High magnetic fields and the correlation gap in SmB_6

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We report the results of electrical resistivity measurements on SmB_6 in magnetic fields as large as 18 T and pressures as large as 66 kbar. We find that the activation gap Δ extracted from these measurements has an anomalously weak field dependence, even when pressures of ~ 50 kbar are used to tune Δ to instability. Our results imply the existence of strong exchange interactions in the low-pressure, gapped state, and argue that Δ originates with many-body correlations. Nonetheless, the small, negative magnetoresistance $\Delta\rho(H)/\rho_{H=0} \propto -H^2$ observed below 50 kbar is completely accounted for by the weak field variation of Δ. A sign change in the magnetoresistance accompanies the gap collapse, with the high-pressure, gapless metal having $\Delta\rho(H)/\rho_{H=0} \propto H^{3/2}$. The Fermi surface deduced from these measurements is similar to that of noninteracting, trivalent LaB_6, but with a volume which increases rapidly with pressure.

I. INTRODUCTION

Despite more than 20 years of experimental effort, many fundamental aspects of the mixed valent-Kondo intermetallic SmB_6 continue to elude understanding. Infrared absorption,[1] inelastic neutron scattering,[2] optical conductivity,[3] electron tunneling,[4] and electrical transport[5] measurements all detect a small electronic gap $\Delta = 40-100$ K. However, the low-temperature transport properties of SmB_6 are manifestly metallic, having a large but finite resistivity below ~ 4 K,[5] indicating that the Fermi level is pinned within the gap. There is considerable controversy whether these in-gap states are intrinsic, present in pristine SmB_6,[2,6,7] or extrinsic, dictated entirely by sample quality.[5,8]

We have recently performed a series of high-pressure electrical resistivity and Hall-effect measurements on SmB_6 to address this issue.[6] We find that the concentration of the gap states is extremely pressure dependent, and that these states dominate the transport to increasingly high temperatures as the gap is suppressed by pressure. We argue that the states are primarily intrinsic and share stability with that of the activation gap itself, which electrical resistivity measurements find collapses at ~ 50 kbar. Since there is no accompanying crystal symmetry change or cell volume instability at the critical pressure,[9] the disappearance of the gap cannot be interpreted as a simple band-crossing transition.

Our results indicate that the gap in SmB_6 is not a feature of the single-particle band structure, but instead arises from strong electronic correlations. What is more, the relationship between the concentration of gap states and the magnitude and stability of the gap itself is highly reminiscent of another correlation gap system, the Mott-Hubbard insulator. Here, the electronic gap originates with a strong on-site Coulomb interaction, and the insulator-metal transition is thought to be mediated by intrinsic states appearing in the preformed gap.[10] Despite these similarities, there is no microscopic reason to believe that SmB_6 is literally a Mott-Hubbard system akin to transition-metal oxides. That is, while our high-pressure transport measurements argue that Δ cannot be a feature of a noninteracting electronic structure in SmB_6, the origin of the gap remains in question. We address this issue here in a series of high-pressure resistivity measurements which assess the effects of large magnetic fields both on Δ and also on the low-temperature transport attributed to the in-gap states. A preliminary report of these results has already appeared.[11]

II. EXPERIMENTAL PROCEDURE

Single crystals of SmB_6 were prepared from an aluminum flux, and were carefully screened for a resistivity in-

crease of at least 10^4 between room temperature and 4.2 K. Pressures as large as 100 kbar were generated in a miniaturized Bridgman anvil cell. A superconducting lead manometer[12] was used initially to establish the pressure dependence of the SmB_6 activation gap and residual resistivity, which served as secondary pressure standards in subsequent high magnetic-field measurements. Magnetic fields as large as 18 T were generated in a superconducting solenoid at the National High Magnetic Field Laboratory-Pulsed Field Facility at Los Alamos National Laboratory. Measurements up to 7 T were performed in our home laboratory at the University of Michigan. In every case, the measuring current was applied in the plane perpendicular to the magnetic field, which lay along one of the principle axes of the cubic crystal. Temperatures were stabilized in field by correcting the control sensor for magnetoresistance using standard calibrations.[13] This technique was found to provide no more than 20–30 mK reproducible temperature offset in a field of 18 T at a fixed temperature of 4.2 K.

