Pressure effect on large magnetoresistance in the lower charge-density-wave transition of $NbSe_3$

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A large magnetoresistance appearing below the T_{c2}-CDW (charge-density wave) transition temperature in $NbSe_3$ has been studied under various pressures including pressure above the critical pressure where the T_{c2}-CDW phase is suppressed totally. The large magnetoresistance appears as long as the T_{c2}-CDW phase exists. However, when the phase is suppressed totally by high pressure, neither large magnetoresistance nor any anomalies are induced by the magnetic field up to 12 T, suggesting that it is negative for the formation of a magnetic-field-induced CDW. The origin of the large magnetoresistance is discussed in terms of normal carriers on the ungapped Fermi surface created on the CDW transition compared with the pressure-induced magnetoresistance observed newly above T_{c2}. [S0163-1829(99)13227-2]

I. INTRODUCTION

Niobium triselenide, $NbSe_3$, which is known as a typical quasi-one-dimensional (Q1D) metal, exhibits two incommensurate charge-density-wave (CDW) transitions at $T_{c1} = 142$ K and $T_{c2} = 58$ K.[1–3] In most CDW materials, the formation of a CDW gap results in a complete destruction of the Fermi surface, ending in a semiconducting ground state at temperatures below the transition. However, $NbSe_3$ exhibits a metallic or rather a semimetallic behavior at the low-temperature regime. This has been understood by the existence of normal carriers in small pockets created by the two CDW phase transitions. Band calculations[4,5] of $NbSe_3$ by Shima and Kamimura demonstrate that the T_{c1}-CDW transition is attributable to a nearly perfect nesting between two quasiparallel sheets of the Fermi surface, while the T_{c2}-CDW transition is due to an imperfect nesting of the remaining part of the Fermi surface. In fact, large Shubnikov–de Haas oscillations due to electron and hole ellipsoidal pockets have been observed at low temperatures well below T_{c2}.[6] So it is accepted that the ground state of $NbSe_3$ is a semimetallic CDW state at low temperatures.

The study of the magnetic-field dependence of CDW in $NbSe_3$ has received considerable attention since the discovery of a large magnetoresistance below the T_{c2}-CDW transition by Coleman *et al.*[6,7] They observed that when a magnetic field is applied perpendicular to the high conductivity axis, the large magnetoresistance appears just below the T_{c2}-CDW transition and increases rapidly with decreasing temperature, and its value is above 400% at low temperatures around 10 K. It was also found that the large magnetoresistance is very anisotropic to the direction of the applied magnetic field, suggesting that the orbital response of the electrons to the magnetic field plays an important role.[6]

Balseiro and Falicov[8,9] proposed a theory in which the magnetic field constrained the electronic motion to improve the nesting condition of a Q1D Fermi surface and resulted in the enhancement of the CDW gap. Similar models have been accepted as a standard model for a formation mechanism of the field-induced spin-density-wave (SDW) observed in Q1D organic conductors.[10] Many transport experiments such as narrow-band noise,[11,12] Hall effect,[13] and the thermopower[14] have been tried in the transverse magnetic fields to investigate whether the CDW order parameter of $NbSe_3$ is enhanced by the magnetic field. However, these results were negative for the change in the CDW order parameter with magnetic field.

In addition to the transport data, recently Kiryukhin *et al.*[15] have performed an x-ray-scattering study in magnetic fields up to 10 T to investigate a magnetic-field-induced shift of the nesting vector and the T_{c2}-CDW order parameter in $NbSe_3$. They found that there was $\Delta q_b / q_b \leqslant 2.5\times 10^{-3}$ on the magnetic-field-induced shift of the nesting vector of the T_{c2}-CDW, and the magnetic-field dependence of the T_{c2}-CDW order parameter was independent on the magnetic field to within 10%. These results may also be negative for the change in the CDW order parameter with magnetic field. Both the results of the magnetotransport and the x-ray scattering lead us to the prediction in which the large magnetoresistance appearing below T_{c2} could be simply explained by considering light carriers in small ungapped pockets of the Fermi surface generated by the imperfect nesting on the formation of the T_{c2}-CDW state, as pointed out by Tritt *et al.*[11,12] However, quantitative estimates of such effects have not yet been reported.

As shown in the P-T-H phase diagram of SDW in Q1D organic conductors,[10] the field-induced SDW phase appears in the high magnetic field near the critical pressure where the SDW phase is suppressed totally in zero magnetic field. On the other hand, although the P-T-H phase diagram of CDW in $NbSe_3$ is not yet settled, it is well known that in zero magnetic field the T_{c2}-CDW phase of $NbSe_3$ is suppressed totally by pressure above the critical pressure of 7.5 kbar.[16] It is important to investigate magnetic-field effects on CDW of $NbSe_3$ in the region near the critical pressure in order to see whether the CDW order parameter is changed by the magnetic field.

