

# Coherent Tunneling between Elementary Conducting Layers in the NbSe<sub>3</sub> Charge-Density-Wave Conductor

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Characteristic features of transverse transport along the  $a^*$  axis in the NbSe<sub>3</sub> charge-density-wave conductor are studied. At low temperatures, the  $I$ - $V$  characteristics of both layered structures and NbSe<sub>3</sub>-NbSe<sub>3</sub> point contacts exhibit a strong peak of dynamic conductivity at zero bias voltage. In addition, the  $I$ - $V$  characteristics of layered structures exhibit a series of peaks that occur at voltages equal to multiples of the double Peierls gap. The conductivity behavior observed in the experiment resembles that reported for the interlayer tunneling in Bi-2212 high- $T_c$  superconductors. The conductivity peak at zero bias is explained using the model of almost coherent interlayer tunneling of the charge carriers that are not condensed in the charge density wave. © 2002 MAIK "Nauka/Interperiodica".

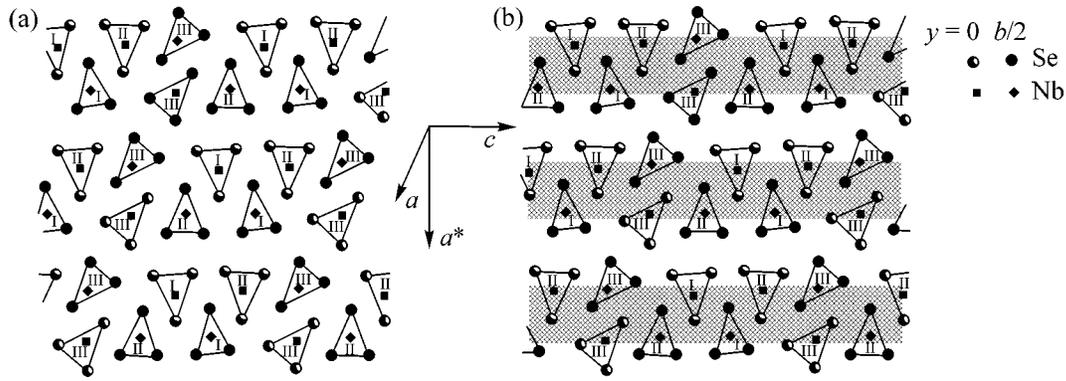
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It is well known that the crystal structure of BSCCO high- $T_c$  superconductors consists of atomically thin superconducting cuprate layers spatially separated by atomically thin insulating layers of BiO and SrO. The corresponding spatial modulation of the superconducting order parameter in the direction across the layers (along the  $c$  axis) leads to a discrete description of the transverse transport (the Lawrence–Doniach model [1]) with the neighboring superconducting layers being coupled through Josephson tunneling junctions. The validity of this approach is confirmed by the results of the experiments on natural Bi-2212 layered structures of small lateral size [2, 3]. Currently, the study of the interlayer tunneling of Cooper pairs and quasiparticles [4, 5] is one of the new original methods for investigating high- $T_c$  superconductors.

In this paper, we study the interlayer tunneling in a layered system with an electron condensate of a different type, namely, in a charge-density-wave (CDW) conductor. The material chosen for our experiments is NbSe<sub>3</sub>. This compound is characterized by two Peierls transitions, which occur at the temperatures  $T_{p1} = 145$  K and  $T_{p2} = 59$  K. In the low-temperature Peierls state, the Fermi surface retains some regions where the nesting conditions are not satisfied (the "pockets") and, hence, the Peierls gap is absent. Therefore, NbSe<sub>3</sub> does not undergo transition to the insulating state and retains its metallic properties down to the lowest temperatures [6]. From the analysis of both the crystal structure of

