Anomalous asymmetry of magnetoresistance in NbSe$_3$ single crystals

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A pronounced asymmetry of magnetoresistance with respect to the magnetic field direction is observed for NbSe$_3$ crystals placed in a magnetic field perpendicular to their conducting planes. It is shown that the effect persists in a wide temperature range and manifests itself starting from a certain magnetic induction value $B_0$, which at $T = 4.2$ K corresponds to the transition to the quantum limit, i.e., the state where the Landay level splitting exceeds the temperature.

PACS numbers: 71.45.Lr, 73.40.Ns, 74.80 Fp

The NbSe$_3$ material is one of the most popular quasi-one-dimensional conductors with charge density waves (CDWs). The crystal lattice of NbSe$_3$ is monoclinic with the $b$ axis being parallel to the CDW chains and corresponding to maximum conductivity. The anisotropy of conductivity in the $(b - c)$ plane is $\sigma_b/\sigma_c \sim 10$, whereas the conductivity ratio $\sigma_b/\sigma_{c//}$ reaches a value of $\sim 10^4$ at low temperatures. The material experiences two Peierls transitions at the temperatures $T_{p1} = 145$ K and $T_{p2} = 59$ K, below which the spectrum of single-particle excitations develops energy gaps $\Delta_{p1}$ and $\Delta_{p2}$ at the Fermi level. However, the electron spectrum does not become completely dielectric. As a result of the incomplete nesting, normal carriers, i.e., electrons and holes, are retained in small pockets formed at the Fermi level. The shape of the pockets was determined from the angular dependences of Shubnikov-Haas oscillations. According to the data reported in Refs. 1-3, the Fermi surface areas that are not covered by the energy gap are shaped as ellipsoids with a maximum axial ratio of $8 - 10$ and with the major axes being parallel to the $c$ axis of the crystal. The concentration of both types of carriers is $n \sim 10^{18} \text{cm}^{-3}$, their mobility at low temperature is $\mu \sim 10^4 \text{cm}^2/\text{V} \cdot \text{s}$ (Ref. 4), and the effective mass is $m^* \sim 10^{-1}m_e$ (Ref. 5). Many of the experimental data can be adequately explained under the assumption that, in NbSe$_3$ at low temperatures, the two-dimensional nature of the electron spectrum is realized. In view of the aforementioned characteristics of the material, this suggests that the state of the carriers should be close to that of a 2D electron gas. The metal properties of NbSe$_3$ are retained down to the lowest temperatures. Studies of this compound with an immobile CDW revealed some unusual features of the transport properties due to the carriers not condensed into the CDW. Primarily, these properties include the effect of internal correlated interlayer tunneling and the presence of localized states within the Peierls energy gap.

Magnetotransport properties of this material also exhibit unusual behavior. When magnetic field is perpendicular to the $b$ axis of the crystal, the magnetoresistance of NbSe$_3$ first rapidly increases with magnetic field, then, at a certain field, its growth becomes much slower. However, the resistance is not saturated and, in high magnetic fields, the nonoscillating component of magnetoresistance linearly varies with magnetic field. In weak magnetic fields, a quantum size effect is observed for magnetic field orientations along the conducting layers (in the $(bc)$ plane of the crystal). Note that the effects described above are caused by the carriers that are not condensed into the CDW. Any direct effect of magnetic field on the properties of CDWs, including the Peierls transition temperature, has never been observed.

In this paper, we report on the unusual behavior of magnetoresistance of NbSe$_3$ in a magnetic field whose orientation is perpendicular to the conducting $(b - c)$ planes.

For our study, we used high-quality NbSe$_3$ single crystals with the ratio $R(300 K)/R(4.2 K) > 50$. The resistance was measured by the standard four-terminal method with a current flowing along the chains (along the $b$ axis); the current was from 1 to 100 $\mu$A, depending on the cross-sectional area of the sample, and, in all of the cases, it was $2 - 3$ orders of magnitude smaller than the current corresponding to the onset of the CDW slip. The magnetic field with an induction up to 9 T was generated by a superconducting solenoid. The measurements were performed with the magnetic field orientation perpendicular to the $(bc)$ plane of the crystal, while the sample could be rotated about the $c$ axis. The temperature range of measurements was $4.2 \div 60$ K.

