Pressure-induced Fermi-liquid behavior in the Kondo insulator SmB_6: Possible transition through a quantum critical point

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We have investigated the electrical resistivity of the intermediate valence narrow-gap semiconductor SmB_6 at temperatures below 80 K and under pressure in the range between 1 bar and 70 kbar. We report on a continuous suppression of the gap under pressure, and on the observation of a critical point at $p_{cr} \approx 40$ kbar, characterizing the pressure induced transition from a Kondo insulator below p_{cr} to a metallic heavy fermion liquid well above p_{cr}. In the metallic phase close to p_{cr} strong indications for a non-Fermi-liquid region, in which the electrical resistivity can be described by a power law $\rho(T) = \rho_0 + AT^n$ with $1 \leqslant n \leqslant 2$, were observed.

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Strongly correlated, low carrier density systems exhibiting a small transport and spin gap close to the Fermi level, have attracted much attention in the last few years.[1–3] The formation of this small gap is expected to originate from the hybridization of the narrow and well localized f states with the broad s-, p-, or d-type conduction band, with each unit cell containing an even number of electrons. The intermediate valence compound SmB_6, in which the oxidation state of samarium ions is intermediate between the Sm^{2+} ($4f^6$) and Sm^{3+} ($4f^5 5d^1$) configurations, is one of the most studied materials of this class of heavy fermion semiconductors, also denoted as Kondo insulators. Its magnetic susceptibility reveals a Curie-Weiss-like dependence for $T > 100$ K indicative of local-moment behavior, and a Pauli-like spin susceptibility for the lowest temperatures due to a non-magnetic configuration for $T \rightarrow 0$.

Recent experiments on SmB_6 (Refs. 4–11) showed that in the low-energy excitation spectrum of this material several energy scales and several regimes of low-temperature electron kinetics exist. The properties of SmB_6 are governed by the hybridization gap $E_g \approx 10$–20 meV in the temperature range $70\ \text{K} > T > 15$ K; between 15 and 5 K a narrow in-gap band separated from the bottom of the conduction band by a direct activation energy of $E_d \approx 3$–5 meV was observed. Below about 5 K the electrical conductivity saturates, indicating a small conductivity channel within the E_d in-gap states, where the Fermi level is pinned. While Ref. 10 favors hopping processes of electrons, results of Refs. 11,12 imply the residual conductivity of SmB_6 being nonactivated and the corresponding state, which is formed within the in-gap states, having a type of metallic nature. An enhancement of the specific heat of SmB_6 below about 2 K has been attributed to the formation of a coherent state within this metallic-like state.[13] Moreover, a comprehensive study of low temperature characteristics, thermopower, microwave conductivity, and millimeter wavelength range quasioptical spectra of high quality single crystals of SmB_6 have allowed the authors of Refs. 10,11 to choose among a variety of models of fast charge fluctuations in favor of a formation of exciton-polaron complexes[14,15] in the temperature range $T < 15$ K. In addition, very recent spectroscopic measurements[16–18] indicate that the above described insulating gap at E_F rather has a pseudogap structure.

Variations in pressure, composition, and magnetic fields are expected to alter the magnitude or the energy spectrum of the gap (pseudogap) structure due to the mutual shift of the $4f$ states and the $5d$ band, thus giving rise to an insulator to metal transition in SmB_6. Resistivity studies of Berman *et al.*[19] and Beille *et al.*[20] showed that the behavior of $R(T)$ changes continuously from that of a narrow gap semiconductor to that of a metal in the range of pressure between 0 and 70 kbar. Results of Cooley *et al.*,[6] on the other hand, have revealed the activation gap in SmB_6 vanishing discontinuously between 45 and 53 kbar, yielding at high pressures a mass-enhanced Fermi liquid phase. Thus, in spite of a lot of effort, the nature of the crossover from semiconducting into metallic state, which also provides evidence whether the hybridization gap description is complete or not, remains still unclear. Properties of this compound close to the pressure induced metal-insulator transition have not been investigated so far either.

