Pressure and magnetic field effect on transport gap of black SmS

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Abstract. We measured the electrical resistivity of black SmS at high magnetic fields up to 30 T. We observed that the magnetic field drastically reduces the energy gap \( E_g(H, P) \) in a similar way to that the pressure does. From the finding that data points of \( E_g(H, P) \) lie on a single line as functions of \( H \) and \( P \), we argue that the giant negative magnetoresistance is intrinsic to black SmS and the magnetic field plays a similar role to the pressure.

1. Introduction
Pressure effect on strongly correlated 4f-electron systems is one of the most attractive topics in solid state physics. SmS is a prototypical system that exhibits a pressure-induced phase transition at a critical pressure \( P_{c1} \) (\( \sim 6 \) kbar at room temperature) from a semiconducting “black phase” to a valence fluctuating “golden phase” accompanied by an isostructural volume collapse of \( \Delta V/V \sim 0.08 \) [1]. At higher pressures than about 20 kbar, the electrical resistivity shows metallic behavior at low temperatures [2]. Recently, a second critical pressure \( P_{c2} \) (\( \sim 20 \) kbar) above which an antiferromagnetic ordered state emerges was detected by nuclear forward scattering, magnetic susceptibility, heat capacity, thermal expansion, and Hall effect experiments [3 - 7]. It is interesting to note that an intermediate state between \( P_{c1} \) and \( P_{c2} \) is a nonmagnetic, pseudogapped state, as evidenced by thermal expansion and specific heat experiments [8]. On the other hand, black phase seems not to have attracted so much interest, possibly because problems concerning black phase are considered to be settled; the temperature dependence of the electrical resistivity can be understood as a conventional narrow-gap semiconductor, and the magnetic susceptibility can be explained by Van Vleck paramagnetism of Sm\(^{2+} \) ionic state (4f\(^6 \), \( J = 0 \)) [9]. From the theoretical study of the electronic structure on black SmS, the semiconducting energy gap \( E_{gap} \) which is determined by the energy difference between the localized 4f level and the 5d conduction band opens and this energy gap shows pressure dependence with the rate of \( dE_{gap}/dp \sim -130 \) K/kbar which is in good agreement with experiments [10, 11, 12]. Interestingly, a strong magnetic field effect on the electrical resistivity was observed by Konczykowski et al.; at ambient pressure the large negative magnetoresistance was observed below 280 K [13]. They attributed the observation to an impurity effect, because it is difficult to explain the huge magnetic field effect on the electrical resistivity based on a simple nonmagnetic Sm\(^{2+} \)

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state. In our previous report [12], we showed that the giant negative magnetoresistance arises from the energy gap reduction by a magnetic field. In the present paper, we argue that the magnetoresistance can be intrinsic to black SmS.

2. Experimental technique
A single crystal of SmS was grown by the Bridgman method. Note that the electrical resistivity strongly depends on stoichiometry [11]. A sample used for the present measurement (with a dimension of $2.0 \times 2.1 \times 0.36 \text{ mm}^3$) was cleaved from a bulk ingot having a nominal composition of Sm$_{1.00}$S$_{1.00}$ (sample #8), and is different from a sample (Sm$_{1.01}$S$_{1.00}$, sample #10) that was used for our previous investigation [12]. The transverse magnetoresistance measurements under pressure were carried out by usual DC van der Pauw method. The magnetic field applied perpendicular to the current was generated by a superconducting magnet up to 15 T and a hybrid magnet up to 30 T installed at National Institute for Materials Science. Pressure was applied at room temperature using a piston cylinder clamp cell, which consists of a NiCrAl inner and CuBe outer cylinder. Daphne oil 7373 was used as a pressure transmitting medium. Pressure at low temperatures was determined from a superconducting transition temperature of lead.

3. Results and Discussion
Figure 1 shows the temperature dependence of the electrical resistivity under magnetic fields at 1.4 kbar in Arrhenius plot ($\rho \propto \exp(E_g/2k_B T)$): To avoid complexity as reported in Ref. [14], we first cooled the sample at a magnetic field depicted in Fig. 1, and then measured the electrical resistivity at the field in a warming process. At every magnetic field, the slope of the $\rho(T)$ curve deviates from the linear behavior and bends to saturate at low temperature, which can be ascribed to variable range hopping mechanism as suggested by the so-called $T^{-1/4}$-law of $\rho(T)$ (not shown here). This feature becomes enhanced with increasing magnetic fields possibly

![Figure 1](image-url)

Figure 1. Temperature dependence of electrical resistivity of SmS (sample #8) in the Arrhenius plot at a fixed magnetic field at 1.4 kbar. Note that the electrical resistivity decreases drastically by application of a magnetic field; for example, a magnetic field of 30 T reduces the resistivity by 6 decades at 20 K.
because of reduced gap energy.