NbSe<sub>3</sub> and the anisotropy of its properties, it follows that this material can be classed with quasi-two-dimensional layered compounds. In fact, its conductivity anisotropy in the ( $b$ - $c$ ) plane is determined by the chain conductivity along the  $b$  axis and is estimated as  $\sigma_b/\sigma_c \sim 10$ , whereas the conductivity ratio  $\sigma_b/\sigma_{a^*}$  is determined by the layered character of the structure and reaches the values  $\sim 10^4$  at low temperatures [7, 8]. Figure 1a shows the crystal structure of NbSe<sub>3</sub> in the ( $a$ - $c$ ) plane. One can see that, in this material, the layered structure is formed as a result of a pairwise coordination of selenium prisms with the predominant orientation of their bases in the ( $b$ - $c$ ) plane. In Fig. 1b, the shaded areas indicate the elementary conducting layers in which the prisms are rotated and shifted with their edges toward each other. In these layers, the distances between the niobium chains are relatively small, whereas the neighboring conducting layers are separated by an insulating layer formed as a double barrier by the bases of the selenium prisms.

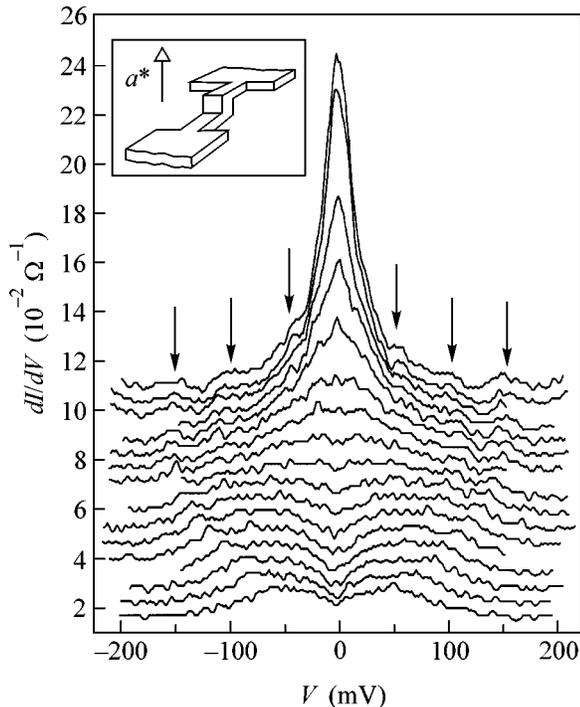
This type of layered structure in combination with the conductivity anisotropy offers the possibility for the CDW condensation in the elementary conducting layers spatially separated by atomically thin insulating layers. In this case, as in layered high- $T_c$  superconductors, one can expect that the CDW order parameter will be modulated along the  $a^*$  axis, and the transport across the layers will be determined by the intrinsic interlayer



**Fig. 1.** (a) Schematic representation of the  $\text{NbSe}_3$  structure in the  $(a-c)$  plane; (b) the same structure with the conducting planes indicated by shading.

tunneling between the elementary layers with the CDW.

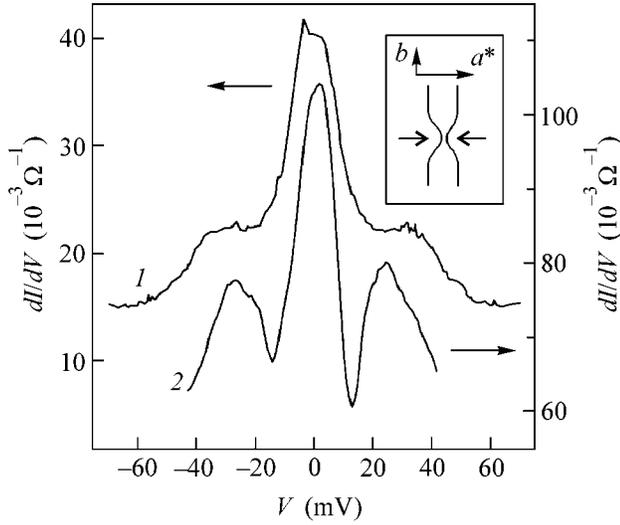
To verify these speculations, we carried out an experimental study of the transport across the layers in  $\text{NbSe}_3$  in the condensed CDW state.