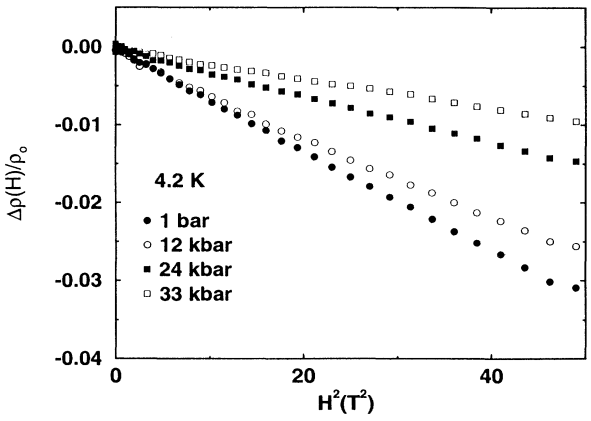

FIG. 2. Pressure dependence of the 4.2 K magnetoresistance in the low-pressure gapped state, $\Delta\rho(H) = -B(P,T)H^2$. ●=1 bar, ○=12 kbar, ■=24 kbar, □=33 kbar.

III. EXPERIMENTAL RESULTS

As we previously reported,[11] the temperature dependence of the electrical resistivity $\rho(T)$ was measured in zero field and in 18 T, at the relatively low pressure of 12 kbar, and in the vicinity of the gap collapse, 45 kbar. $\rho(T)$ is activated at both pressures and the transport gap Δ was extracted from fits to $\rho(T)=\rho_0(P)\exp(\Delta(P)/k_BT)$. Our surprising result is that the electronic gap Δ is almost completely unaffected by large magnetic fields, decreasing at 12 kbar by less than 4% from 30.7±0.4 K at $H=0$ to 29.5±0.4 K at $H=18$ T. This insensitivity to magnetic fields is even more strikingly demonstrated when a pressure of 45 kbar is used to tune Δ to the gap instability, resulting in a zero-field value of 11.6±0.2 K. Here, Δ increases ~20% from its $H=0$ value to 13.9±0.2 K at 18 T. Again, the effect of magnetic fields is surprisingly weak, since the Zeeman splitting associated with a field of 18 T and a free-electron Landé g factor of 2 is 25 K, more than twice the magnitude of the measured gap.

The detailed field dependence of the electrical resistivity measured at fixed temperature argues that the magnetoresistance has different origins at low and high pressures. To investigate this hypothesis, we have carried out a detailed study of the magnetoresistance for pressures both above and below the gap instability at ~50 kbar, and for temperatures between 1.2 and 40 K. These observations are summarized in Fig. 1, which reports the sign and field dependence of the 7 T magnetoresistance $\rho(7T)-\rho(0T)/\rho(0T)$, at all the temperatures and pressures studied. The pressure-dependent, zero-field activa-

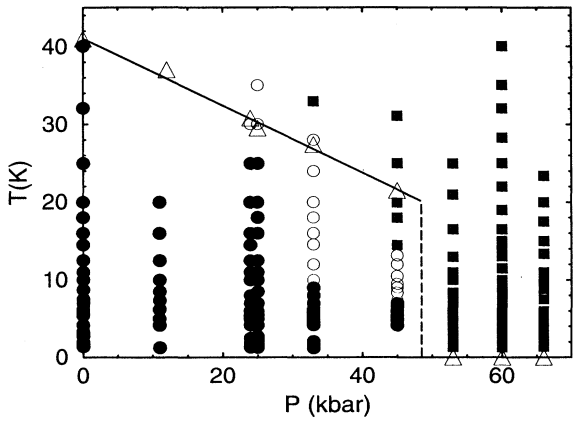

FIG. 1. Sign and field dependence of the electrical resistivity ρ as a function of pressure and temperature. $\Delta\rho(H)/\rho_{H=0} \propto -H^2$ (●), $\Delta\rho(H)/\rho_{H=0} \propto H^{3/2}$ (■), $\Delta\rho(H)/\rho_{H=0}$=admixture of $-H^2$, $H^{3/2}$ (○). Maximum field is 7 T. Also depicted is the pressure-dependent activation gap $\Delta(P)$(△), which vanishes near 50 kbar.

