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Suppression of the energy gap in SmB_6 under pressure

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The electrical resistance R of SmB_6 as a function of temperature T and pressure P has been measured in the range $1 \text{ K} \leq T \leq 300 \text{ K}$ and $0 \leq P \leq 220 \text{ kbar}$. The behavior of $R(T)$ changes continuously from that of a narrow gap semiconductor to that of a metal in the range of $0 \leq P \leq 70 \text{ kbar}$. The dependence of R on T and P can be analyzed phenomenologically within the context of a thermal activation model with an activation energy that decreases linearly with pressure from $\sim 33 \text{ K}$ at zero pressure to zero at $\sim 70 \text{ kbar}$. The data resemble those of SmS and SmSe under pressure and suggest a general behavior of $R(T, P)$ for intermediate-valence Sm compounds.

The intermediate-valence (IV) compound SmB_6 has attracted the attention of both experimentalists and theoreticians alike because of its striking physical properties, some of which are indicative of a poor metal, and others which are characteristic of a semiconductor with a small energy gap of several meV.¹⁻³ A number of theoretical models⁴⁻¹⁰ have been advanced to account for the unusual physical properties of SmB_6 such as the $d-f$ hybridization gap model proposed by Mott,⁶ and the disordered Wigner lattice model of Kasuya *et al.*⁹

The first evidence for semiconducting behavior of SmB_6 was provided by the temperature dependence of the electrical resistivity. The resistivity increases with decreasing temperature in a thermally activated manner and then, below 3 K, saturates to a value that can be as large as 10^4 times the room-temperature value.^{1,11-13} NMR,¹⁴ electron tunneling,^{1,13,15-17} far-infrared absorption,^{1,13} and low-temperature specific-heat¹ measurements are all consistent with the existence of a small energy gap of several meV.

Recently, x-ray diffraction measurements on SmB_6 under pressure were carried out by King, LaPlaca, Penney, and Fisk¹⁸ in a diamond anvil cell at room temperature. The results indicate that the valence of the Sm ions changes from 2.8 at zero pressure to 2.9 at 60 kbar, the highest pressure attained in the experiment. A transition to a fully trivalent state at higher pressure could conceivably occur and should lead to magnetic order since trivalent Sm is a Kramer's ion.

In this paper, we report the results of measurements of the pressure dependence of $R(T)$ of SmB_6 up to $\sim 220 \text{ kbar}$. The experiment was undertaken in order to (1) determine how the energy gap varies with pressure, (2) search for evidence of phase transitions (e.g., crystallographic, valence, insulator-metal, magnetic) under pressure, and (3) compare the pressure dependences of $R(T)$ of SmB_6 and

the samarium monochalcogenides SmS (Refs. 19-21) and SmSe (Ref. 21) in their IV phases.

Four separately grown samples of SmB_6 were investigated at high pressures— samples 1 and 2 at the University of California at San Diego (UCSD) and samples 3 and 4 at Kernforschungsanlage (KFA), Jülich. All samples were in the form of coarse powders obtained by crushing small single crystals of SmB_6 which were formed in an Al flux.

A Bridgman anvil technique²² was employed in attaining quasihydrostatic pressures $P \leq 160 \text{ kbar}$ at UCSD and $P \leq 220 \text{ kbar}$ at KFA, Jülich. The SmB_6 powder and a Pb manometer were sandwiched between two steatite disks and contained within a pyrophyllite gasket, although in the case of sample 1, the Pb manometer was omitted. Pressures relevant to sample 1 were estimated from the applied pressure load during pressurization using a previously established calibration which was based on the T_c vs P behavior of Pb. A Pb manometer was included in the pressure cell in the experiments on sample 2. The higher-pressure measurements at KFA, Jülich were also calibrated via Pb manometers assuming a linear relationship for T_c (Pb) vs P between the Pb (I-II) transformation fixed point at 130 kbar and the GaP transformation fixed point at 220 kbar.