When looking at the electrical resistivity as a function of the magnetic field at a fixed temperature, the resistivity appears to be almost unchanged at room temperature but decreases drastically at low temperatures. This feature is remarkable at 30 T; the electrical resistivity is even metallic in the sense that the low-\(T\) resistivity is almost independent of temperature, although the system is considered to be still in black phase.

We estimated the gap energy \(E_g\) from the slope of the Arrhenius plot. The obtained \(E_g\) is summarized in Fig. 2 as a function of magnetic field at a fixed pressure. We clearly observe that \(E_g\) decreases drastically by application of the magnetic field, almost consistent with our previous measurements made up to 6.8 T for the different sample [12]. It is surprising that at 1.4 kbar, for instance, \(E_g \simeq 730\) K at \(H = 0\) is reduced down to \(E_g \simeq 50\) K at \(H = 30\) T, reflecting the 6 decades reduction of the electrical resistivity mentioned above. We anticipate that some discontinuous behavior will occur at high magnetic fields corresponding to the black to golden phase transition, but it has not been observed yet. A further experiment at higher

![Figure 2. Magnetic field dependence of the energy gap \(E_g\) at a fixed pressure. The error bar was estimated from the ambiguity in determination of the slope of the Arrhenius plot. Note that the gap decreases drastically by application of the magnetic field.](image-url)

![Figure 3. Pressure and magnetic field dependence of the energy gap \(E_g\). Closed diamonds show the pressure dependence of \(E_g\) at a zero field (bottom axis). Closed squares show the magnetic field dependence of \(E_g\) at ambient pressure (top axis). Data points of \(E_g\) at high pressures are plotted on a horizontal \(H\)-axis starting from a relevant \(P\) value. Note that the scaling between \(H\) and \(P\) is the same among all the pressures, \(H(T) = 10 \times P(\text{kbar})\).](image-url)
pressure and magnetic fields is in progress.

The reduction of $E_g$ by magnetic field seen in Fig. 2 may indicate that the magnetic field plays a similar role to the pressure. To check this possibility, we present $E_g$ as functions of the pressure and magnetic field in Fig. 3: Data points at zero field ($E_g(H = 0, P)$) denoted by closed diamonds are plotted on a bottom $P$-axis. Data points at ambient pressure ($E_g(H, P = 0)$) denoted by closed squares are plotted on a top $H$-axis, where $P$ and $H$ are related with each other via the relation of $H(T) = 10 \times P$(kbar). Data points at 0.7 kbar ($E_g(H, P = 0.7)$) denoted by open squares are plotted on a horizontal $H$-axis starting from $P = 0.7$ kbar on the bottom $P$-axis. Here, the same scaling relation between $H$ and $P$ was used. Other data points at 1.4, 2.0, and 2.2 kbar respectively denoted by closed circles, open circles, and closed triangles are similarly plotted with the same conversion relation. Note that the scaling factor mentioned above is an only adjustable parameter used for this plot, nevertheless we find all the data points to lie on a single line. This finding seems to indicate that the magnetic field actually serves as the pressure on resistivity, suggesting that if the pressure effect on black phase is intrinsic then the magnetic field effect is also intrinsic to black SmS. Indeed, the pressure effect is intrinsic, thus it is very likely that the negative magneto- resistance is inherent to SmS. One possibility of the magnetic field effect on resistivity is the Zeeman effect on the conduction band. In this case, although the $g$-factor is estimated for very large value, the external pressure and magnetic field show the equivalence on resistivity. To confirm this, we need a further experiment to observe the black to golden phase transition by application of magnetic field.

4. Conclusion
We measured the magnetoresistance of black SmS at high magnetic fields up to 30 T. We observed that magnetic fields drastically reduce the energy gap $E_g(H, P)$ in a similar way to that the pressure does, and that all the data points of $E_g(H, P)$ presented here lie on a single line as functions of $H$ and $P$. From this observation, we argued that the giant negative magneto-resistance is intrinsic to black SmS and the magnetic field plays a similar role to the pressure. We hope that the present result stimulates further experimental and theoretical investigations.

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