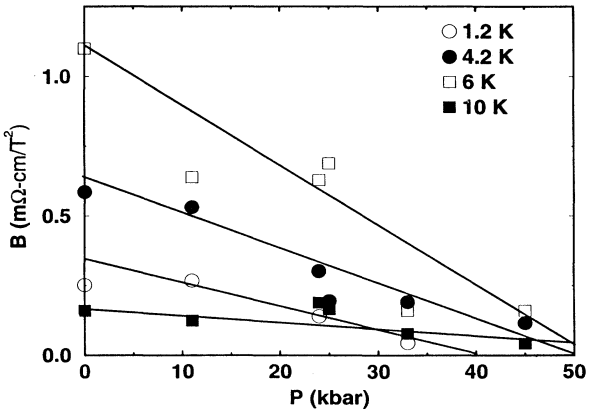

FIG. 3. Pressure dependence of the coefficient $B(P,T)$, for temperatures ranging from 1.2 to 10 K.

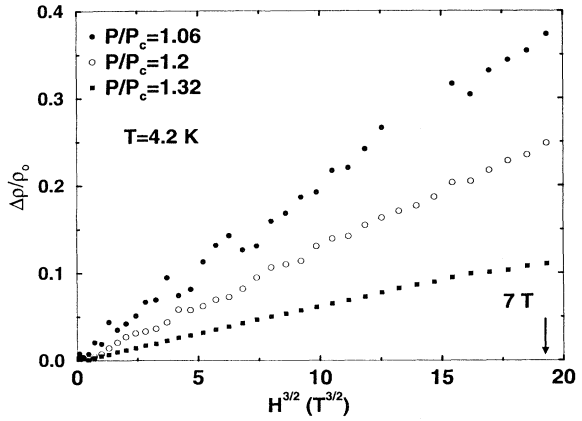

FIG. 4. Pressure dependence of the 4.2 K magnetoresistance in the high-pressure gapless state: $\Delta\rho(H)/\rho_{H=0}=C(P)H^{3/2}$. ●=53 kbar, ○=60 kbar, ■=66 kbar. Critical pressure for gap suppression $P_c\sim 50$ kbar.

tion gap $\Delta(P)$ is plotted for comparison. Roughly speaking, a negative, quadratic magnetoresistance is found as long as the experimental temperature is less than $\Delta(P)$ and a positive magnetoresistance proportional to $H^{3/2}$ is found both at high temperature and for pressures at which Δ has been completely suppressed. The sign change in the magnetoresistance does not herald a sharp phase transition, as complex field dependencies resulting from almost equivalent positive and negative components are found along the interface of the two regions.

Our measurements indicate that the overall magnetoresistance of SmB_6 can be explained very naturally as the result of two pressure-dependent components, one positive and associated with the high-pressure metallic state, and one negative, associated with the low-pressure gapped state. Distinctive field dependencies for the electrical resistivity in SmB_6 can be identified in each regime. The magnetoresistance data plotted in Fig. 2 for $T=4.2$ K and pressures ranging from 1 bar to 33 kbar are representative of the low-temperature, low-pressure gapped regime. In agreement with a previous report in fields as large as 50 T,[14] here the magnetoresistance is negative and quadratic in field $\Delta\rho(H)/\rho_{H=0}=1-B(P)H^2$. The pressure dependence of the coefficient B at fixed temperatures ranging from 1.2 to 10 K is plotted in Fig. 3, demonstrating that at all temperatures this component of the magnetoresistance drops approximately linearly as the pressure is increased from 1 bar to 45 kbar, apparently vanishing as the gap instability at $\sim$50 kbar is approached. At all pressures, $B(P)$ achieves a maximum magnitude between 4.2 and 6 K, perhaps reflecting a crossover in the zero-field transport from the high-temperature regime, dominated by excitations across the gap, to the low-temperature metallic regime.[6]

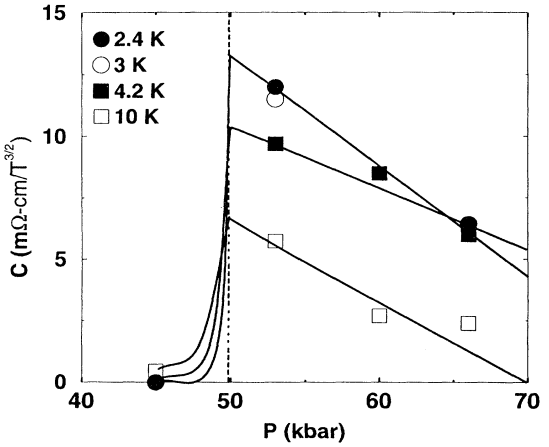

FIG. 5. Pressure dependence of the coefficient $C(P,T)$ for the high-pressure magnetoresistance $\Delta\rho(H=0)/\rho_{H=0}=C(P,T)H^{3/2}$.