Shown in Fig. 1 are R vs T data between 1 and 300 K and pressures $P \leq 145 \text{ kbar}$ for sample 1. The temperature dependence of R at the lowest pressure (18 kbar— curve A) is similar to that previously observed at zero pressure¹¹; specifically, R increases with decreasing T , rapidly for $3 \leq T \leq 50 \text{ K}$, and more slowly for $T < 3 \text{ K}$, while

$$R(1 \text{ K})/R(300 \text{ K}) \approx 500.$$

With increasing pressure, the $R(T)$ curves gradually change shape, passing through complex variations at intermediate pressures to metallic character at the highest pressures. At the highest pressure (145 kbar— curve G), R decreases

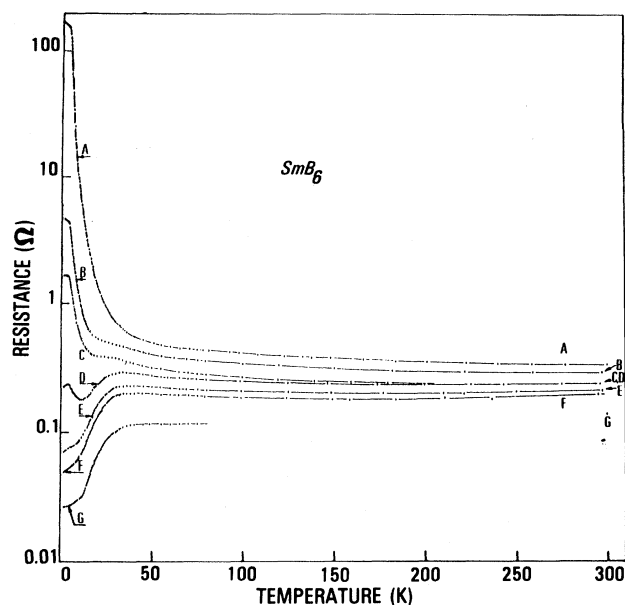


FIG. 1. Electrical resistance vs temperature for SmB_6 at various pressures (A—18 kbar, B—33 kbar, C—47 kbar, D—62 kbar, E—75 kbar, F—89 kbar, and G—145 kbar).

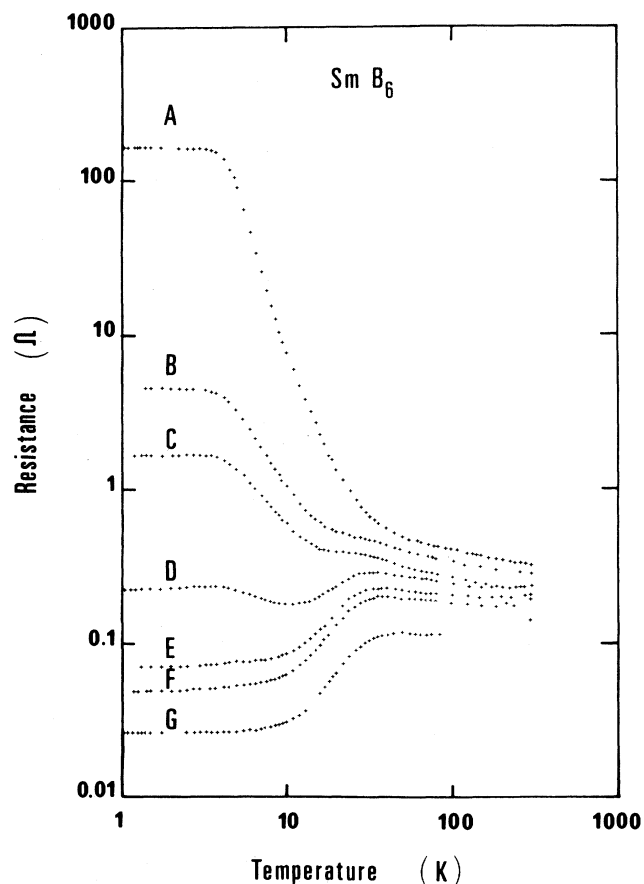


FIG. 2. Electrical resistance (R) vs temperature (T) data of Fig. 1 plotted on logarithmic R and T scales.

with decreasing temperature and

$$R(1\text{ K})/R(300\text{ K}) \approx 0.2.$$

The data of Fig. 1 are also displayed as $\log R$ vs $\log T$ and $\log R$ vs T^{-1} in Figs. 2 and 3, respectively.

A second set of experiments on sample 2 revealed the same general behavior displayed by curve E of Fig. 1 at the same applied pressure, although the low-pressure value $R(1\text{ K})/R(300\text{ K}) \approx 10$ indicated that sample 2 was not as pure as sample 1. Samples 3 and 4 were measured up to pressures of ~ 190 kbar and ~ 216 kbar, respectively. The $R(T)$ curves for both samples showed the same evolution with pressure from semiconducting to metallic character exhibited by samples 1 and 2.