In order to shed more light onto this problem single crystal resistivity measurements under pressure in the range between 1 bar and 70 kbar were performed. These pressures encompass the entire insulating phase and a part of the high pressure and low-temperature metallic phase. We report on the observation of a continuos suppression of the energy gap in SmB_6, and characterize the metallic phase lying beyond the critical pressure p_{cr}. On this metallic side of the phase boundary, we identify two regimes with different properties: (i) a region close to p_{cr}, in which the resistivity exhibits non-Fermi-liquid phenomena and (ii) a high pressure (heavy) Fermi-liquid state.

Single crystals of SmB_6 were prepared using the floating zone melting technique, their ratio between resistivity at 4.2 K and room temperature was higher than 10^4. Pressure measurements up to 19 kbar were carried out in a piston-cylinder cell, $ErCo_2$ with a first order phase transition served as manometer. High pressures were generated in a Bridgman anvil cell using Al_2O_3 as anvils. In this case the pressure was

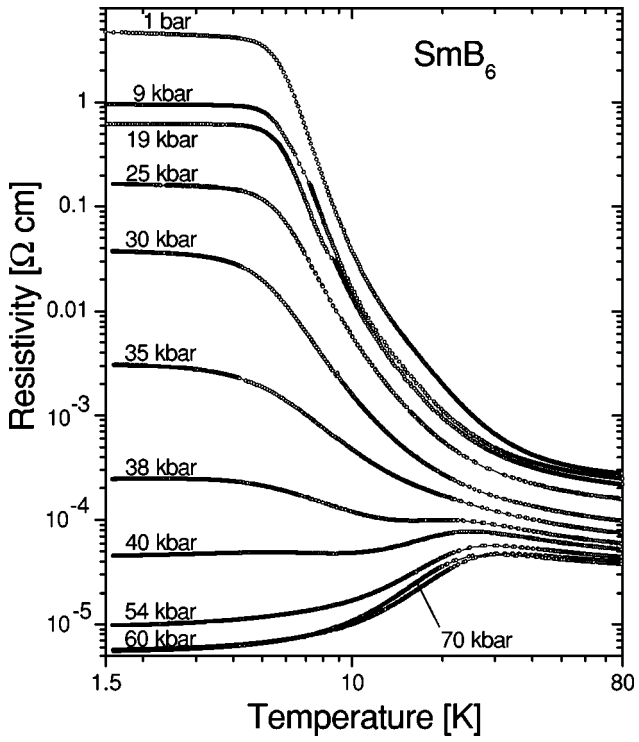

FIG. 1. Temperature dependent electrical resistivity $\rho(T)$ for various values of applied pressure. The insulator to metal transition sets in at $p_{cr} \approx 40$ kbar.

monitored *in situ* by a superconducting Pb manometer. The resistivity was measured in four probe arrangement, with current kept low enough to avoid self-heating of the sample.

