The magnetoresistance of the quasi-one-dimensional conductor $NbSe_3$

2003 J. Phys.: Condens. Matter 15 5353

(http://iopscience.iop.org/0953-8984/15/31/302)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) 5353–5358 PII: S0953-8984(03)63990-1

The magnetoresistance of the quasi-one-dimensional conductor $NbSe_3$

Jing-qin Shen[1], **Xue-zhi Chen**[1], **Yi Zheng**[1], **Hong-tao Wang**[2] **and Zhu-an Xu**[1,3]

[1] Department of Physics, Zhejiang University, Hangzhou 310027, China
[2] Department of Physics and Electronic Information Science, Wenzhou Normal College, Wenzhou 325027, China

E-mail: zhuan@css.zju.edu.cn

Received 29 May 2003
Published 23 July 2003

Abstract

The temperature dependence of the resistivity and magnetoresistance (MR) of quasi-one-dimensional $NbSe_3$ was studied. The sharp increase of the positive MR in the lower charge-density wave (CDW) phase is consistent with previous reports, and the violation of Kohler's rule is obvious. We found that the MR data can be fitted very well with a modified two-band model, and the temperature dependence of the resulting parameters was discussed. The MR, the effect of magnetic field on the CDW gap, and the thermoelectric power of $NbSe_3$ can be coherently understood within this model.

1. Introduction

Quasi-one-dimensional conductors exhibit a variety of phases ranging from charge-density waves (CDW) and spin-density waves (SDW) to a metallic state to superconductivity in the low-temperature ground state. $NbSe_3$ is unique among CDW materials in that its ground state remains metallic or 'semimetallic' even after it has experienced two CDW transitions at $T_1 = 145$ K and $T_2 = 59$ K [1, 2]. It was estimated [2, 3] from the resistivity that only 20% of the Fermi surface (FS) is destroyed by the formation of the CDW gap at the upper transition ($T_1 = 145$ K) and about 60% of the remaining FS at the lower transition. Moreover, a very large magnetoresistance (MR) $\Delta\rho/\rho$ was reported in the temperature range $10\text{ K} < T < 50\text{ K}$ when a magnetic field was applied perpendicular to the high-conductivity axis (b-axis) [3–5].

Up to now, the nature of the large MR in $NbSe_3$ has remained ambiguous. In order to explain the large MR, a theory in which the CDW gap can be enhanced by a magnetic field was developed by Balseiro and Falicov (BF) [6]. Parilla *et al* obtained evidence of magnetic-field-induced enhanced gaps by measuring the variations of the narrow-band noise (NBN) frequency [3] and ac conductivity [7] with the applied magnetic field. They observed a 30%

---

[3] Author to whom any correspondence should be addressed.

increase in CDW carrier density at 30 K under a field of 7.5 T. However, Tritt *et al* [8] presented negative results; they found that even at $H = 10$ T the increase in the number of CDW carriers was less than 5% and showed that the results in [3] were in error, as a two-probe technique was used and H affected the contact resistance. Recently Tian *et al* [9] invoked a modified two-band model to explain the large MR observed in the purple bronzes AMo_6O_{17} (A = Na, K, and Tl) which are quasi-two-dimensional CDW materials. They found that a small decrease in number of the major carriers induced by the enhancement of CDW gap could account for the large MR.

In this paper, we reported the measurement of the MR in the lower CDW phase of $NbSe_3$ for $H \parallel c$. The violation of Kohler's rule [10] is obvious and the data can be fitted very well with the modified two-band model.

2. Experimental details

The $NbSe_3$ crystals were prepared by a two-step vapour transport method [11]. The details of the growth procedure were reported elsewhere [12]. Typical crystal dimensions were 5 mm × 10 μm × 1 μm, estimated from scanning electronic microscopy (SEM) photographs.

The resistivity of $NbSe_3$ was measured by a standard four-probe method. The MR measurement was performed on a Quantum Design PPMS-9 system with the magnetic field parallel to the c-axis. Electric contacts were made by attaching 38 μm gold wires to the sample using silver paint and the contact resistances were estimated as less than 1 Ω. The excitation current applied to the sample was carefully chosen such that the resulting voltages did not exceed the threshold voltage of CDW sliding.

