Coherent-noncoherent tunneling crossover in NbSe₃-NbSe₃ point contacts

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Current-voltage (IV) characteristics of NbSe₃-NbSe₃ artificial contacts (point contacts) formed along the a^* axis have been investigated in a wide range of contact resistances and temperatures. Depending on the contact resistance, two types of IV characteristics have been observed: the first type observed for low contact resistance demonstrates a conducting peak at zero bias, while the second one for large contact resistance is characterized by a large resistance maximum near the zero bias voltage. Below the second charge-density wave (CDW) transition ($T < 59$ K), CDW energy gap singularities were observed in the IV characteristics of both types of junctions at $V = \Delta_{p1}/e$ and $V = 2\Delta_{p2}/e$, while the singularity at $V = \Delta_{p2}/e$ corresponding to the single particle tunneling of carriers noncondensed into the low-temperature CDW is absent. The experimental results are discussed in the frame of the model of interlayer coherent tunneling of the remaining charge carriers that are not condensed into the CDW.

I. INTRODUCTION

Tunneling spectroscopy is one of the best tools for the determination of quasiparticle excitation spectra in metallic and semiconducting systems.[1,2] In the case of conventional superconducting materials, this method reveals the gap character of the electronic spectrum as well as the electron-phonon origin of the Cooper pairing. Practically, tunneling spectroscopy is realized using, for examples, planar (thin film) structures, point contacts, or a scanning tunnel microscope. However, in all these methods, the investigated tunnel junction is formed on the surface of the sample with the consequence that the results strongly depend on the surface quality of the material. Recently, a new method for tunneling spectroscopy has been proposed: This interlayer tunneling method has been used for studying anisotropic materials with a layered crystalline structure where the elementary conducting layers are separated by atomically thin insulating barriers. In these materials, the electrical transport along the conducting layers is provided by the relatively high intralayer conductivity, while the transport across the layers occurs via interlayer tunneling. The main advantage of this method compared with others is the possibility of investigating bulk properties of materials. At first, this technique has been applied for layered high-temperature cuprate superconductors.[3,4] Very rich information about the superconducting energy gap and pseudogap,[5] the symmetry of the superconducting order parameter,[6] the coherence of interlayer tunneling,[7] and others have been obtained using this method.

Recently, this technique has been applied to materials with another (nonsuperconducting) collective electronic ground state, namely, compounds with a charge-density wave (CDW) and specifically to NbSe₃, regarded as the typical example of quasi-one-dimensional (Q1D) systems.[8,9] This compound undergoes two CDW phases occurring at $T_{p1} = 144$ K and $T_{p2} = 59$ K. Below these temperatures, the successive Peierls energy gaps, Δ_{p1} and Δ_{p2}, are opened in the spectrum of the quasiparticle excitations. Distinctively

from most Q1D systems with a CDW, this compound does not undergo a transition into an insulating state but retains its metallic properties down to the lowest temperatures.[10] This results from the remaining normal carriers in a small region of the Fermi surface (pockets), where the nesting conditions are not satisfied, the Peierls energy gap being thus absent. Indeed, large Shubnikov–de Haas oscillations due to electron and hole pockets have been observed at temperatures well below T_{p2}.[11–15] Another characteristic feature of NbSe₃ is its very large conductivity anisotropy due to the layered character of the structure. Evaluated from the chain conductivity along the b axis, the conductivity anisotropy in the b-c plane is estimated as $\sigma_b/\sigma_c \sim 10$, whereas the conductivity ratio perpendicular to the b-c plane, σ_b/σ_{a^*}, may reach the value $\sim 10^4$ at low temperatures.[8,16]

Stacked mesa structures of NbSe₃ with micron in-plane sizes prepared by focused-ion beam[17] were studied in a series of works.[8,9,18–21] It was assumed that the CDW order parameter is modulated in the direction transversely to the layers (along the a^* axis). As a result, the transport in this direction is determined by the intrinsic interlayer tunneling between the elementary conducting layers. It was experimentally shown that this interlayer tunneling has two components: the coherent tunneling of pocket carriers and the ordinary tunneling of CDW quasiparticles through the double Peierls energy gap barrier. The indication of coherent tunneling comes from the anomalously large conductance peak at zero bias voltage (ZBCP) observed at low temperature below T_{p2} in the differential current-voltage characteristics of investigated junctions.[9,18,19] There are two explanations for this peculiarity: tunneling with conservation of the in-plane momentum[9] or tunneling considered as a ballistic transport.[22] In addition, the conductance peaks at $|V| = 2\Delta_{p2}/e$ expected for a single CDW-I-CDW (I stands for insulator) tunnel junction were observed in such mesa structures of NbSe₃. The ZBCP is suppressed when the temperature is increased or under the application of a high magnetic field. In this case, additional singularities of IV characteristics indicating the

existence of localized states inside the CDW energy gaps appear.[20,21] According to Ref. 9, the coherent tunneling of the pocket electrons takes place if the interlayer and in-plane scattering energy are small relative to other energy parameters of the electronic system.

