetically previously (Ref. 3).

indicative of the second phase transition, observed magnetically at low temperature and pressure, has been detected unambiguously. However, a close examination of the  $R$ - $T$  curves, e.g., those in Fig. 1, revealed that  $R$  exhibits a subtle different  $T$  behavior near the phase transition depending on the pressure applied. For instance,  $R$  always shows a negative curvature on cooling following the initial linear  $T$  dependence of  $R$  and before entering the phase transition when the pressure is below  $\sim 11$  kbar, whereas a positive curvature when above  $\sim 11$  kbar (see the dashed lines as a guide in Fig. 1). This observation suggests that the phase transitions below and above  $\sim 11$  kbar may be of different nature, in agreement with the phase diagram previously proposed.<sup>3</sup> The clear break in slope of the  $T_0$ - $P$  plot shown in Fig. 2 lends further support to such a suggestion. Results from the earlier magnetic study of  $\text{CrBe}_{12}$  were also included as curves in Fig. 2 for comparison. The solid curve represents the I-II phase boundary with a transition temperature  $T_{C1}$ , and the dashed curve denotes the II-III or I-III phase boundary with a transition temperature  $T_{C2}$ . The slight disagreement between  $T_0$  and  $T_{C1}$  or  $T_{C2}$  is common for a transition temperature when determined by different methods.

The  $T_{C1}$ - $P$  curve, separating the paramagnetic I phase from the itinerant ferromagnetic II phase in Fig. 2, has been shown<sup>3</sup> to fit well the expression  $T_{C1}(P) = T_{C1}(0) \times (1 - P/P_c)^\tau$  with  $T_{C1}(0) = 46.2$  K,  $P_c = 22.3$  kbar, and  $\tau = 0.244$ . The above expression was originally proposed<sup>7</sup> to account for the pressure effect on the Curie temperature of an itinerant ferromagnet but with  $\tau = 0.5$  (or 0.75 when spin fluctuation dominates). The nature of phase III remains enigmatic. By comparing this phase diagram with that of

the  $\text{Cr} + 0.2$  at. % Ru, it was therefore postulated<sup>3</sup> that the I, II, and III phases in  $\text{CrBe}_{12}$  might be associated with the paramagnetic, commensurate (or incommensurate) ferromagnetic, and incommensurate (or commensurate) ferromagnetic states, respectively.

In the present study, the characteristic of the  $R$  anomaly accompanying the phase transition and the  $P$  dependence of  $T_0$  demonstrates that two different types of phase transition are observed, one below  $\sim 11$  kbar and the other above. They correspond to the I  $\rightarrow$  II and I  $\rightarrow$  III transitions in Fig. 2, respectively. The failure to detect resistively the II  $\rightarrow$  III transition at low temperature below  $\sim 11$  kbar may be attributed to our inability to separate the weak  $R$  anomaly associated with the II  $\rightarrow$  III transition from that with the I  $\rightarrow$  II transition. It should be noted that the II  $\rightarrow$  III phase transition also failed to be detected<sup>4</sup> resistively in the  $\text{Cr} + 0.2$  at. % Ru alloy.

In spite of the gross similarity between the  $T$ - $P$  phase diagrams of the  $\text{CrBe}_{12}$  compound and Cr-based alloys, difference does exist between the two systems. The  $R$  anomaly in the Cr-based alloys displays a rapid rise (first order for the I  $\rightarrow$  II transition and second order for the I  $\rightarrow$  III transition) following the formation of spin-density waves associated with the removal of conduction electrons from the Fermi surface resulting from the nesting of the electrons and holes. On the contrary, the  $R$  anomaly for  $\text{CrBe}_{12}$  always exhibits a drop. The difference may be ascribed to the different magnetic long-range orders involved in the two systems: ferromagnetic in  $\text{CrBe}_{12}$  and antiferromagnetic in the Cr-based alloys. An  $R$  drop is typical of a ferromagnetic ordering as a result of the removal of the spin-disorder scattering.

In conclusion, the present resistance study under pressure on the  $\text{CrBe}_{12}$  compound shows that there are two types of phase transition: one below  $\sim 11$  kbar and the other above, in agreement with the earlier magnetic investigation under pressure. While difference exists between the  $\text{CrBe}_{12}$  compound and the Cr-based alloys, the similarity between the  $T$ - $P$  phase diagram cannot be ignored. The different electron-scattering processes near the I  $\rightarrow$  II and I  $\rightarrow$  III transition in  $\text{CrBe}_{12}$ , evident from the different  $R$  anomalies detected, should be taken into consideration in future model calculations. More careful transport and magnetic experiments under pressure are in progress. However, a neutron-diffraction study under pressure to determine the magnetic structure of this compound is most worthwhile.

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<sup>6</sup>We are indebted to Professor M. Croft for suggesting such a method of defining a transition temperature which yields a better agreement with the magnetically determined results.

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