**Fig. 2.** Dependences of  $dI/dV$  on the voltage  $V$  for an overlap junction (sample No. 1) at different temperatures: 59.5, 55.6, 52.1, 48.0, 43.7, 40.1, 37.0, 34.3, 31.0, 28.0, 25.1, 22.6, 19.1, 15.7, 12.8, 8.0, and 4.2 K. The dynamic conductivity scale corresponds to the curve at  $T = 59.5$  K, and other curves are shifted upwards for clarity. The inset shows the sample configuration.

The samples used in the experiment were layered structures with a small area through which the current flows across the layers,  $S = 2 \times 2 \mu\text{m}$ , and with the number of elementary layers  $\sim 30$  (the overlap junctions). They were made from a thin  $\text{NbSe}_3$  single crystal by the focused ion beam processing technique developed for fabricating similar structures from Bi-2212 [9]. Schematically, the structure under investigation is shown in the inset of Fig. 2. In addition, we studied the characteristics of  $\text{NbSe}_3$ - $\text{NbSe}_3$  point contacts oriented in the direction of the  $a^*$  axis. The configuration of such a contact is shown in the inset of Fig. 3. The contact was formed directly at low temperature by bringing two  $\text{NbSe}_3$  whiskers together with high accuracy. In both cases, we performed four-terminal measurements of the  $I$ - $V$  characteristics and their derivatives at low temperatures, i.e., below the second Peierls transition  $T_{p2} = 59$  K.

The main results of the experiment are shown in Fig. 2, which, for one of the virtually perfect structures (no. 3), displays the dynamic conductivity along the  $a^*$  axis versus the bias voltage  $V$  at different temperatures from 59.5 to 4.2 K. It can be seen that, for  $T < T_{p2}$ , the differential  $I$ - $V$  characteristics are of a tunneling character. When the temperature decreases below  $\sim 35$  K, the  $I$ - $V$  characteristics begin to exhibit a conductivity peak at zero bias voltage. As the temperature decreases further, this peak becomes a dominant feature. The peak amplitude reaches saturation when the temperature decreases below 6–8 K, and at  $T = 4.2$  K, it is almost 20 times as great as the conductivity observed at high bias voltages. Note that this anomaly cannot be attributed to Joule heating. The estimate of the sample heating for a typical value of heat transfer to helium yields a conductivity decrease of less than 10% for the whole range of measured voltages. In addition, the  $I$ - $V$  characteristics show a clearly defined set of conductivity peaks that are symmetric about zero voltage and at low temperature correspond to  $|V| = 50, 100,$  and  $150$  mV, i.e., to  $|V| = nV_0$ , where  $V_0 = 50$  mV and  $n = 1, 2, 3$ . As the temperature increases above 25 K, these



**Fig. 3.** Dependences  $dI/dV(V)$  (1) for a  $\text{NbSe}_3$ - $\text{NbSe}_3$  point contact and (2) for an overlap junction (sample No. 2) at  $T = 4.2$  K. The inset shows the point contact configuration.

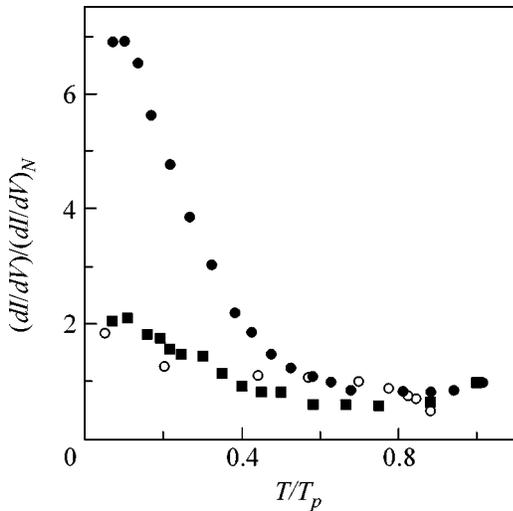
peaks move to lower energies, and, at the temperature  $T = 59$  K corresponding to the second Peierls transition for  $\text{NbSe}_3$ , the values of  $V_n$  vanish.