Figure 1 shows the normalized resistance $\delta R = R(B)/R(0) - 1$ versus the magnetic field $B$ oriented along the $a^*$ axis for four different single crystals at $T = 4.2$ K. Qualitatively, the behavior of magnetoresistance is the same for all of the samples. In weak magnetic fields, the magnetoresistance is symmetric with respect to the direction of magnetic field, i.e., $R(B) = R(-B)$, and obeys the classical dependence $R \propto B^2$. In the fields from 0.2 to 1 T, the $R(B)$ dependence changes fundamentally: from quadratic in low magnetic fields to linear high magnetic fields. Precisely in this field interval, starting from a certain magnetic induction value $B_0$, the field reversal symmetry of the $\delta R(B)$ dependences fails. The asymmetry that appears in the $\delta R(B)$ dependences is not affected by changes in the direction and magnitude of the transport current and is only determined by the relative orientation.
FIG. 1: Normalized resistance $\delta R = R(B)/R(0) - 1$ versus magnetic field for four different NbSe$_3$ single crystals; $B \parallel a^\ast$.

FIG. 2: Angular dependence of magnetoresistance for a NbSe$_3$ single crystal (sample no. 10) rotated about the $c$ axis at $T = 4.2$ K in magnetic fields $B = 8.6$ and 1.7 T (the upper and lower curves, respectively). The angles $\theta = 90^\circ$ and $270^\circ$ correspond to the magnetic field orientation parallel to the $a^\ast$ axis.

FIG. 3: Angular dependence of the magnetoresistance difference $\Delta R = R(B) - R(-B)$ for a NbSe$_3$ single crystal (sample no. 10) at $T = 4.2$ K in magnetic field $B = 8.6$ T. The dashed curve represents the function $\Delta R_{\text{max}} \sin \theta$.

FIG. 4: Magnetoresistance of sample no. 13 at different temperatures; $B \parallel a^\ast$.

the angle between the $a$ and $c$ axes in the monoclinic crystal structure of NbSe$_3$. Note that, to make our analysis correct, we present only the results obtained with the samples that exhibited the aforementioned feature, although the asymmetry under discussion was observed by us in all other cases as well.

Figure 3 displays the dependence of the difference $\Delta R = R(B) - R(-B)$ on the angle $\theta$ at $B = 8.6$ T. The experimental dependence is adequately described by the function $\Delta R_{\text{max}} \sin \theta$ (the dashed curve in Fig. 3). This means that only the presence of the field component parallel to the $a^\ast$ axis gives rise to the asymmetry of

of the crystal and the magnetic field. This is illustrated by Fig. 2, which shows the angular dependence of magnetoresistance obtained by rotating the sample about the $c$ axis for two values of magnetic field: $B = 1.7$ and 8.6 T. The presence of characteristic local maxima of magnetoresistance at the angles $\theta = 109^\circ$ and $\pi - 109^\circ$ testifies to the fact that the sample under study truly is a single crystal, because the angle $\theta = 109^\circ$ corresponds to
magnetoresistance. For magnetic field orientations along the c and b axes, the effect is completely absent.

The effect is also independent of history; i.e., it does not depend on the direction of the field applied immediately after cooling the sample to the low temperature.