The effect of intrinsic tunneling in NbSe$_3$ was also recently observed for another experimental geometry, in longitudinal nanoconstrictions with submicron scale.[23] In contrast to stacked junctions, only in-plane electrical transport occurs in such a geometry. It was shown that such a constriction behaves like a CDW-I-CDW tunnel junction, and the observed energy gap singularities correspond well with the full CDW gaps $2\Delta_{p1}$ and $2\Delta_{p2}$.

As a rule, a mesoscopic stacked junction contains several tens of the elementary tunneling junction formed by the layered crystalline structure of the material. For comparison, it is very interesting to investigate the properties of only a single interlayer connection. One can then expect to observe similar effects in artificial NbSe$_3$-I-NbSe$_3$ junctions as in stacked junctions.

Till now, only nonsymmetrical tunnel structures N-I-NbSe$_3$ (N stands for normal metal)[24–26] or direct point contacts N-NbSe$_3$ (Refs. 27 and 28) were investigated. Theoretical calculations[29] predict unusual *IV* characteristics of symmetrical tunnel junctions for CDW materials with a partly gapped electronic spectrum. The differential conductivity of such structures must have a root singularity at $eV=\Delta_p$, corresponding to tunneling of N-CDW type of the gapless carriers to the CDW, and a jump at $eV=2\Delta_p$, corresponding to CDW-I-CDW-type tunneling of quasiparticle excitations through the double Peierls energy gap. Investigation of NbSe$_3$-I-NbSe$_3$ junctions allows us to experimentally prove these predictions.

Another motivation for studying artificial NbSe$_3$-NbSe$_3$ junctions is the investigation of phenomena possibly caused by Joule self-heating. In a recent work,[30] it was assumed that the two exceptionally sharp gaplike features observed in Ref. 19 may not be necessarily related to CDW gaps but could originate from self-heating. It is well recognized, however, that self-heating should not play any role with point contacts in the ballistic regime, i.e., when the point-contact diameter d is less than the mean free path l. As previously estimated (Ref. 28), taking for mobility $\mu=10^6$ cm^2/V s (Ref. 31), for effective mass $m^*=0.24m_e$ (Ref. 11), and for Fermi velocity $v=1.4\times10^7$ cm/s (Ref. 10), one obtain the estimation $l\approx20$ μm in NbSe$_3$. One can expect that the ballistic transport condition will also be fulfilled for the NbSe$_3$-NbSe$_3$ point contacts oriented along the a^* direction with $d<20$ μm and that self-heating will be negligible.

In the present paper, we report the results of detailed investigations of NbSe$_3$-NbSe$_3$ point-contact characteristics, the resistance of which varying in the range $10-10^3$ Ω and at temperatures $T<65$ K.

II. EXPERIMENT

Only NbSe$_3$ single crystals with a perfect surface in the b-c plane were selected for the experiments. The typical sizes of the investigated wiskers were, along the b axes,
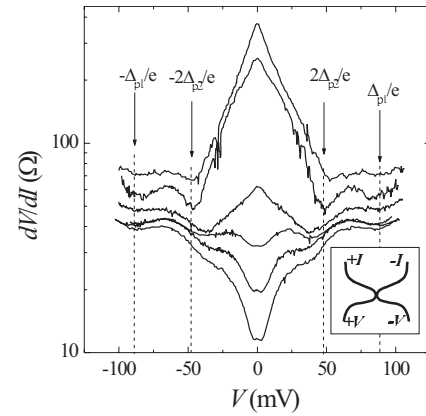

FIG. 1. Dependences of the differential resistance $R_d(V)$ (in a logarithmic scale) measured at $T=4.2$ K for several NbSe$_3$-NbSe$_3$ point contacts with different zero bias resistances R_{d0} (from 10 to 400 Ω), oriented along the a^*-axis direction. The voltages corresponding to the mean value of the energy gaps, Δ_{p1}/e $=88.9$ mV and $\Delta_{p2}/e=47.4$ mV, are indicated by arrows. Inset illustrates schematically the contact geometry.

$L_b\approx3-4$ mm, along the c axes, $L_c\approx20-30$ μm, and, along the a^* axes, $L_{a^*}\approx1-5$ μm. To form a NbSe$_3$-NbSe$_3$ contact along the a^* axis, two bent crystals with parallel b axis were brought together (see inset of Fig. 1), directly at low temperature, with the help of a precise mechanical motion transfer system. The maximal effective size of such type of contact was in the order of tens of microns. The measurements of the *IV* characteristics and of its first derivative R_d $=dV/dI$ of the prepared junctions were carried out with the standard modulation technique using a lock-in detector.