The picture described above was observed only for perfect structures. The presence of structure defects, such as a twin boundary, leads to a considerable reduction of the conductivity peak at zero bias and to the appearance of a peak at  $V = 25\text{--}27$  mV  $\approx V_0/2$  (curve 2

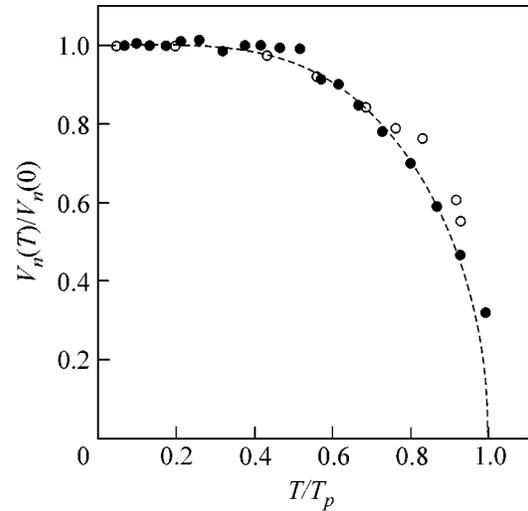
in Fig. 3). Qualitatively similar dependences were observed for some of the  $\text{NbSe}_3$ - $\text{NbSe}_3$  point contacts. Their  $I$ - $V$  characteristics also exhibit a conductivity peak at zero bias with an amplitude approximately equal to that observed for defect overlap junctions, as well as a conductivity peak at  $V \approx 25$  mV (curve 1 in Fig. 3). Figure 4 presents the temperature dependences of the normalized dynamic conductivity at zero bias voltage for all types of samples studied in the experiment. One can see that, for a perfect overlap junction, the amplitude of the zero bias anomaly is more than three times greater than the amplitudes observed for the defect layered structures and point contacts. Note that the voltage value  $V_0 = 50$  mV is close to twice the value of the low-temperature energy gap,  $2\Delta_{p2}/e$ , for  $\text{NbSe}_3$  [10, 11], while the dependence  $V_n(T)/V_n(0)$  obtained for both overlap junctions and point contacts (Fig. 5) agrees well with the temperature dependence of the energy gap predicted by the BCS theory (the dashed line in Fig. 5). This result suggests that the conductivity features observed in the experiment are governed by a gap mechanism.

It is important to note that all effects described above were observed exclusively for the transport across the layers (along the  $a^*$  axis). The extra experiments performed by us on specially fabricated bridges and point contacts oriented along the  $c$  axis (with the transport across the chains in the layer plane) showed no conductivity peak at  $V = 0$ .

Let us first analyze the results obtained for perfect overlap junctions. The most prominent feature of the  $I$ - $V$  characteristics of these structures is the strong con-



**Fig. 4.** Temperature dependences of the dynamic conductivity at zero bias voltage for two overlap junctions: sample No. 3 (full circles) and sample No. 1 (full squares), and for a  $\text{NbSe}_3$ - $\text{NbSe}_3$  point contact (empty circles). The conductivity values are normalized to the value of  $dI/dV$  ( $T = 62$  K).



**Fig. 5.** Temperature dependence of the second Peierls energy gap for an overlap junction (sample No. 3, full circles) and a  $\text{NbSe}_3$ - $\text{NbSe}_3$  point contact (empty circles).

ductivity peak at zero bias voltage. In addition, the characteristics exhibit a periodic sequence of peaks at  $|V| = nV_0$ , which resembles the series of quasiparticle branches observed in the  $I$ - $V$  characteristics of Bi-2212 layered structures when the measurements are performed across the layers [3]. As was noted above, the value  $V_0 = 50$  mV is close to twice the value of the low-temperature CDW gap in NbSe<sub>3</sub>, and the temperature dependence  $V_0(T)$  follows the prediction of the BCS theory. However, in contrast to the case of a superconductor, the conductivity observed for NbSe<sub>3</sub> at zero bias voltage is finite. This fact suggests that the interlayer conduction at zero bias does not originate from the collective CDW contribution of the Josephson tunneling type.