Figure 4 shows that the high-pressure gapless regime at 4.2 K is characterized by a positive magnetoresistance, $\Delta\rho(H)/\rho_{H=0}=C(P)H^{3/2}$. Similar results are found at higher temperatures. As demonstrated in Fig. 5, this positive component of the measured magnetoresistance has a pressure dependence complementary to that of the low-pressure state. At all temperatures, $C(P)$ increases sharply as the critical pressure is approached from above, while increasing temperature reduces the overall pressure dependence. Significantly, $C(P)$ is entirely absent in the low-pressure, gapped regime, except at the highest temperatures, indicating that the positive magnetoresistance is entirely a property of the ungapped state in SmB_6.

IV. DISCUSSION

A. The field dependence of the activation gap Δ

We begin our discussion of these experimental results by considering the weak magnetic-field dependence of the activation gap Δ. The magnetic-field dependence of electronic gaps is inevitably traced to Zeeman splitting of the gap-edge states, with a magnitude given by their Landé g factor g:

$$\Delta(H)\sim\Delta_{H=0}\pm g\mu_B H \ . \tag{1}$$

The properties of the gap-edge states can be expected to differ considerably, depending on the microscopic origin of the gap and possible many-body interactions among the states in the conduction and valence bands. In this spirit, the magnetic-field dependence of an electronic gap can give important insight into its origin, particularly in systems in which Δ is suspected to result from electronic correlations. We specialize our discussion here to SmB_6, and consider in detail several scenarios for the gap origin, each leading to distinct field dependencies which can be compared to our experimental results.

The first possibility that we consider is that the gap in SmB_6 is a feature of the single-electron band structure. Self-consistent calculations of the electronic structure of cubic rare-earth hexaborides find that only one band crosses the Fermi level.[15] The valence and conduction

bands correspond to the bonding and antibonding orbitals of the B_6 octahedra, and are completely filled by two electrons. While substantial admixture of extended $5d$ character from the rare earth is expected at the X points, in this view the rare-earth ions serve merely as an electronic reservoir, with their valence setting the band-filling.[16] Single-particle excitations necessarily leave unpaired electrons, which couple to the magnetic field with a g factor given by the local band structure, ~ 2 for nearly free conduction electrons.[17] Narrow band gap InSb provides a classic example of this effect, with an applied magnetic field of 0.8 T increasing the activation gap by more than a factor of 2.[18]

The situation is somewhat similar for the hybridization gap phase found in the mean-field limit of the Kondo lattice model.[19] Here, Δ arises from the coherent hybridization of a lattice of Sm^{3+} moments, partially compensated through their interaction with the conduction electrons.[20] Since Δ is associated with the single-ion Kondo temperature T_K, single-particle excitations correspond to the unbinding of a Sm $4f$ moment–conduction-electron singlet, resulting in a magnetic triplet state.[21] Unlike the simple band gap described above, these gap-edge states in both the valence and conduction bands have substantial local moment character. For SmB_6, the Γ_8 ground state of the $Sm^{3+}(J=5/2)$ ions in a cubic crystal field has $g=2/7$, although the larger g factors of the relatively low-lying excited Γ_7 level as well as those of the admixed conduction-electron states suggest this is a lower bound on the measured g factor.[22,23] These arguments very successfully describe the magnetic-field suppression of the gap in YbB_{12},[24] where a magnetic field of ~ 20 T is found to continuously close the 50 K zero-field gap, corresponding to $g=3.5$, in good agreement with the g factor expected for Yb^{3+} ions in a cubic crystal field.[23] Gap anisotropy in CeNiSn,[25] and a complicated, temperature-dependent magnetoresistance[26] in $Ce_3Bi_4Pt_3$ have hindered similar analyses in these proposed Kondo insulators.

The g factor we deduce from the magnetic-field dependence of the activation gap in SmB_6 is too small to be accommodated by either of these models. Specifically, at 12 kbar $\Delta(H=0)-\Delta(18\text{ T})=1.2\pm0.8$ K, giving a Landé g factor $g=0.1\pm0.06$. At 45 kbar, $\Delta(H=0)-\Delta(18\text{ T})=-2.3\pm0.4$ K, yielding slightly larger $g=0.19\pm0.03$. In both cases, the coupling of the magnetic field to the electronic structure is anomalously weak, and cannot be explained within the context of either the single electron-band structure, or the Kondo hybridization gap models described above.