3. Results and discussion

Figure 1 shows the temperature dependence of the resistivity of the $NbSe_3$ sample from room temperature down to 5 K. The inset of figure 1 shows the $\rho(H, T)$ curves at $H(\parallel c) = 0, 2, 5$ T for temperatures ranging from 60 to 10 K. We can see clearly that there are two sharp increases of the resistivity at $T_1 = 57$ K and $T_2 = 144$ K, corresponding to the two CDW transitions. The room temperature resistivity $\rho(300\text{ K})$ is 2.18 $\mu\Omega$ m and the residual resistivity ratio, $\text{RRR} = \rho(300\text{ K})/\rho(5\text{ K})$, is about 223, which are in agreement with other reports on high-quality pure $NbSe_3$ crystals [1–5]. The large positive magnetoresistance appears as soon as the temperature drops below T_2 and the position of the maximum of $\rho(T)$ is shifted from $T \sim 40$ to 30 K by a magnetic field of 5 T. T_2 is independent of the magnetic field within our experimental error of ±0.2 K.

On the basis of the increase of the resistivity at CDW transitions, Ong's group [2] and Parilla *et al* [3] made a simple estimate of the ratio (α) of destroyed FS to total FS, and proposed a model to use for calculating the density of condensed electrons n_{CDW}. Ong and Monceau [2] reported α_1 for the first CDW transition to be about 0.2, which means about 20% of the FS is destroyed by the CDW transition. And α_2 for the second phase transition is about 0.62, which means another 62% of the area of the rest of the FS is destroyed after the second transition. Without an applied magnetic field, the calculated values of α_1 and α_2 for our $NbSe_3$ sample are 0.22 and 0.63 respectively, which are close to Ong and Monceau's results. If we assume that the large MR is caused by the conversion of normal carriers to CDW carriers induced by the magnetic field, which was proposed by BF [6], a 5 T magnetic field (along the c-axis) increases $\alpha_2(H)$ from 0.63 to 0.78, i.e., causes a 25% increase in number of the CDW carriers, which is quite comparable to the 30% increase under a field of 7.5 T reported by Parilla *et al* [3]. However, this estimate of $\alpha_2(H)$ is based on the assumption that the applied

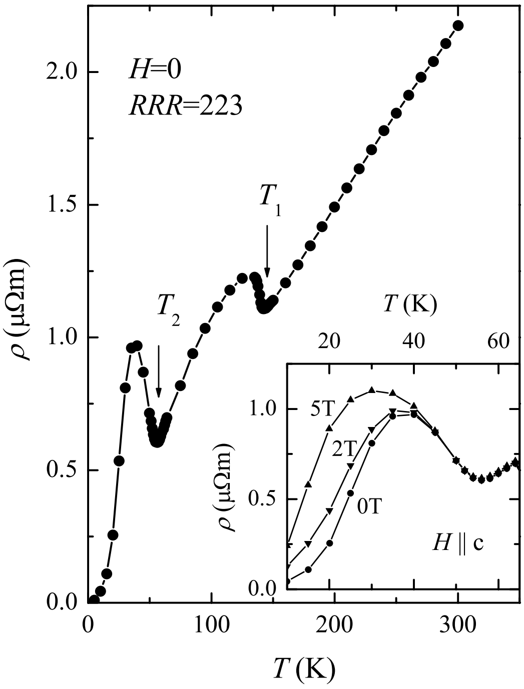

Figure 1. The temperature dependence of the resistivity of $NbSe_3$ for temperatures ranging from 300 to 5 K without an applied magnetic field. Inset: resistivity versus temperature for magnetic fields of 2 and 5 T for 10 K $< T <$ 60 K. The two arrows indicate the positions of the two CDW transitions.

magnetic field mainly causes the reduction in the area of the remaining FS by enhancing the CDW gap. According to the NBN measurement reported by Tritt *et al* [8], the increase of n_{CDW} is less than 5% under a magnetic field of 10 T, inconsistent with above assumption.

