Electrical resistivity and thermal expansion investigation of golden SmS under pressure

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Abstract. We measured the electrical resistivity and the thermal expansion of SmS at high pressures. It is known that the golden SmS undergoes an antiferromagnetic phase transition at a critical pressure ($P_{c2} \sim 18$ kbar). At pressures slightly below $P_{c2}$, the temperature dependence of the electrical resistivity shows a local maximum (hump). Combining the thermal expansion results showing the two-phase mixture of the paramagnetic and antiferromagnetic phase in the vicinity of $P_{c2}$, we argue that the hump in the resistivity is caused by the pressure inhomogeneity. We also argue that the inhomogeneity gives rise to a striking effect on the pressure dependence of the residual resistivity, and speculate that in the limit of hydrostatic condition, the residual resistivity may show a rapid drop when the pressure is increased across $P_{c2}$.

1. Introduction

SmS undergoes an insulator-to-metal transition at a critical pressure ($P_{c1} \sim 7$ kbar at room temperature (RT)). This first-order phase transition involves a large volume collapse of about 8%, valence change from divalence to intermediate valence, and a color change from black to golden yellow \cite{1, 2}. It is noteworthy that the high pressure “golden phase” is not a simple metal but a pseudo-gapped state at low temperature \cite{3}. Upon further increasing pressure, an antiferromagnetic (AFM) phase transition takes place at another critical pressure ($P_{c2} \sim 18$ kbar at low temperature), below which the electrical resistivity $\rho(T)$ shows metallic behavior \cite{4, 5, 6}. According to our recent experiments of thermal expansion, there is a tricritical point in the pressure-temperature phase diagram, at which the Néel transition changes from first-order to second-order \cite{7}.

Although extensive efforts have been made for the last four decades, the nature of golden phase, especially of the paramagnetic (PM) phase below $P_{c2}$, is not clarified yet; for example, it is unclear whether the presence of a hump structure in $\rho(T)$ at pressures slightly below $P_{c2}$ is intrinsic or extrinsic (see Fig. 1) \cite{8, 9}. This arises from difficulties in high pressure experiments for SmS that is very sensitive to pressure inhomogeneity.

From our previous study on the thermal expansion under pressure, we showed that there is a two-phase mixture region (arising from the inhomogeneity) in the vicinity of $P_{c2}$ \cite{7}. This motivates us to study the relationship between the hump structure and the two-phase mixture state. In this report, we show that the hump of $\rho(T)$ is extrinsic due to the inhomogeneity.
further argue that as pressure increases, the residual resistivity \( \rho_0(P) \) may drop at \( P_{c2} \) in the limit of hydrostatic pressure.

2. Experimental technique

Single crystals of SmS were grown by the Bridgman method [10]. We note that physical properties including the electrical resistivity strongly depend on a starting composition of Sm and S. The samples used in this study were grown from a nominally stoichiometric composition. Electrical resistivity was measured by van der Pauw method (for samples \#A and \#B) or usual 4-probe method (for sample \#C). For sample \#A, we made a simultaneous measurement of the electrical resistivity and the thermal expansion (by the strain gauge technique). Daphne oil 7373 and Fomblin Y140/13 were used as a pressure transmitting medium for samples \#A and \#B and for sample \#C, respectively. Pressure was applied at RT for the experiments of samples \#B and \#C, and at 320 K for sample \#A to reduce the pressure inhomogeneity arising from the freezing of Daphne oil 7373 [7]. For all the samples, an outer BeCu and inner NiCrAl cylinder was used. Pressure at low temperatures was determined from a superconducting transition temperature of indium. The detailed description of the experimental technique is given elsewhere [11].