The conductivity peak observed at zero bias cannot be explained by the regular (incoherent) single-particle tunneling as well. If this were the case, the conductivity peak could be attributed to the energy dependence of the density of states of electrons that are not condensed in the CDW. Then, the conductivity feature under discussion should also be observed for man-made N-I-CDW tunnel junctions. However, no such effects were revealed by the detailed experimental studies of this kind of tunnel junctions oriented along the  $a^*$  axis [10, 11].

We believe that the conductivity peak at zero bias is caused by the coherent interlayer tunneling of the charge carriers localized in the pockets of the Fermi surface, and below we thoroughly substantiate this statement.

In the general case, the tunnel current between two nonsuperconducting layers,  $a$  and  $b$ , is described by the expression [12]

$$I = \frac{4\pi es}{\hbar} \sum_{\mathbf{p}, \mathbf{q}} |t_{ab}(\mathbf{p}, \mathbf{q})|^2 \times \int_{-\infty}^{+\infty} d\omega \left( \tanh \frac{\omega}{2T} - \tanh \frac{\omega - eV}{2T} \right) \times \text{Im}G_a^R(\mathbf{p}, \omega) \text{Im}G_b^R(\mathbf{q}, \omega - eV), \quad (1)$$

where  $G^R$  is the retarded Green function of an electron with the momentum  $\mathbf{p}$  in the layer  $a$ ,  $\text{Im}G^R(\mathbf{p}, \omega)$  is the spectral density of the Green function,  $t_{ab}(\mathbf{p}, \mathbf{q})$  is the matrix element characterizing the tunneling from the state with the momentum  $\mathbf{p}$  in the layer  $a$  to the state with the momentum  $\mathbf{q}$  in the layer  $b$ ,  $V$  is the voltage between the adjacent layers, and  $T$  is the temperature.

Note that the contribution of the collective (moving) CDW mode [13] to the interlayer tunneling is absent, because the tunnel current is determined by the Green functions in different layers, whereas the collective mode propagates within the layers.

Below, we consider the coherent tunneling, when the electron momentum in the plane does not change under the tunneling:  $\mathbf{p} = \mathbf{q}$ . Then, we have

$$I = \frac{es}{\pi\hbar} \int d\mathbf{p} |t_{ab}(\mathbf{p})|^2 \times \int_{-\infty}^{+\infty} d\omega \left( \tanh \frac{\omega}{2T} - \tanh \frac{\omega - eV}{2T} \right) \times \text{Im}G_a^R(\mathbf{p}, \omega) \text{Im}G_b^R(\mathbf{p}, \omega - eV). \quad (2)$$

Taking into account that the scattering in a layer is determined by the collision frequency  $\nu$ , we obtain

$$\text{Im}G^R(\mathbf{p}, \omega) = \frac{\gamma}{\pi(\omega - \epsilon(\mathbf{p})^2 + \gamma^2)}, \quad (3)$$

where  $\epsilon(\mathbf{p})$  is the electron spectrum and  $\gamma = \hbar\nu$ . We can use this expression with  $\gamma = \gamma_{sc} + \gamma_{inc}$ , which takes into account the change in the momentum because of the scattering within the layers and the change due to the tunneling  $\gamma_{inc} = \langle \epsilon(\mathbf{p}) - \epsilon(\mathbf{q}) \rangle$ . In both cases, we take into account the energy uncertainty for the state characterized by the momentum  $\mathbf{p}$ . We define the tunneling as almost coherent when  $\gamma_{inc}$  and  $\gamma$  are small relative to other energy parameters of the electron system. In our case, such parameters are the Peierls gap and the width of the electron band in the pockets. Replacing  $t_{ab}(\mathbf{p})$  by the momentum-independent quantity  $t$  and integrating with respect to  $\mathbf{p}$ , we obtain

$$\frac{1}{4\pi^2} \int d\mathbf{p} |t_{ab}(\mathbf{p})|^2 \text{Im}G_a^R(\mathbf{p}, \omega) \text{Im}G_b^R(\mathbf{p}, \omega - eV) = \frac{2\gamma N(0) |t|^2}{e^2 V^2 + 4\gamma^2}, \quad (4)$$