It is well known that the semiclassical transport theory predicts Kohler's rule to hold if there is a single type of charge carrier and the scattering time is isotropic on the FS [10]. Kohler's rule is given by

$$\frac{\Delta\rho(H,T)}{\rho(0,T)} = F(\omega_c\tau) = f\left(\frac{H}{\rho(0,T)}\right). \tag{1}$$

Here ω_c is the cyclotron frequency, τ the scattering time, $\rho(0,T)$ the zero-field resistivity, and $\Delta\rho(H,T) \equiv \rho(H,T) - \rho(0,T)$. The corresponding plots are known as Kohler's plots. Figure 2 (lower panel) shows the Kohler's plot of the MR for 10 K $\leqslant T \leqslant$ 60 K. The upper panel shows the semilogarithmic plot of $\Delta\rho(H,T)/\rho(0,T)$ versus T. $\Delta\rho(H)/\rho$ at 10 K is as large as 440% when $H = 5$ T. From figure 2, we can conclude that Kohler's rule is obviously violated in this temperature range (the curves for 2 and 5 T cannot be scaled to a universal curve). It has also been pointed out in [5] that the large MR is not associated with ordinary MR, i.e., Kohler's rule is not satisfied. Such a violation of Kohler's rule should be a rational consequence if there are two types of charge carrier and their ratio is affected by the magnetic field.

Stimulated by the reports of the unusual temperature dependence and sign change of the thermopower S in the lower CDW phase of $NbSe_3$ [13–15], we invoked a modified two-band model to fit the MR data. It has been reported that the MR of the quasi-two-dimensional purple bronzes AMo_6O_{17} (A = Na, K, and Tl) can be fitted well with this model [9]. Assuming that the

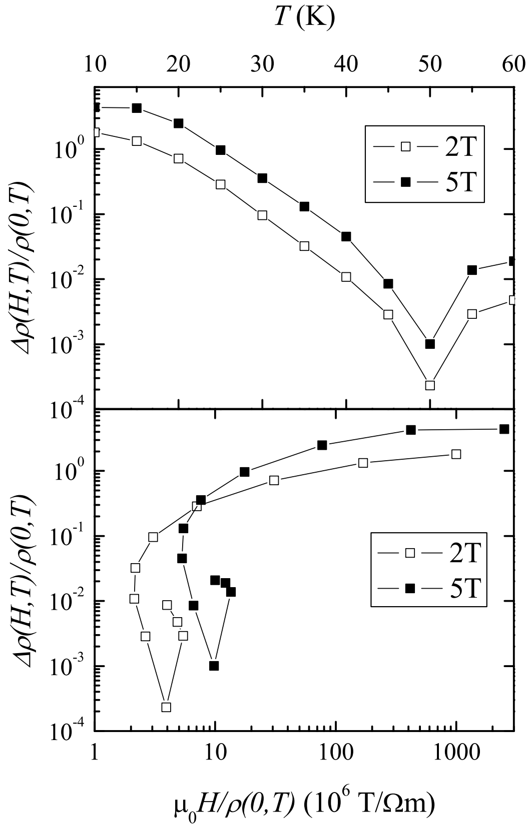

Figure 2. The lower panel shows the Kohler's plot of the MR for 10 K $< T <$ 60 K. The upper panel shows the semilogarithmic plot of $\Delta\rho(H, T)/\Delta(0, T)$ versus T.

two types of carrier have the same relaxation time and mass in the two-band galvanomagnetic effect model of Noto and Tsuzuku [16], the MR can be expressed as [9]

$$\frac{\Delta\rho}{\rho} = \frac{4(\beta - \gamma H)\bar{\mu}^2 H^2}{(1 + \beta - \gamma H)^2 + (1 - \beta + \gamma H)^2\bar{\mu}^2 H^2}. \tag{2}$$

Here β is the ratio of two types of carrier at $H = 0$. $n_2/n_1 = \beta - \gamma H$, where γ is a constant representing the effect of magnetic field on the ratio of carrier concentrations. n_1 and n_2 are the concentrations of the two types of carrier. $\bar{\mu}$ is the mobility for both types of carrier (we have assumed that the mobilities of the two carrier types are the same). It should also be noted that the parameter β is symmetrical for interchange of n_1 and n_2. Figure 3 shows the field dependence of the MR at T = 20, 40, and 60 K and the curves for the best fit with equation (2). The solid squares represent the experimental data and the solid lines are the fitting curves. The MR data for different temperatures can all be fitted with equation (2) very well, and the resulting parameters β, γ, and $\bar{\mu}$, determined from the best fit, are listed in table 1.

