Tunneling spectroscopy of $NbSe_3$ in a high magnetic field

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We have measured the differential current-voltage characteristics of normal-metal–$NbSe_3$ direct point contacts (without an insulating barrier) formed along the *b* axis under applied magnetic field with different orientations. At $T=4.2$ K two charge-density-wave (CDW) gaps, Δ_{p1} and Δ_{p2}, corresponding, respectively, to the high- and low-temperature CDW's were observed simultaneously as singularities of the excess resistance, which is attributed to the reflection injected from normal-metal carriers on the Peierls energy-gap barriers. The applied magnetic field up to 8.5 T does not lead to a change in the density of states and the Peierls energy gaps, suggesting that the large magnetoresistance observed in $NbSe_3$ might not result from the change in the CDW order parameter with magnetic field, but rather from the increasing scattering of carriers which are not condenced to CDW.

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$NbSe_3$ is known as a typical quasi-one-dimensional conductor, exhibiting two incommensurate charge-density-wave (CDW) transitions at $T_{p1}=145$ K and $T_{p2}=59$ K.[1,2] In contrast to most CDW materials, for which the Peierls transition completely destroys the Fermi surface, resulting in a semiconducting ground state, $NbSe_3$ exhibits metallic or semimetallic behavior up to very low temperatures. Such a behavior occurs because normal carriers in small pockets remain, due to imperfect nesting. Indeed, large Shubnikov–de Haas oscillations resulting from electron and hole pockets have been observed at temperatures well below T_{p2}.[3–5]

The magnetic-field dependence of the CDW properties has been of interest since the discovery of very large magnetoresistance in $NbSe_3$ below T_{p2} with the magnetic field, *H*, (more than 20 T) applied perpendicular to the conducting chains.[6] It was also found that the large magnetoresistance is very anisotropic with respect to the direction of *H*, suggesting that the orbital response of electrons to the magnetic field plays an important role. It was assumed that this effect is not caused by a direct interaction of *H* with the CDW but by the magnetic-field dependence on either the number or the mobility of noncondensed normal carriers.

Balseiro and Falicov[7,8] proposed a theory describing the effect of a magnetic field on the electronic structure of very anisotropic systems with a CDW. It was shown that in materials where there is lack of perfect nesting ($NbSe_3$ is just one), a high magnetic field applied along certain directions destroys electron and hole pockets and induces gaps in the spectrum at the Fermi level, decreasing the density of states (DOS) and thus, transforming a semimetallic behavior into a semiconducting one.

Many experiments were performed in transverse magnetic field to investigate whether the CDW energy gap of $NbSe_3$ is increased and the DOS of pocket carriers is decreased by the magnetic field. In Ref. 9 two-probe resistance and narrow-band-noise (NBN) measurements in $NbSe_3$ for *H* up to 75 kG were reported. It was concluded that a 30% increase in the CDW carriers took place: this was obtained from the slope of the CDW current versus NBN frequency. However, this result was not been confirmed in works[10,11] in which a similar experiment in $NbSe_3$ was performed with the help of a four-probe method for *H* up to 9.6 T. It was found that such a magnetic field induces a change of less than 5–6% in the CDW carrier concentration. But according to Ref. 12, the concentration of noncondensed electrons at *T* well below T_{p2} is 10^{18} cm^{-3} while the concentration of the CDW electrons is 10^{21} cm^{-3}. A 30% increase in the CDW carrier concentration means that the concentration of noncondensed carriers is more than 3×10^{20} cm^{-3}. This value is more than two orders-of-magnitude larger than the real concentration of noncondensed carriers. Therefore the result obtained in Ref. 9 seems unrealistic. On the other hand, the accuracy of 10^{-2} achieved in Refs. 10 and 11 corresponds to concentration of order 10^{19} cm^{-3}, which is larger than the concentration of noncondensed carriers, too, and, therefore, is not sufficient for reaching a definite conclusion about the existence or the absence of condensation of pocket electrons in the CDW ground state induced by *H*. Hall-effect measurements at high magnetic fields cannot give direct information regarding normal carrier concentration, because of quantum oscillating behavior of the Hall voltage (equivalent to the Shubnikov–de Haas oscillations of the resistivity).[5] The x-ray-scattering measurements of a possible magnetic-field-induced shift of the nesting wave vector in $NbSe_3$ (Ref. 13) show that $\Delta q_b/q_b<2.5\times10^{-3}$. But again, the accuracy achieved is not sufficient to draw definite conclusions about the existence or the absence of condensation of pocket electrons in the CDW.