In this paper, we report on the experimental results on the

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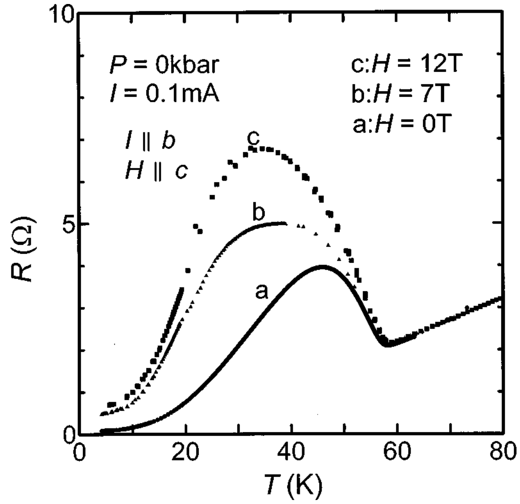

FIG. 1. Temperature dependence of the resistance in the range 2–80 K at ambient pressure at $H=0$, 7, and 12 T.

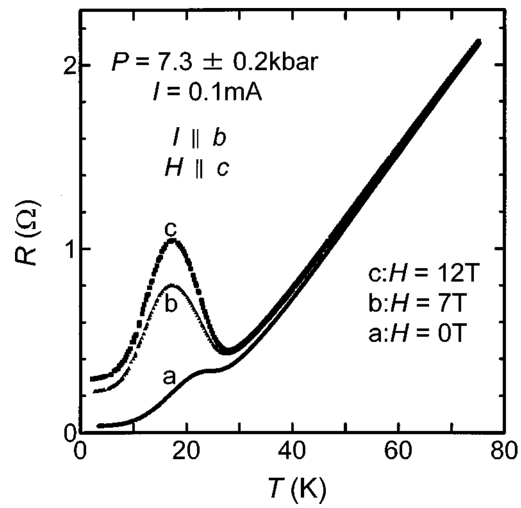

FIG. 2. Temperature dependence of the resistance in the range 2–80 K at $P=7.3\pm0.2$ kbar at $H=0$, 7, and 12 T.

large magnetoresistance appearing at the T_{c2}-CDW transition as a function of temperature under various pressures including pressure above the critical pressure, which suppresses totally the T_{c2}-CDW phase. It is found that neither large magnetoresistance nor any anomalies appear in a high magnetic field when the T_{c2}-CDW phase is totally suppressed. This is contrary to the case of the field-induced SDW, supporting the idea that the origin of the large magnetoresistance may be due to light carriers in small pockets created at T_{c2} rather than the enhancement of the CDW gap with magnetic field. Furthermore, we find newly a pressure-induced magnetoresistance which appears from a temperature well above T_{c2}, although the value is two orders smaller than that of the large magnetoresistance below T_{c2}. The origin is discussed with comparing behaviors of the large magnetoresistance.

II. EXPERIMENT

The $NbSe_3$ crystals were grown by vapor transport in sealed evacuated quartz tubes using Nb wire of 99.9% purity and Se shots of 99.999% and heated to 740 °C for two weeks at a temperature gradient of approximately 1 °C/cm. The pure crystal gave residual resistance ratios $R(300\,\mathrm{K})/R(4.2\,\mathrm{K})$ in the range 50–60. The resistance of $NbSe_3$ was measured by a usual four-probe dc method, where a current is parallel to the *b* axis and a magnetic field is parallel to the *c* axis. The samples were attached to four gold wires with Dupont silver paint (No. 4929). A superconducting magnet was used for magnetic fields in the range 0–12 T. Pressures were generated by use of a WC piston and a beryllium-copper clamp type cylinder with the internal diameter of 6 mm ϕ. Transmitting liquid was a 1:1 mixture of Fluorinert FC70 and FC77. Pressures were estimated from the pressure dependence of T_{c1} and T_{c2} in $NbSe_3$ based on a previous result.[16]

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the resistance of $NbSe_3$ in the magnetic fields at ambient pressure. The magnetic fields enhance the resistance anomaly associated with the formation of the lower CDW state. It is noted that the large magnetoresistance appears as soon as the T_{c2}-CDW phase is formed, and it is negligibly small above T_{c2}. These results agree well with previous results.[6,7] T_{c2} is independent of the magnetic fields within our experimental error of ±0.5 K. This is not inconsistent with previous results by Coleman *et al.*, in which the T_{c2} increases only by 0.5 K, or less, in a field in excess of 20 T.[6,7] Thus, T_{c2} remains essentially unchanged by magnetic fields.