As the temperature decreases from 60 K, the parameter β drops fast, γ changes nonmonotonically, and $\bar{\mu}$ increases (as listed in table 1). We should point out that the resulting parameters are reasonable and meaningful. The sharp drop of the ratio $\beta = \frac{n_2}{n_1}|_{H=0} = \frac{n_2(0)}{n_1(0)}$ with temperature after the second CDW transition at T_2 is consistent with the studies of the thermopower in NbSe$_3$ [13–15] and band-structure calculations [17]. According to band-structure calculations [17], the normal-state FS consists of several open sheets (corrugated

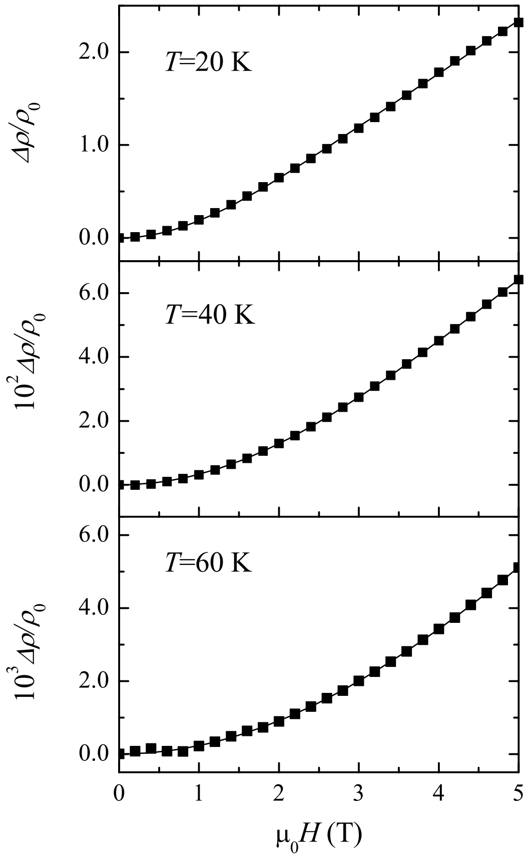

Figure 3. Magnetoresistance $\Delta\rho/\rho$ versus H at $T = 20$, 40, and 60 K. The solid squares represent the experimental data and the solid lines are the curves for the best fit with equation (2).

Table 1. Fitting parameters estimated from the modified two-band model for NeSe$_3$ at $T = 20$, 40, and 60 K.

Temperature (K)	$\beta = n_1(0)/n_2(0)$	γ	$\bar{\mu}$ (m^2 V^{-1} s^{-1})
20	3.27	0.135	0.522
40	29.46	1.12	0.160
60	100.36	0.080	0.077