The $R(T)$ data of sample 4 are displayed in Fig. 4. Between 125 and 216 kbar, the transition towards more metallic behavior with pressure is apparent from the disappearance of the maximum in $R(T)$ above 125 kbar and the relative steepening of the $R(T)$ curves for $50 \leq T \leq 300$ K. The $R(T)$ data depicted by curve A were taken at 21 kbar before the excursion up to 216 kbar, while the $R(T)$ data represented by curve B were taken at 21 kbar upon reloading following a complete release of pressure. The disparity between curves A and B reflects the permanent damage in-

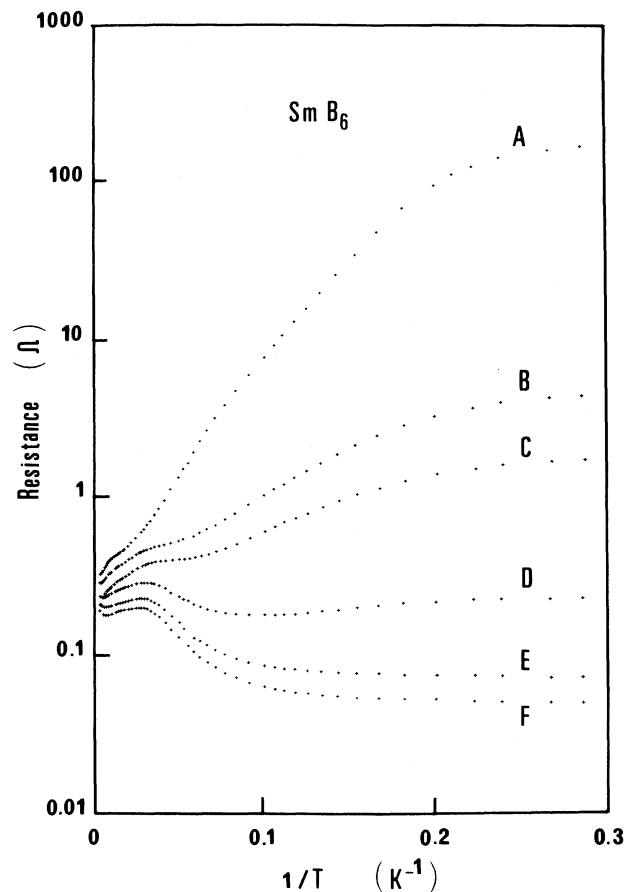


FIG. 3. Electrical resistance (R) vs inverse temperature (T^{-1}) data of Fig. 1 plotted on logarithmic R and T^{-1} scales (Arrhenius plots).

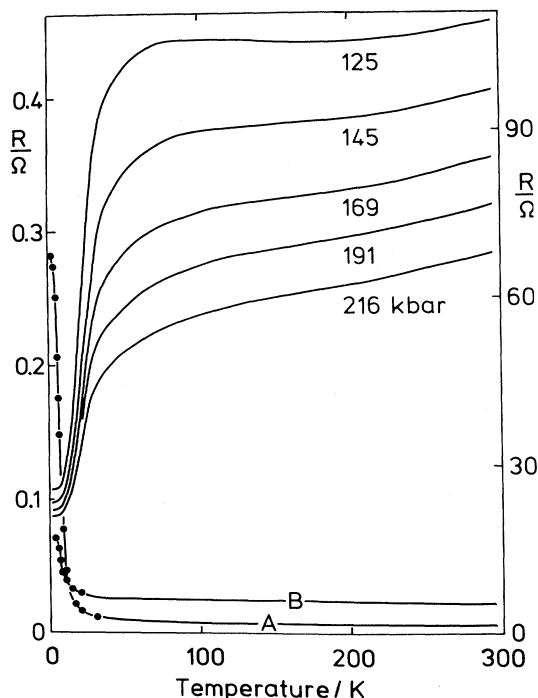


FIG. 4. Electrical resistance R vs temperature isobars for SmB_6 at various pressures between 125 and 216 kbar (left scale) and at 21 kbar (right scale—curves A and B). Curve A represents data taken before excursion to 216 kbar, while curve B denotes data taken upon reloading following a complete release of pressure.

curred by the SmB_6 sample after it had been subjected to an inhomogeneous pressure of 216 kbar.