Figure 2 shows the temperature dependence of the resistance in the magnetic fields under pressure at $P=7.3\pm0.2$ kbar. The lower CDW transition is observed at $T_{c2}=25$ K under zero magnetic field. The large magnetoresistance is also observed below T_{c2} under pressure. We find that the temperature where the large magnetoresistance appears runs after T_{c2}, which decreases with increasing pressure. The value of T_{c2} under various pressures is found to be almost constant to the magnetic field up to 12 T. This result is contrary to the case of the field-induced SDW, whose transition temperature strongly depends on magnetic fields.[10] Moreover, it is found under pressure that a small enhancement of the resistance with the magnetic fields is observed even above T_{c2}. The small magnetoresistance above T_{c2} tends to increase with increasing pressure. So we call it the pressure-induced magnetoresistance.

Figure 3 shows the temperature dependence of the resistance R and its temperature derivative dR/dT for (a) $H=0$ T and (b) $H=12$ T under applied pressure at $P=8.3\pm0.8$ kbar, which is larger than the critical pressure (7.5 kbar) of the T_{c2}-CDW phase.[16] In zero magnetic field [Fig. 3(a)], there is no anomaly associated with the T_{c2}-CDW phase in both curves of R versus T and dR/dT versus T. At the same time, the superconductivity occurs at $T_s=2.5$ K. Here, the superconducting transition temperature T_s is defined as the temperature where the resistance becomes one-half of the resistance in the normal state. The value of T_s is slightly smaller than that of the earlier result,[16] which may be due to pressure inhomogeneity in a pressure cell. In the high magnetic field [Fig. 3(b)], we also find no sign for the superconductivity and no anomaly associated with the formation

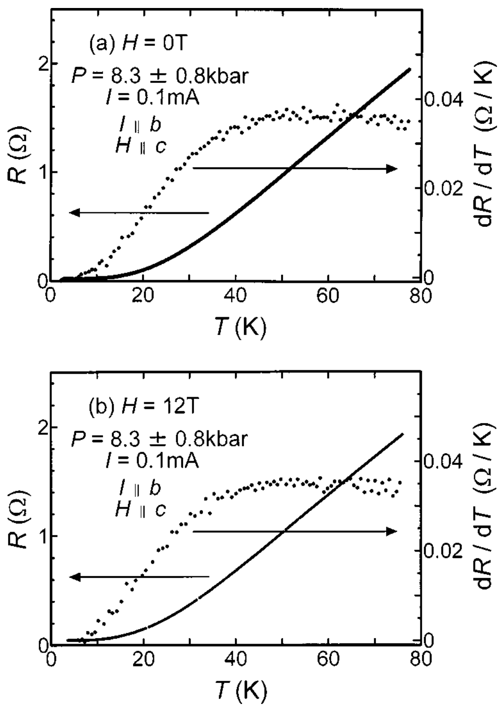

FIG. 3. Temperature dependence of the resistance R and its temperature derivative dR/dT at (a) $H=0$ T and (b) $H=12$ T at $P=8.3\pm0.8$ kbar. The superconductivity appears at $T_s=2.5$ K at $H=0$ T.

of the CDW gap induced by the magnetic field (the field-induced CDW) over the whole temperature range measured. This is contrary to the case of the field-induced SDW, which appears in the region near the critical pressure,[10] suggesting that it is negative for the formation of the field-induced CDW, at least within the magnetic field measured up to 12 T.

Figure 4 shows the temperature dependence of the magnetoresistance $\Delta\rho/\rho_0$ under various pressures in the magnetic field of $H=12$ T. In the case of the presence of the T_{c2}-CDW phase, the large magnetoresistances rapidly appear just below T_{c2} for the applied pressure from 0 to 7.3 ±0.2 kbar. These values are very large and are from 400% to 700% at low temperatures. At ambient pressure, $\Delta\rho/\rho_0$ is very small above T_{c2}, which is consistent with previous reports.[6,7] Under the pressures, however, the pressure-induced magnetoresistance is observed above T_{c2} and its appearance temperature increases with increasing pressure. On the other hand, in the case of the absence of the T_{c2}-CDW phase, the large magnetoresistance disappears totally under the high pressure of $P=8.3\pm0.8$ kbar even in the strong field of $H=12$ T, while the pressure-induced magnetoresistance is observed below 77 K. The temperature dependence of the pressure-induced magnetoresistance at $P=8.3\pm0.8$ kbar increases gradually as temperature decreases. The value at the low-temperature range is below 200%, quite a bit smaller than that of the large magnetoresistance, namely, the behavior of the pressure-induced magnetoresistance is found to be clearly different from that of the large magnetoresistance appearing below T_{c2}.