The received resistivity data as a function of temperature, for various pressure values, are plotted in Fig. 1. The observed temperature dependence of the resistivity below 40 kbar exhibits an increase as the temperature is lowered from 70 to 3 K, indicating a gap structure in the density of states. Below 3 K a saturation is observed. To derive more information about the influence of pressure on the gap structure and about the carrier kinetics below 70 K, the local activation energy $W(T) = d[\ln \rho(T)]/d(1/k_B T)$ [derivative of the $\ln \rho(T)$ vs $1/k_B T$ dependence] as a function of temperature was determined from the above data by means of a standard numerical derivative analysis, see Fig. 2. At ambient pressure and in the temperature range between 70 and 15 K the resistivity dependence is characterized by an activation energy with a value of about 5.9 meV. This value can be attributed to intrinsic semiconductorlike conductivity of the form $\rho(T) \propto \exp(E_g/2k_B T)$, where $E_g \approx 11.8$ meV corresponds to the width of the hybridization gap. Between 15 and 3 K a second pronounced activation energy appears. Its value is $E_d \approx 5.6$ meV, and represents the distance between the bottom of the conduction band and the in-gap states that are formed within this energy gap. Below about 3 K the local activation energy decreases rapidly to values lower than the available thermal energy $k_B T$ implying that the residual conductivity of SmB_6 is of nonactivated (metalliclike) nature. The fact that E_g and E_d do not clearly level off is most probably connected with the pseudogap structure of the energy gap. With increasing pressure (see Fig. 2) the local activation energy maxima become lower, the structures are broadened and shifted to lower temperatures, reflecting a gradual suppression of the gap (pseudogap) width in the density of states. For pressures of 38 kbar and 40 kbar the activation energy forms only one broad maximum, thus making it difficult to distinguish between E_g and E_d. In this case we consider these activation energies to have merged, i.e., $E_g/2 = E_d$. Since for $p = 40$ kbar E_g and E_d are very close to zero (they seem to disappear together), we estimate the critical pressure p_{cr}, where the semiconductorlike conductivity and the local activation energies disappear, to be about 40 kbar. The inferred (E_g, E_d) vs p phase diagram is shown in Fig. 3, a continuous suppression of the gap in SmB_6 with increasing pressure can be observed. Equivalent E_g and E_d pressure dependencies can be obtained from the slopes of an Ahrenius plot [i.e., from the $\ln \rho(T)$ vs $1/k_B T$ plot].

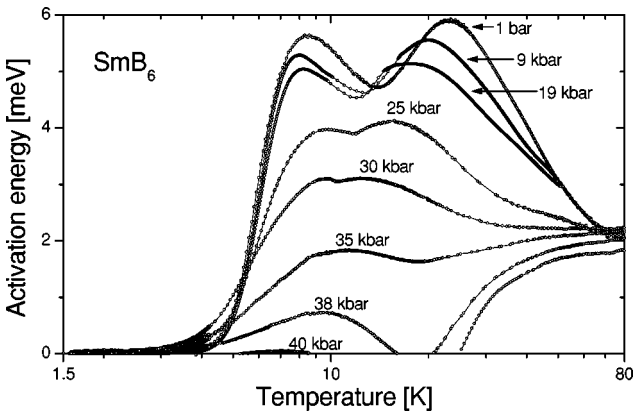

FIG. 2. Temperature dependence of the local activation energy W of SmB_6, calculated for various pressures from the resistivity data in Fig. 1.

Particularly interesting are the results of the analysis of the low temperature and pressure induced metallic phase. Here, at the lowest temperatures and for $p \geq p_{cr}$ resistivity does not follow a characteristic Fermi liquid behavior $\rho(T) = \rho_0 + AT^2$, with ρ_0 being the residual resistivity. Instead, it appears to be well described by a power law $\rho(T) = \rho_0 + AT^n$ and $1 \leq n \leq 2$. For pressure $p = 40$ kbar $n \approx 1$, for $p = 54$ kbar $n \approx 1.56$, for $p = 60$ kbar $n \approx 1.9$ and for $p = 70$ kbar $n \approx 2$ (Fig. 4). Although the temperature range in which these dependencies are observed is relatively narrow, the manifestation of this kind of dependence at all successive pressures between 40 and 70 kbar shows strong indications for a non-Fermi-liquid behavior.

For $p = 60$ kbar and $p = 70$ kbar, where the exponent n is very close or equal to 2, the coefficient A appears to have a magnitude of 0.052 and 0.044 $\mu\Omega\,\mathrm{cm\,K^{-2}}$, respectively. These values are lower than those deduced by Cooley *et al.*[6] According to the Kadowaki and Woods plot[21] the corresponding electronic specific heat coefficient γ should have a value of about 60 $\mathrm{mJ\,mol^{-1}\,K^{-2}}$, which shows that high pressure metallic SmB_6 exhibits properties similar to that of a normal Kondo lattice: A coherent Fermi liquid behavior with a moderate enhancement of the quasiparticle mass at low temperatures and the $\rho(T) \propto -\ln(T)$ resistivity dependence at higher temperatures (between 50 and 300 K). It is