Direct measurements of the energy gap and DOS in the presence of a magnetic field, for example, using tunneling spectroscopy would be a way to clarify the physical picture. In most known works with the aim of investigating the DOS behavior in $NbSe_3$ by means of tunneling spectroscopy, planar CDW-insulator-superconductor or CDW-insulator-normal-metal junctions were used.[14–16] But, the uncertainty of the quality of the insulating barrier does not allow to obtain reproducible tunneling spectra with a clear energy-gap structure. In our previous work we reported experimental

results of the point-contact spectroscopy of the energy gap in $NbSe_3$ in different crystallographic orientations.[17] The tunneling spectra obtained show a perfect energy-gap structure with good reproducibility. In the present paper we report point-contact measurements of the energy gaps in $NbSe_3$ in magnetic fields up to 8.5 T at $T=4.2$ K.

The experimental method we used is based on reflection injected from normal-metal carriers on the barrier associated with the Peierls energy gap in direct contacts formed between normal metals and the CDW. It was shown experimentally[18–20] and theoretically[21,22] that at the normal-metal (N)-CDW boundary, the injected carriers from normal metals are reflected due to the Peierls energy-gap barrier when their energy is less than Δ_p. The result of such an interaction of injected particles with a CDW is the appearance of an excess resistance at the N-CDW interface at $V \leqslant \Delta_p/e$, which provides the possibility to determine the energy gap directly from the N-CDW point-contact characteristics.

For our investigations, we chose the contact direction along the *b* axis (chain direction) because this direction is perpendicular to the Fermi surface. Another reason for choosing such a direction is the very large conductivity anisotropy of $NbSe_3$. In fact, according to Ref. 23, the conductivity anisotropy along the *b* and a^* axes is $\sigma_b/\sigma_{a^*} \simeq 10^3 - 10^4$, and that along the *b* and *c* axes is $\sigma_b/\sigma_c \simeq 10$ (see Ref. 12). Therefore, the contacts oriented along the *b* axis should be the most directional. At the same time, for the contacts oriented along the a^* axis, which are more easily prepared, the electric-field distribution near the point contact is strongly modified because of this very large anisotropy.

The $NbSe_3$ samples used in the present experiment had sizes along the *b* axis of $L_b \approx 3-4$ mm, along the *c* axis $L_c \approx 10 \div 20$ μm, and along the a^* axis $L_a \approx 1$ μm. Only single crystals with native-grown boundaries perpendicular to the *b* axis were selected.

To form a point contact the tip we used was a thin gold strip with a width of 50 μm and a thickness of 4 μm. The electrical contact of the sample with the normal-metal counterelectrode was formed at $T=4.2$ K with the use of a precision mechanical motion transfer system. The measurements of the *IV* characteristics and their first derivative $R_d = dV/dI$ have been carried out with the standard modulation technique. The point-contact *IV* characteristics were measured with three orientations of the magnetic field: parallel to the *b*, *c*, and a^* axes.

The *IV* characteristics for all the investigating contacts in zero magnetic field show the clear energy-gap structure, as illustrated by the lower curves shown in Figs. 1–3. At high voltages ($V>100$ mV) the increment of R_d is proportional to the square of the bias voltage, typical for Joule heating of metal-metal contacts (dotted lines in Fig. 1). This fact demonstrates that investigated contacts are of a direct type (without an insulating barrier). At $|V|<100$ mV the curves exhibit two steps in the differential resistance, associated with the excess resistance arising from the reflection of injected quasiparticles from the normal metal on the Peierls energy-gap barrier. The first step of the excess resistance corresponding to the carrier reflection on the first Peierls gap, Δ_{p1}, appears