The $\log R$ vs T^{-1} data (Arrhenius plots) at the lower pressures shown in Fig. 3 are consistent with conduction by thermal activation of electrons across an energy gap. Therefore we have analyzed the data *phenomenologically* with an activation law $R = R_0 \exp(\Delta E/kT)$, where ΔE is the activation energy. This equation describes the 18-kbar data (curve A of Fig. 3) with $\Delta E \approx 27$ K in the temperature ranges 6–14 and 140–300 K. It is interesting to note that thermally activated behavior of $R(T)$ with the same activation energy ΔE below ~ 20 and above ~ 50 K has been observed in single-crystal specimens of SmB_6 at ambient pressure with values of ΔE between 28 and 41 K.¹³ At higher pressure, ΔE for the low-temperature linear part of the Arrhenius plot decreases rapidly with pressure and vanishes at ~ 70 kbar, as can be seen in Fig. 5 where ΔE is plotted versus P . Within experimental uncertainty, ΔE decreases linearly with P at a rate $d(\Delta E)/dP \approx -0.5$ K/kbar. Extrapolation of the data to $P=0$ yields $\Delta E = 33 \pm 5$ K. This value is in reasonable agreement with previously reported zero-pressure values of ΔE that range from 21 to 41 K.^{5,11–13} Although ΔE has been deduced in a temperature range where $kT \approx \Delta E$, the energy gap of SmB_6 is expected to be comparable in magnitude to ΔE and to close with pressure at about the same rate.

In the pressure range investigated here, the pressure dependence of $R(T)$ of SmB_6 bears a striking resemblance to that of SmS in its high-pressure (≥ 6.5 kbar) IV “gold” phase and SmSe . Whereas the character of the $R(T)$

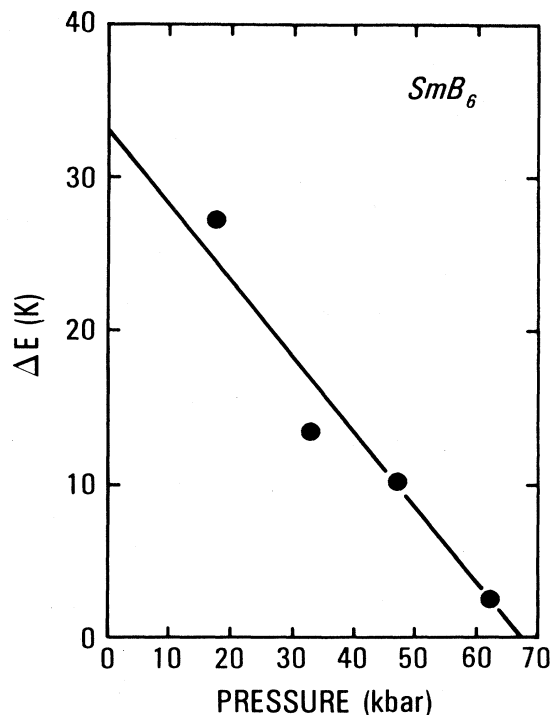


FIG. 5. Low-temperature activation energy ΔE vs pressure for SmB_6 .

curves changes from semiconducting to metallic near ~ 70 kbar for SmB_6 , it occurs near ~ 20 kbar for SmS (Refs. 19–21) and ~ 100 kbar for SmSe .²¹ Moreover, the overall shapes of the $R(T)$ curves of SmB_6 above ~ 70 kbar, gold SmS above ~ 20 kbar, and SmSe above ~ 100 kbar are surprisingly similar to one another and to those of many metallic IV compounds whose R vs T curves exhibit strong negative curvature and saturation in the neighborhood of or below room temperature.³ An interesting feature in the $R(T)$ curve of SmB_6 is the maximum that disappears above ~ 125 kbar. A corresponding maximum in the $R(T)$ data of SmS vanishes completely by ~ 108 kbar.²¹

Recent electron tunneling experiments have revealed the appearance of a small energy gap ~ 1.7 meV at 4.2 K for SmS above the pressure at which the “black” phase transforms into the gold phase.¹⁷ Thus the energy gap for gold SmS is comparable to the energy gap of SmB_6 .

The fact that SmB_6 , SmS , and SmSe all display similar pressure-induced transitions from a narrow gap semiconductor to a metal suggests that this may be a general behavior of IV Sm compounds with an underlying common mechanism that remains to be elucidated. Finally, the IV compound TmSe has also been found to exhibit a small energy gap ~ 2 –3 meV as well as an insulator-metal transition near 32 kbar, although this case is complicated by the occurrence of several types of magnetic order.²³

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