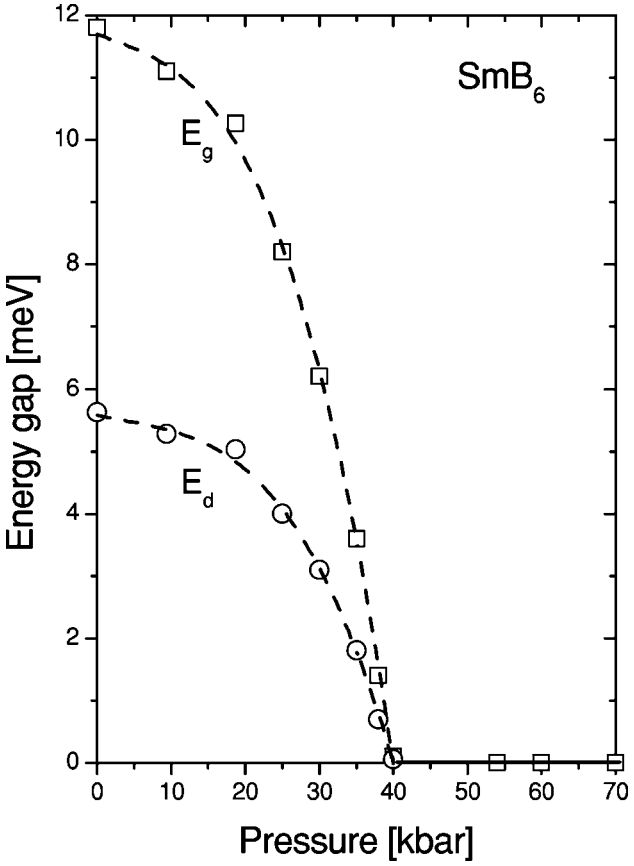

FIG. 3. Pressure dependence of the activation energies E_g (related to the width of the hybridization gap) and E_d (related to the excitations between the in-gap states and the bottom of the conduction band). The dashed lines are guides for the eye.

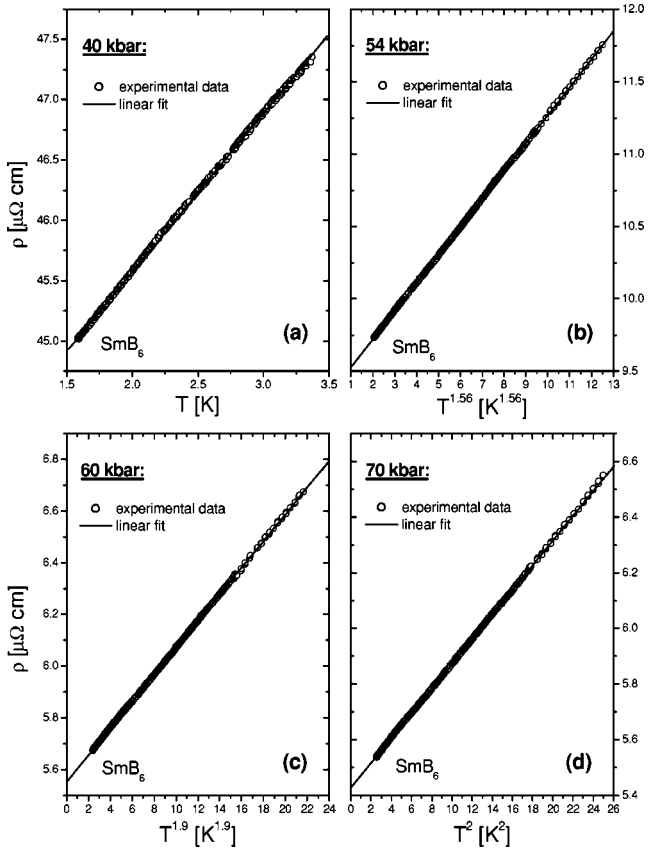

FIG. 4. Temperature dependent resistivity at (a) $p=40$ kbar, (b) $p=54$ kbar, (c) $p=60$ kbar, and (d) $p=70$ kbar. The dashed lines are least squares fits (see text).