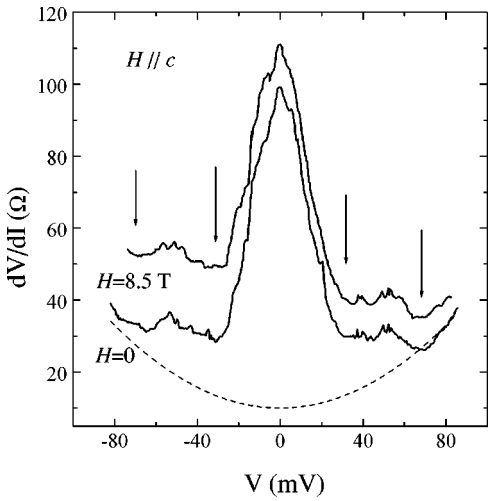

FIG. 1. Differential resistance $R_d(V)=dV/dI$ for a point contact Au-$NbSe_3$ oriented along the *b* axis and *H* parallel to the *c* axis. The lower curve: $H=0$; the upper curve: $H=8.5$ T. The voltages corresponding to the energy-gap positions are indicated by arrows. The dotted line is the normal-state background curve. The differential resistance scale corresponds to the lower curve; the upper curve is offset for clarity.

at $V=70-80$ mV. The second step is connected with the reflection of injected carriers on the second Peierls gap, Δ_{p2}, at $V=27-32$ mV. It can be seen from the figures that R_d decreases rapidly as voltage increases from zero. Such a shape of the excess resistance maximum is attributed to the two-dimensional character of the electronic spectrum,[17,24,25] and is in qualitative agreement with Huang and Maki's calculations[26] of the density of states in $NbSe_3$.

The results of the application of the magnetic field along different crystallographic orientations are shown in the upper curves in Figs. 1–3 for the same contacts. It is seen that for

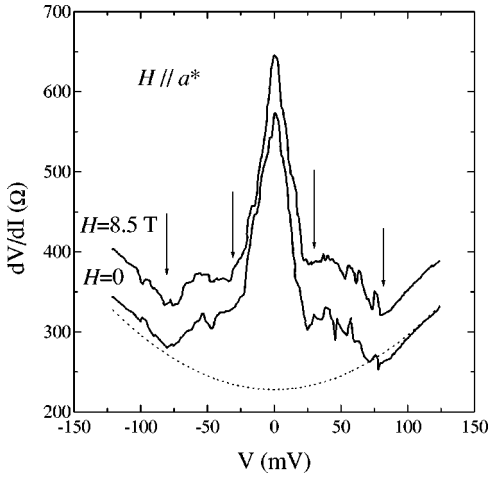

FIG. 2. $R_d(V)$ for a point contact Au-$NbSe_3$ oriented along the *b* axis and *H* parallel to the a^* axis. The lower curve: $H=0$; the upper curve: $H=8.5$ T. The voltages corresponding to the energy-gap positions are indicated by arrows. The dotted line is the normal-state background curve. The differential resistance scale corresponds to the lower curve; the upper curve is offset for clarity.

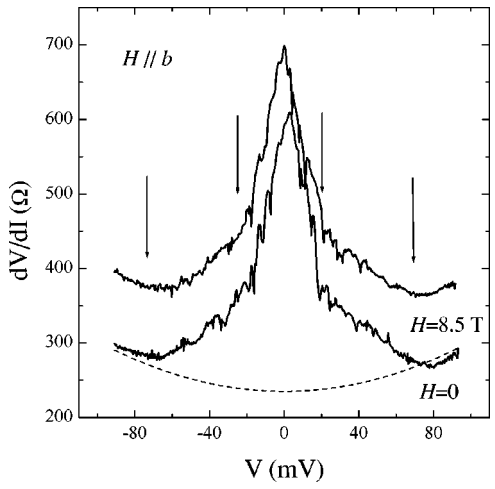

FIG. 3. $R_d(V)$ for a point contact Au-$NbSe_3$ oriented along the b axis and H parallel to the b axis. The lower curve: $H=0$; the upper curve: $H=8.5$ T. The voltages corresponding to the energy-gap positions are indicated by arrows. The dotted line is the normal-state background curve. The differential resistance scale corresponds to the lower curve; the upper curve is offset for clarity.