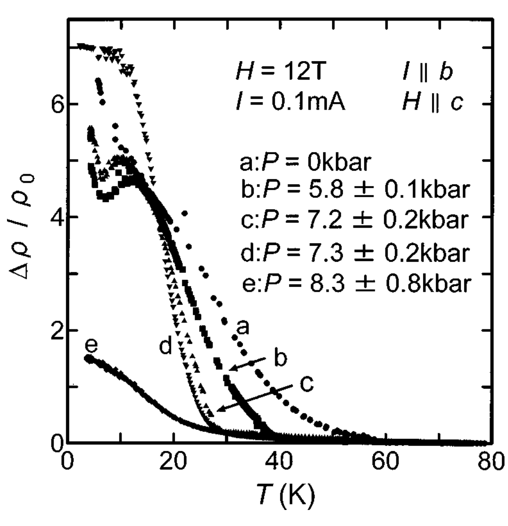

FIG. 4. Temperature dependence of the magnetoresistance $\Delta\rho/\rho_0$ at various pressures in the magnetic field of 12 T.

As shown by Shima and Kamimura,[4,5] the T_{c2}-CDW transition is attributable to an imperfect nesting. As a result, small ungapped pockets of the Fermi surface are created below T_{c2}, while there are few pockets above T_{c2} because of a nearly perfect nesting of the T_{c1}-CDW phase. It is natural to assume that carriers with light mass are generated at the ungapped Fermi surface below T_{c2}. If the large magnetoresistance is caused by these carriers, we can easily understand the appearance of the large magnetoresistance below T_{c2} and the absence of that above T_{c2}. Moreover, when the T_{c2}-CDW phase is suppressed totally under high pressure, the pockets will also disappear at the same time, leading to the absence of the large magnetoresistance. Thus, we can explain consistently our results by assuming the existence of carriers generated at the ungapped Fermi surface.

We discover the pressure-induced magnetoresistance above T_{c2}. The pressure-induced magnetoresistance is observed in spite of an absence of the T_{c2}-CDW phase and a disappearance of the large magnetoresistance. The pressure-induced magnetoresistance always appears above T_{c2} and its temperature dependence is independent of the value of T_{c2}. Band-structure calculations[4,5] demonstrate that the T_{c1}-CDW transition corresponds to a nearly perfect nesting between two quasiparallel sheets of the Fermi surface, while the T_{c2}-CDW transition is due to an imperfect nesting. This means that above T_{c2} there are few electron and hole pockets in the T_{c1}-CDW phase. However, applying pressure leads to a collapse of the nearly perfect nesting of the Fermi surface in the T_{c1}-CDW transition, that is, the decrease in the nesting size of the Fermi surface. As a result, the number of the normal carriers will increase with increasing pressure. Thus, it is considered that the pressure-induced magnetoresistance might result from carriers in pockets created by an imperfect nesting induced by pressure. Here, it should be emphasized that our explanation for the pressure-induced magnetoresistance is not inconsistent with changes in the Fermi surface associated with the chain corresponding to the T_{c1}-CDW, which has been pointed out in the NMR studies by Shi *et al.*[17] and uniaxial stress studies under magnetic fields with fermiology changes by Kuh *et al.*[18]

IV. CONCLUSIONS

In conclusion, we have investigated the magnetoresistance as a function of temperature under various pressures in

$NbSe_3$. It is found that the large magnetoresistance always appears as long as the T_{c2}-CDW phase exists, while when the T_{c2}-CDW phase is suppressed totally by high pressure, neither large magnetoresistance nor any field-induced anomalies are observed even in the high magnetic field of $H=12$ T. This suggests that the large magnetoresistance might not result from the change in the CDW order parameter with magnetic field but rather from light carriers in small ungapped pockets of the Fermi surface generated by an imperfect nesting of the Fermi surface. Furthermore, we discover the pressure-induced magnetoresistance above T_{c2}. It can be explained by assuming the existence of carriers in small pockets created by a pressure-induced imperfect nesting of the T_{c1}-CDW. These results may be attributable to the semimetallic CDW character of $NbSe_3$. We believe that when the T_{c1}-CDW phase is suppressed totally by higher pressure,[19] the pressure-induced magnetoresistance will disappear similar to the case of the large magnetoresistance. Such a study is in progress now.

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