also interesting to note that the derived γ value is comparable to the enhanced electronic specific heat coefficient observed in heat capacity measurements at very low temperatures.[13] Moreover, despite of limited data, it can be seen that the characteristic energy scale for quasiparticle interactions in the Fermi liquid $T^* \propto 1/\sqrt{A}$ increases, and the mass enhancement $m^*/m_e \propto \sqrt{A}$ correspondingly decreases as p is driven beyond 60 kbar.

The above behavior of $\rho(T)$ near the critical point p_{cr} reminds of that observed e.g., in antiferromagnetic *f*-electron systems such as $CePd_2Si_2$ (Ref. 22) and of that in the Mott insulator $BaVS_3$,[23] a compound that is dominated by antiferromagnetic spin-spin interactions at lowest temperatures.

It should be noted that non-Fermi-liquid behavior is often found experimentally near a magnetically ordered (ferromagnetic or antiferromagnetic) state in the phase diagram, indicating that non-Fermi-liquid behavior in those systems may be linked to a magnetic instability that arises at $T=0$. The transition from a magnetically ordered to a non-Fermi-liquid state is driven by a control parameter other than temperature, e.g., external pressure, doping or magnetic field at absolute zero, by quantum-mechanical fluctuations. The control parameter thus tunes a system at the zero temperature boundary from an ordered ground state towards a nonordered state crossing a quantum critical point (see, e.g., Refs. 24,25). In the case of SmB_6 the overall appearance of the susceptibility curve[2,9] shows that the ground state is formed by the nonmagnetic $^7F_0$ state of Sm^{2+} ($4f^6$) and by the nonmagnetic $^7H_2$ state in which the $5d^1$ electrons are coupled to the $4f^5$ electrons of the Sm^{3+} configuration (only with increasing temperature electrons are transferred from ground state to excited magnetic energy levels). Thus, in this compound, under the influence of external pressure at $T \rightarrow 0$, a transition from an insulating to a Fermi-liquid behavior, characterized by an enhanced electronic density of states at E_F and also most probably by enhanced Pauli paramagnetism, is observed. The non-Fermi-liquid-like behavior in between both of these regimes would then be a consequence of a pressure driven crossover from a non-magnetic to a magnetic state. This behavior, together with the dependence of $E_g(p)$, if the width of the energy gap is taken as an order parameter, resembles very much the case of a pressure induced insulator (semiconductor) to metal transition through a quantum critical point as it was observed in $BaVS_3$.[23] To prove the presence of a quantum critical point in SmB_6, however, further pressure experiments (above all at lower temperatures) are needed. Moreover, one should keep in mind that deviations from the T^2 behavior at p_{cr} may originate from residues of the former gap or pseudogap structure in the density of states as well. To resolve a T^2 dependence in $\rho(T)$ requires an energy independent density of states at the Fermi level which, however, seems not to be found at the critical pressure.

The inferred continuous vanishing of the energy gap of SmB_6 with pressure seems to support the hybridization gap scenario of this Kondo insulator. Moreover, the almost parallel continuous decrease of the activation energy E_d with pressure suggests the corresponding in-gap states being very tightly connected with the hybridization gap and thus seem to exhibit intrinsic nature. Their nonexternal nature (even if impurities and imperfections can slightly alter their position in the gap[12]) can also be derived from the fact that up to now these states have been observed in all investigated samples, regardless of their quality, way and place of their production. Our results thus seem to support the model recently proposed by Curnoe and Kikoin[15] in which self-trapped electron-polaron complexes rather than simply electrons in an impurity band are responsible for the low temperature properties of SmB_6. However, also other models which include the presence of in-gap states cannot be ruled out at present.

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