3. Results and Discussion

Figure 1 shows the temperature dependence of the electrical resistivity \( \rho(T) \) of sample \#A at pressures in the vicinity of \( P_{c2} \). Note that for a sample used here (sample \#A), \( P_{c2} \) was estimated as 18.1 ± 0.8 kbar [7]. At 16.6 kbar \(( < P_{c2} )\), \( \rho(T) \) increases monotonically with lowering temperature, possibly reflecting the pseudo gap formation. By contrast, at 19.1 kbar \(( > P_{c2} )\), \( \rho(T) \) initially increases with lowering temperature but turns to decrease below about 12 K, that is slightly higher than \( T_N \sim 10 \text{ K} \). Here, the Néel temperature \( T_N \) is defined by a temperature at which the thermal expansion coefficient exhibits a peak, as shown in the inset

![Figure 1. Temperature dependence of the electrical resistivity of sample \#A at several pressures. Note that the critical pressure \( P_{c2} \) above which AFM phase appears is evaluated as approximately 18.1 kbar. Inset shows the temperature dependence of the thermal expansion coefficient at the same pressures as the resistivity [7]. The shallow minimum at 16.6 kbar reflects the pseudogap formation in the PM state [3], while the sharp peak at 19.1 kbar corresponds to the Néel transition. Note that the peculiar feature of the curve at 18.0 kbar suggests the mixture of the PM and AFM state [7].](image-url)
of Fig. 1. (Note that the Néel transition corresponds to the steepest point of the $\rho(T)$ curve rather than the peak of the curve.) Remarkably, at 18.0 kbar, which is below $P_{c2}$ but very close to it, $\rho(T)$ forms a broad peak (hump) at approximately 10 K. Note that similar features were reported in the literature [8, 9].

We show in the inset of Fig. 1 the temperature dependence of the thermal expansion coefficient $\alpha(T)$ at pressures in the vicinity of $P_{c2}$. At 16.6 kbar (dotted curve), $\alpha(T)$ exhibits a shallow minimum at approximately 10 K, while at 19.1 kbar (broken curve), $\alpha(T)$ exhibits a peak at $T_N \sim 10$ K. At 18.0 kbar (solid line), these two features, i.e., the shallow minimum of the PM state and the anomaly at $T_N$, are superposed due to the inhomogeneity [7].

Let us examine the effect of this two-phase mixture on the electrical resistivity. As noted above, the hump structure of the $\rho(T)$ curve at 18.0 kbar emerges at approximately 10 K, and $T_N$ deduced from $\alpha(T)$ is about 9 K. Remembering that the hump appears slightly above $T_N$, we attribute the hump of the $\rho(T)$ curve to the Néel transition, and thus to the two phase mixture.

Figure 2 shows the pressure dependence of the residual resistivity $\rho_0(P)$ of the three samples investigated here. Here, $\rho_0$ is defined as $\rho(T)$ at 2 K, the base temperature of the present experiment. Upon increasing pressure toward $P_{c2}$, $\rho_0(P)$ initially increases and then passes through a broad maximum at $P_{\text{max}}$ ($\sim 13 - 16$ kbar) before steeply decreasing. At pressures above $P_{c2}$, $\rho_0$ becomes to be as small as the order of several tens of $\mu\Omega\text{cm}$, which may be a typical value of a metal.

It is noteworthy that $P_{\text{max}}$, and thus the transient region of $\rho_0(P)$, depends on the sample and/or pressure transmitting medium (see Fig. 2). It is reasonable to assume that the pressure inhomogeneity gives rise to the mixing of the high resistivity PM phase and the low resistivity AFM phase. This suggests that for sample #A (for which the most hydrostatic condition is satisfied among the samples), the transient region will be narrowest. Actually, sample #A exhibits the narrowest transient region. Furthermore, we note that the magnitude of the resistivity drop is greater for sample #A than for sample #C; Here, Fomblin (used for sample #C) yields less hydrostatic condition than Daphne oil (used for sample #A) because the former has the higher viscosity and lower freezing pressure ($\sim 10$ kbar) than the latter [11, 12].