planes) and an inner multiply connected surface. Each corrugated surface is, generally speaking, associated with one chain and one band. The thermopower S becomes a large negative value after the first CDW transition at $T_1 = 145$ K, which implies that the electrons become the majority carriers. That is, the formation of the first CDW gap removes most parts of the hole-like sheets and leaves some electron pockets on the FS. In the lower CDW phase, the absolute value of S decreases and finally becomes positive at about 10 K, which was explained as reflecting another CDW gap developing to destroy the electronic pockets on the FS and the concentration of electrons dropping. In this scenario, there are two types of charge carrier and their concentrations are comparable when $T > T_1$. When $T_2 < T < T_1$, the majority charge carriers are electrons due to the formation of a CDW gap, and the ratio β increases (n_2 represents the concentration of electrons in this case) and may go to a maximum at a temperature just above T_2. The value of β is about 100 at $T = 60$ K from our study, which means the concentration of electrons is 100 times larger than that of holes. When $T < T_2$, the concentration of electron-like carriers drops due to the destruction of the electron pockets on the FS by the formation of the second CDW gap, and holes finally become the dominant carriers at $T \sim 10$ K. Therefore the ratio β should drop fast in the lower CDW phase. We estimated β to be 29.46 and 3.27 at $T = 40$ and 20 K respectively, quite consistent with the above analysis. The increase of the average mobility $\bar{\mu}$ with decreasing temperature, which means the increase of the scattering time or electron mean free path, can be well understood within semiclassical transport theory. As regards the constant γ, its nonmonotonic variation with temperature suggests that the effect of the magnetic field is very subtle. To obtain equation (2), it has been assumed that the magnetic field preferentially affects one type of carrier. A small change in the ratio of two types of carrier caused by the magnetic field can account well for the large MR, and thus it is not necessary to assume a large increase of n_{CDW} with applied magnetic field. Accordingly it can be understood why the increase of n_{CDW} measured by NBN is much less than that estimated from the magnetotransport measurement. The increase of n_{CDW} estimated from MR data has been exaggerated in the previous reports [3–5].

In conclusion, the large MR in the lower CDW phase of $\mathrm{NbSe_3}$ was studied in the framework of a modified two-band model and the temperature dependence of the resulting parameters was qualitatively consistent with the results of thermopower studies [13–15] and band-structure calculations [17]. Our results suggest that the observed large MR can be explained well by the effect of magnetic field on the subtle compensation of two types of charge carrier without assuming an enormous increase in the concentration of condensed CDW electrons. The mechanism for the MR relating to the magnetic-field-induced enhancement of the CDW gap survives, but the effect on the normal carriers is smaller than that expected from the BF theory.

Acknowledgments

We are grateful to Dr Chunmu Feng for help with using Quantum Design PPMS system. This work was supported by the National Science Foundation of China under Grant No 10225417.

References

[1] Monceau P, Ong N P, Portis A M, Meerschaut A and Rouxel J 1976 *Phys. Rev. Lett.* **37** 602

[2] Ong N P and Monceau P 1977 *Phys. Rev.* B **16** 3443

[3] Parilla P, Hundley M F and Zettl A 1986 *Phys. Rev. Lett.* **57** 619

[4] Coleman R V, Eiserman G, Everson M P, Johnson A and Falicov L M 1985 *Phys. Rev. Lett.* **55** 863

[5] Coleman R V, Everson M P, Lu H A, Johnson A and Falicov L M 1990 *Phys. Rev.* B **41** 460

[6] Balseiro C A and Falicov L M 1985 *Phys. Rev. Lett.* **55** 2336
Balseiro C A and Falicov L M 1986 *Phys. Rev.* B **34** 863

[7] Hundley M F, Parilla P and Zettl A 1986 *Phys. Rev.* B **34** 5970

[8] Tritt T M, Gillespie D J, Ehrlich A C and Tessema G X 1988 *Phys. Rev. Lett.* **61** 1776

[9] Tian M, Yue S and Zhang Y 2002 *Phys. Rev.* B **65** 104421

[10] See, e.g., Ziman J M 1963 *Electrons and Phonons* (London: Clarendon) p 492

[11] Thorne R E 1992 *Phys. Rev.* B **45** 5804

[12] Wang H T, Zheng Y, Chen X Z and Xu Z A 2003 *Chin. J. Mater. Res.* **17** 247

[13] Kwak J F, Greene R L, Chaikin P M and Ong N P 1979 *Proc. Int. Conf. on Quasi-One-Dimensional Conductors* ed S Barisic, A Bjelis, J R Cooper and B A Leontic (Berlin: Springer) p 348

[14] Takagaki T, Ido M and Sambongi T 1978 *J. Phys. Soc. Japan Lett.* **45** 2039

[15] Hundley M F and Zettl A 1987 *Solid State Commun.* **61** 587

[16] Noto K and Tsuzuku T 1975 *Japan. J. Appl. Phys.* **14** 46

[17] Shima N and Kamimura H 1985 *Theoretical Aspects of Band Structure and Electronic Properties of Pseudo-One-Dimensional Solids* ed H Kamimura (Dordrecht: Reidel) p 231