all investigated contacts there no detectable effect of the magnetic field up to $H=8.5$ T whatever the orientation H. Indeed, the amplitude of the excess resistance at zero bias as well as the position of the Peierls energy-gap singularities remain unchanged. The effect of application of the magnetic field is observed only for H oriented along the c axis with the appearance of an asymmetry of the IV curve (Fig. 1); the asymmetry results from the Hall electric field, which is largest for this direction of H. Indeed, the Hall electric field is directed in this case along the a^* axis for which the sample size is smallest. This electric field is transverse to the contact orientation. So, it modifies the point-contact electric field and leads to a change in the effective point-contact diameter, which depends on the electric-field distribution near the point contact.

According to Ref. 8, when perfect nesting is no longer present, the CDW induces a direct gap of value $2\Delta_p$ at the Fermi level, but the spectrum now has a smaller indirect gap of $2\Delta_g$. Depending on the degree of the ''lack of nesting,'' the system in the presence of a CDW may be a semiconductor ($\Delta_g \neq 0$) or a semimetal ($\Delta_g = 0$). The model[8] predicts that when a high magnetic field is applied, the semimetal-semiconductor transition occurs in $NbSe_3$. It means that the magnetic field induces a gap which increases rapidly to its maximum value $\Delta_g = \Delta_p$. In this case, the appearance of a new energy-gap singularity in the point-contact spectra should be observed in the magnetic field. But we do not observe this effect in our experiments nor any change in the position of the CDW energy-gap singularities when a magnetic field is applied.

Only two parameters of pocket carriers may be affected by the magnetic field, namely, the number of noncondensed normal carriers and their mobility. In the ballistic regime the differential resistance of point contacts at zero bias is given by the well-known Sharvin formula[27]

$$R_d(0) = \frac{1}{2N(0)e^2 v_F(\pi/4)d^2}, \tag{1}$$

where $N(0)$ is the density of states on the Fermi level and d is the contact diameter. Evidently, $R_d(0)$ depends only on the density of states and is independent of the relaxation time. Let us show that our point contacts satisfy the ballistic regime: the point-contact diameter d is less then the mean free path l. Using data for the mobility of the pocket electrons at low temperature $\mu = 3\times10^5$ cm^2/V s (Ref. 28), for $v_F = 1.4\times10^7$ cm/s (Ref. 29) and for $m^* = 0.24m_e$ (Ref. 3), we estimate $l \approx 6\times10^{-4}$ cm. This is larger than the maximum possible size of the contact in the geometry we used. So, we can conclude that the investigated point contacts are always in the ballistic regime. In $NbSe_3$, in the Peierls state, there are no carriers on the Fermi level except those of pocket electrons. So, in this case, $N(0)$ describes the density of states of uncondensed carriers to CDW carriers only. So, if the Balseiro-Falicov model is valid, the evolution of the differential point-contact resistance in magnetic field should be the same as the magnetoresistance in transport measurements. In our experiments the magnetoresistance of the point contacts is absent, while in the transport measurements it achieves 400% in transverse magnetic field of $H=7$ T at $T\sim4.2$ K (Ref. 3). Taking into account the noise level of our point-contact spectra at zero bias ($\sim5\%$), we can estimate that the changing in the density of states of uncondensed to CDW carriers at $H=8.5$ T (if this effect indeed exists) is no more than 5×10^{16} cm^{-3}, that is, more than an order-of-magnitude less than what must follow from the Balseiro-Falicov model and the experimental results of Ref. 3.

In summary, we have shown that an applied transverse magnetic field does not significantly affect the density of states of noncondensed normal carriers and the CDW energy gaps in $NbSe_3$ of low temperatures. So, the anomalously large magnetoresistance observed in this quasi-one-dimensional conductor below the second Peierls transition temperature most probably is the result of the increase of scattering induced by the magnetic field.

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