![Figure 2](image_url) **Figure 2.** Pressure dependence of the electrical resistivity at $T = 2$ K of golden SmS. A shaded region denotes the two-phase mixture region of the PM and AFM phase in the vicinity of the critical pressure $P_{c2} \sim 18$ kbar for sample #A.
As a result, the resistivity drop in the vicinity of $P_{c2}$ seems to be obscured by the pressure inhomogeneity. This leads us to conjecture that under the ideally hydrostatic condition, the peak position would coincide with $P_{c2}$ and $\rho_0(P)$ would exhibit a discontinuous change at $P_{c2}$.

As mentioned in Introduction, the black-golden phase transition is accompanied by the large volume collapse. Therefore, the pressure inhomogeneity will induce some inhomogeneity in the sample during the transition. To extract intrinsic information of golden SmS, therefore, we need a further investigation under the better hydrostatic condition, although it is extremely difficult to perform.

4. Conclusion

We measured the electrical resistivity and the thermal expansion of golden SmS. At a pressure slightly below the critical pressure $P_{c2}$, the temperature dependence of the electrical resistivity shows the local maximum (hump). On the other hand, the thermal expansion showed the two-phase mixture of the PM and AFM phase at pressures in the vicinity of $P_{c2}$. Combining these results, we concluded that the hump in $\rho(T)$ is caused by the pressure/sample inhomogeneity. We also argued that the pressure dependence of the residual resistivity $\rho_0(P)$ is crucially influenced by the sample/pressure inhomogeneity, and speculated that $\rho_0(P)$ may show a rapid drop under the ideally hydrostatic condition when the pressure is increased across $P_{c2}$. This may lead to an interesting question regarding the relationship between the residual resistivity and the valence transition [7, 13].

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References

[1] Jayaraman A, Narayanamurti V, Bucher E and Maines R G. 1970 Phys. Rev. Lett. 25 1430.
[2] Wachter P: in Handbook on the Physics and Chemistry of Rare Earths, edited by Gschneidner K A, Eyring L Jr., Lander G H and Choppin G R. (North-Holland, Amsterdam, 1994), Vol. 19. p. 383.
[3] Matsubayashi K, Imura K, Suzuki H S, Chen G F, Môri N, Nishioka T, Deguchi K and Sato N K. 2007 J. Phys. Soc. Jpn. 76 033602.
[4] Haga Y, Derr J, Barla A, Salce B, Lapertot G, Sheikin I, Matsubayashi K, Sato N K and Flouquet J. 2004 Phys. Rev. B 70 220406.
[5] Barla A, Sanchez J P, Haga Y, Lapertot G, Doyle B P, Leupold O, Rüffer R, Abel-Elmeguid M M, Lengsdorf R and Flouquet J. 2004 Phys. Rev. Lett. 92 066401.
[6] Matsubayashi K, Imura K, Suzuki H S, Ban S, Chen G F, Deguchi K and Sato N K. 2007 J. Magn. Magn. Mater. 310 408.
[7] Imura K, Matsubayashi K, Suzuki H S, Kabeya N, Deguchi K and Sato N K. submitted to J. Phys. Soc. Jpn.
[8] Lapierre F, Ribault M, Holtzberg F and Flouquet J. 1981 Solid State Commun. 40 347.
[9] Konczykowski M, Morillo and Senator J P. 1981 Solid State Commun. 40 517.
[10] Matsubayashi K, Imura K, Suzuki H S, Mizuno T, Kimura S, Nishioka T, Kodama K and Sato N K. 2007 J. Phys. Soc. Jpn. 76 064601.
[11] Imura K, Matsubayashi K, Suzuki H S, Deguchi K and Sato N K. to appear in Physica B.
[12] Osakabe T and Kakurai K. 2008 Jpn. J. Appl. Phys. 47 6544.
[13] Miyake K, Narikiyo O and Onishi Y. 1999 Physica B 259-261 676.