We hypothesize that the nonmagnetic character of the gap-edge states in SmB_6 results from strong exchange coupling J, which renders them nonmagnetic singlets as in a Mott-Hubbard insulator. J sets the scale for magnetic fields to affect the gap, as minimal Zeeman splitting of the gap-edge states can be expected until the exchange interaction is overcome. Different relationships between J and Δ are expected, depending on the microscopic physics. In a Mott-Hubbard insulator, J and Δ are related through the bandwidth t, $J\sim t^2/\Delta \ll \Delta$. If g is an ionic g factor describing the gap-edge states, a field $g\mu_B H\sim\Delta$ is always sufficient to overcome the exchange coupling in a Mott-Hubbard system with a gap. The field insensitivity of Δ in SmB_6 argues that here the energy scale for exchange is much *larger* than the gap, and confirms that SmB_6 is not a Mott-Hubbard insulator. A more likely scenario has been suggested by Varma,[27] in which the exchange coupling of partially compensated Sm^{3+} moments can be traced to the Ruderman-Kittel-Kasuya-Yosida interaction, which must nevertheless be too weak to drive long-range antiferromagnetic order in SmB_6. We point out that the presence of exchange interactions would distinguish SmB_6 from Kondo insulators like CeNiSn, which inelastic neutron-scattering experiments[28] find has $J\to 0$ as $\omega\to 0$.

B. Origin of the magnetoresistance

Since the temperature dependence of the electrical resistivity remains activated, even in large magnetic fields, there are two possible sources of the magnetoresistance in the gapped state:[29]

$$\rho(T)=\rho_0\exp[\Delta(P,H)/k_BT]\ , \tag{2}$$

$$\frac{d\rho}{dH}=\rho\frac{d\rho_0}{dH}+\Delta\rho\frac{d\Delta}{dH}\ . \tag{3}$$

At 4.2 K, 87% of the measured magnetoresistance at 12 kbar is accounted for by the second term, while at 45 kbar this fraction has dropped to 77%. Regardless of the gap origin, the field suppression of Δ results in the observed negative magnetoresistance, which must necessarily vanish at the critical pressure. Ignoring the field dependence of ρ_0, it is possible to invert (2) and experimentally deduce the magnetic-field dependence of Δ. As demonstrated in Fig. 6, $\Delta(H)$ is reduced quadratically by field. Δ becomes progressively less sensitive to magnetic fields as the critical pressure for gap collapse is approached, providing a natural explanation for the disap-

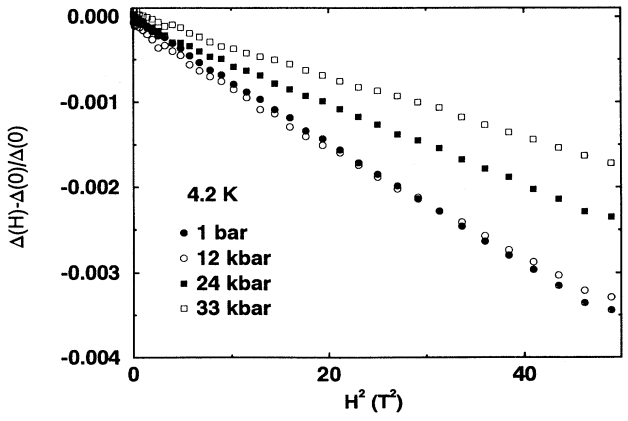

FIG. 6. The magnetic-field dependence of the activation gap Δ deduced from the 4.2 K magnetoresistance using Eq. (2) at 1 bar (●), 12 kbar (○), 24 kbar (■), and 33 kbar (□).

pearance at P_c of the coefficient of the negative magnetoresistance depicted in Fig. 3.