Note that the presence of a similar asymmetry of magnetoresistance in the given geometry of the experiment can be found in other publications, for example, in Ref. [3], where an obviously asymmetric angular dependence of magnetoresistance is presented for NbSe$_3$ in magnetic field $B = 1.5$ T rotating about the c axis of the crystal. The evolution of the $R(B)$ curves with temperature is shown in Fig. 4. At a first glance, it may seem that, as the temperature grows, the asymmetry of magnetoresistance decreases with the variation of the magnetic field direction. However, one can see that the magnetic induction $B_0$ corresponding to the appearance of asymmetry of magnetoresistance increases with temperature. The behavior of this parameter as a function of temperature is shown in Fig. 5. Let us normalize the resistance by the its value $R(0)$ at $B = 0$ and normalize the magnetic induction by $B_0$. As a result, we obtain a universal dependence shown in the inset in Fig. 5. Thus, as the temperature increases, the effect persists, and, at high temperatures, the asymmetry possibly arises beyond the field interval under study. As one can see from Fig. 5, at all the temperatures, the value of $B_0$ falls within the region of the qualitative change in the behavior of the $R(B)$ dependence (deviation from quadratic dependence). At $T = 4.2$ K, the value $B_0 = 0.2$ T is very close to the magnetic field at which the Landau level splitting becomes equal to temperature: $B_0 = 0.3$ T. This indicates a possible quantum nature of the phenomenon under study.

Let us introduce a parameter to characterize the quantitative variation of the effect. For this purpose, we use the magnetic field dependence of the parameter:

$$ r(B) = \frac{|R(+) - R(-)|}{R(0)} $$

As one can see from Fig. 6, which shows the behavior of this parameter for several samples, the function $r(B)$ is linear to a good accuracy in high magnetic fields. Hence, as a quantitative measure of the asymmetry under observation, it is reasonable to choose the slope, $\kappa$, of this linear dependence. We revealed no correlation of the asymmetry with the thickness or width of the crystals under investigation. However, from the inset in Fig. 6, one can see that the parameter $\kappa$ monotonically increases with increasing crystal volume enclosed between the potential contacts, which testifies to the bulk nature of the effect.

An adequate explanation of the phenomenon described in this paper is yet to be found. Formally, the behavior of magnetoresistance observed in our NbSe$_3$ samples means violation of the time reversal invariance, which is impossible. In the quantum limit, such an effect could be expected in the case of a spatially inhomogeneous distribution of magnetic field formed in the sample in the presence of local magnetic moments that may be caused by, e.g., magnetic impurities. However, according to the data of magnetic susceptibility measurements$^{22}$, such impurities are absent in NbSe$_3$. We measured the magnetic properties of NbSe$_3$ with a high-sensitivity SQUID magnetometer in the temperature range $4.2 \div 300$ K. The
data of this experiment will be published in a separate paper. Here, we only note that these measurements also revealed no traces of magnetic impurities in the NbSe$_3$ single crystals.

Another possible origin of a spatially inhomogeneous distribution of magnetic field may be the formation of toroidal magnetic moments $\mathbf{T}(\mathbf{r})$ in NbSe$_3$ crystals.\footnote{Yu. I. Latyshev, P. Monceau, O. Laborde, B. Pennetier, V. Pavlenko, T. Yamashita, J. Phys. IV France 9, Pr10-165 (1999).} For the case under consideration, it is important that $\mathbf{T}(\mathbf{r})$ is a polar vector, which changes sign under time reversal. The presence of toroidal moments is allowed for 31 magnetic symmetry classes.\footnote{Yu. I. Latyshev, P. Monceau, A. A. Sinchenko, L. N. Bulaevskii, et al., Pisma Zh. ksp. Teor. Fiz. 75, 103 (2002) [JETP Lett. 75, 93 (2002)].} However, NbSe$_3$ does not belong to these kinds of magnets.

Possibly, a certain role is played by the fact that the system is in the state with a CDW. If we consider CDW as the result of the singlet pairing of electrons and holes, the CDW should possess no magnetic properties. However, near the inhomogeneities of the CDW, charge and spin density oscillations may arise, which may give rise to local magnetic moments.\footnote{Yu. I. Latyshev, P. Monceau, S. A. Brazovskii, A.P. Orlov and T. Fournier, Phys. Rev. Lett. 95, 266402 (2005).} To determine the physical mechanism of the phenomenon observed in our experiments, further experimental and theoretical studies are necessary.

We are grateful to S.A. Brazovski and V.F. Gantmakher for useful discussions and to A.V. Kuznetsov for magnetic susceptibility measurements. This work was supported by the Russian Foundation for Basic Research (project nos. 05-02-17578 and 03-02-22001 CNRS) and the INTAS (grant no. 05-7972).

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