where  $N(0)$  is the density of states of electrons in the pockets. Finally, for the interlayer current when  $eV < 2\Delta$ , we obtain the expression

$$I(V) = \frac{N(0) |t|^2 \gamma e V}{2\pi^3 (e^2 V^2 + 4\gamma^2)}, \quad (5)$$

and the dynamic conductivity has the form

$$\frac{\sigma(V)}{\sigma(0)} = 4\gamma^2 \frac{4\gamma^2 - e^2 V^2}{(e^2 V^2 + 4\gamma^2)^2}. \quad (6)$$

Note that the temperature dependence is present in  $\gamma(T)$  only. One can see that the dynamic conductivity has a peak of width  $\approx \gamma$  at  $V = 0$  and becomes negative and unstable when  $eV > 2\gamma$ . For a thirty-layer structure, the experimentally observed peak width  $\approx 10$  mV corresponds to  $\gamma \approx 0.3$  meV. For comparison, we estimate the parameter  $\gamma_{sc}$  characterizing the scattering within the layers. Using the known mobility data  $\mu = e/\nu_{sc} m^* \approx 4 \times 10^4$  cm<sup>2</sup>/V s [7, 14], where  $m^* = 0.24m_e$  [15], we obtain  $\gamma_{sc} \approx 0.13$  meV; i.e., the changes in the electron

momentum because of tunneling are approximately equal to changes caused by the scattering within the layers.

Now, we explain why the interlayer current decreases with increasing voltage  $V$ . The electron tunneling between the layers must obey the energy conservation law; i.e.,  $\epsilon(\mathbf{p}) = \epsilon(\mathbf{q}) - eV$  to within  $2\gamma$ . For coherent tunneling, we have  $\epsilon(\mathbf{p}) = \epsilon(\mathbf{q})$ , and for  $eV \ll \gamma$ , tunneling is possible and we have the conventional Ohm's law. When  $eV > 2\gamma$ , tunneling is impossible up to the voltage  $V$  reaching  $2\Delta/e$ . In this case, electrons condensed in the CDW begin to contribute to the interlayer current in the form of a regular tunneling of CDW quasiparticles through the double Peierls gap  $2\Delta$ . Thus, the interlayer current can be realized by means of only one of the two aforementioned mechanisms. In an actual multilayer structure, because of the geometric nonuniformity of individual layers (the areas of the layers are somewhat different), the voltage value  $V \sim 2\gamma/e$  will not be reached simultaneously for different individual tunneling junctions. When  $V > 2\gamma/e$ , some of the junctions can be in the coherent tunneling regime whereas other junctions can be in the regime of single-particle tunneling through the gap. As a result, a sequence of conductivity peaks must appear in the  $I$ - $V$  characteristic of the compound under study at the voltages  $V = 2n\Delta_p/e$ , where  $n = 1, 2, \dots$ .

In the presence of defects in an overlap junction or in the case of a point contact, the incoherence of tunneling is enhanced, although in the best structures the tunneling remains almost coherent. Therefore, the  $I$ - $V$  characteristics of these structures, along with the conductivity peak at zero bias, contain an additional feature at  $V = \Delta_p/e$ , which is related to the single-particle tunneling of the N-I-CDW type.

Thus, the results of this study show that the interlayer tunneling in natural layered structures obtained from NbSe<sub>3</sub> represents an independent efficient method for investigating the Peierls state in this compound. The tunneling conductivity peak observed at zero voltage can be self-consistently explained by the almost coherent interlayer tunneling of charge carriers that are not condensed in the CDW and are localized in the Fermi surface pockets not covered by the gap. The series of equidistant peaks in the  $I$ - $V$  characteristic can be explained by the quasiparticle tunneling through the

CDW gap under sequential transitions of individual tunneling junctions to the resistive state. Note that the conductivity peak observed at zero bias is a unique manifestation of coherent single-particle transport in solids.

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