As the pressure is increased towards the gap instability, the magnetoresistance—like the zero-field transport—is increasingly dominated by states which pin the Fermi level in the gap. Equations (2) and (3) are no longer adequate to describe the low-temperature transport, either in a field or not. The measured field dependence of the electrical resistivity $\Delta\rho(H)/\rho_{H=0} = C(T,P)H^{3/2}$ provides us insight into the Fermi surface describing these metallic gap states, a conclusion strengthened by the complete absence of this component of the magnetoresistance in the low-pressure, gapped state. Significantly, the simple uncompensated metal LaB_6 has a very similar magnetoresistance $\Delta\rho(H)/\rho_{H=0} \propto H^{1.4}$ for $H\|100\perp I$, and de Haas–van Alphen measurements confirm the presence of an open orbit in this direction on the Fermi surface.[30] We hypothesize that the magnetoresistance in high-pressure SmB_6 results from a similar Fermi-surface orbit. Since both the electrical resistivity and carrier concentration vary considerably with both temperature and pressure in the high-pressure gapless regime of SmB_6, it is difficult to compare orbit parameters in SmB_6 and LaB_6 using the raw magnetoresistance data presented in Fig. 4. Instead, we use Kohler's rule[31]

$$\Delta\rho(H)/\rho_{H=0} = \mathcal{F}(\omega_c\tau) , \tag{4}$$

$$\omega_c = eH/m^*c, \quad \tau = m^*\rho_{H=0}/ne^2 , \tag{5}$$

to scale the 4.2 K magnetoresistance by the pressure-dependent carrier concentration n and zero-field resistivity $\rho_{H=0}$.[6] The magnetoresistance of SmB_6 measured in fields up to 7 T is presented as a function of $\omega_c\tau = H/\rho_{H=0}ne$ in Fig. 7, for pressures from 53 to 66 kbar. Similar data for LaB_6 are shown for comparison.[30] While $\mathcal{F}(\omega_c\tau) \propto (\omega_c\tau)^{3/2}$ in each case, suggesting a common Fermi-surface orbit, it is clear that the orbital parameters themselves vary considerably with pressure in SmB_6. Since a larger $\omega_c\tau$ is required to provide an equivalent magnetoresistance as pressure is increased, we deduce that the Fermi-surface orbit—if not the Fermi surface itself—expands as pressure is increased above the critical pressure. Assuming that the origin of this expansion is the pressure-induced addition of electrons previously localized in Sm orbitals, we predict that the orbit must expand still more and approach the dimensions found in trivalent, isostructural LaB_6 as pressure increases the Sm valence towards 3.[32]

V. CONCLUSION

In summary, neither the single-electron band structure or Kondo insulator models successfully describe the weak magnetic-field dependence of the activation gap we have found in SmB_6. By analogy to Mott-Hubbard insulators, we suggest that the gap-edge states experience exchange coupling, with an energy scale for exchange much larger than the correlation gap itself. Nonetheless, the negative magnetoresistance found in the low-pressure gapped state can be almost entirely attributed to the weak-field dependence of the gap. At higher pressures, the pressure-induced collapse of the gap is accompanied by a sign change in the magnetoresistance. The field dependence of the resistivity above the critical pressure ~50 kbar is very similar to that of LaB_6, indicating carriers in a similar Fermi-surface orbit. Pressure is found to increase the orbital radius, and we conclude that the carriers are being transferred from localized Sm levels to the Fermi level, commensurate with the valence increase found over the same pressure range. Our results indicate that at the highest pressures, the electronic properties of SmB_6 will closely resemble those of LaB_6, with the possible distinction of weak Kondo coupling to the Sm^{3+} moments.

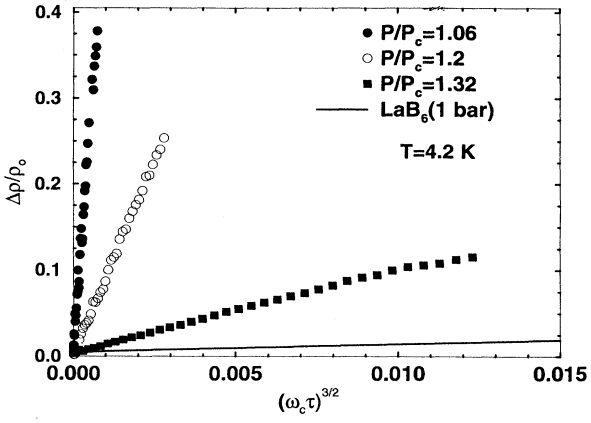

FIG. 7. 4.2 K magnetoresistance from Fig. 5 plotted as a function of $\omega_c\tau = H/ne\rho_{H=0}$. Maximum field is 7 T. LaB_6 data are taken from Arko *et al.* (Ref. 30).

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