III. EXPERIMENTAL RESULTS

Depending on the value of the zero bias resistance R_{d0}, the observed NbSe$_3$-NbSe$_3$ contact characteristics demonstrate, on one hand, a qualitatively different behavior at low bias voltages and, on the other hand, nearly the same behavior at high voltages. The differential *IV* curves at $T=4.2$ K for different contacts with R_{d0} varying from 10 to 400 Ω are shown in Fig. 1. The curves with a low R_{d0} are characterized by a deep minimum of R_d at $V=0$ (ZBCP). When R_{d0} increases, the amplitude of ZBCP decreases and then a zero bias resistance peak (ZBRP) appears. The point contacts with $R_{d0}>100$ Ω exhibit the typical tunneling behavior with a large ZBRP and minima at $|V|=45-51$ mV, the mean value of which (47.4 mV), shown with a dashed line in Fig. 1, coincides with the $2\Delta_{p2}/e$ value known from former works.[24–28] At the same time, independent of the contact resistance, all the curves qualitatively show a similar behavior at $|V|>50$ mV, demonstrating a characteristic minimum at $|V|=80-92$ mV, the mean value of which (88.9 mV) is closely related to the value of the high-temperature CDW energy gap Δ_{p1}.[28] Note that, in spite of the significant difference in the resistance values at $V=0$ (3 orders of magnitude), the positions of the energy gap singularities appear within a relatively small voltage dispersion, demonstrating the good reproducibility of the experimental data.
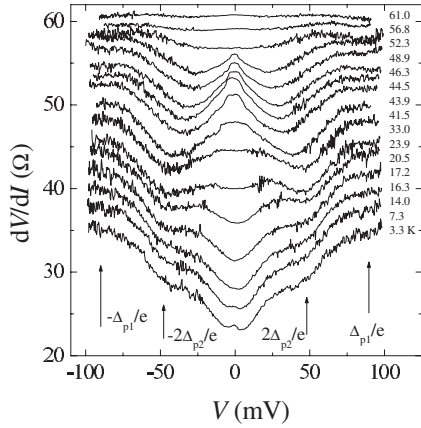
FIG. 2. Dependences of the differential resistance $R_d(V)$ as a function of temperature for a NbSe$_3$-NbSe$_3$ contact demonstrating a zero bias conductance peak (ZBCP). The curves are shifted for clarity and correspond, from the bottom to the top, to T =3.3–61 K. The arrows indicate the positions of mean value of the energy gaps.

FIG. 3. Dependences of the differential resistance $R_d(V)$ as a function of temperature for a NbSe$_3$-NbSe$_3$ contact with a zero bias resistance peak (ZBRP). The curves are shifted for clarity and correspond, from the top to the bottom, to T=7.5–62 K. The arrows indicate the positions of mean value of the energy gaps. Inset shows a typical $R_d(V)$ curve for a Au-NbSe$_3$ contact at T=4.2 K.

Studies of the temperature evolution of the *IV* characteristics were rather difficult because of the mechanical instability of contacts caused by many random factors during the change in temperature. According to Ref. 32, a real point contact consists of multiple microshorts, the main contribution to the point-contact resistance being provided by the biggest microshort. Thus, any change in the point-contact configuration leads to an increase (decrease) of the number of microshorts that, consequently, yields some sharp change in the point-contact resistance easily detectable in the *IV* curve. In some cases, we have successfully measured *IV* characteristics of a point contact without such mechanical instabilities; this indicates that the point-contact configuration was preserved in the whole temperature range from the liquid helium temperature up to above T_{p2}.

Contacts with high and low zero bias resistances have a different temperature evolution of the *IV* characteristics. *IV* curves measured in the temperature range from 3.3 to 60.0 K, for a contact revealing the ZBCP behavior (R_{d0}=22 Ω), are shown in Fig. 2. The amplitude of the ZBCP decreases monotonically with the increase of temperature and becomes nondetectable at $T \approx 30$ K. Above this temperature, the ZBRP appears and increases up to $T \approx 40$ K; after that, it starts to decrease and completely disappears at $T=T_{p2}$=59 K. It can also be noted that the minimum of R_d at $|V|$ close to 47.4 mV (see arrows in Fig. 2), corresponding to the mean value of $2\Delta_{p2}/e$, becomes well pronounced near 15 K; at higher temperatures, the voltage position of this singularity goes monotonically to zero when $T \to T_{p2}$. At T <15 K, the accurate determination of the second energy gap is difficult because the large ZBCP largely masks the gap singularity of the lowest CDW.

Figure 3 displays the dynamic resistance as a function of bias voltage at different temperatures for a contact revealing a zero bias resistance peak at low temperature. In contrast to the contact with ZBCP, R_{d0} decreases monotonically with the increase of temperature. The minimum at $|V|=2\Delta_{p2}/e$ mV is
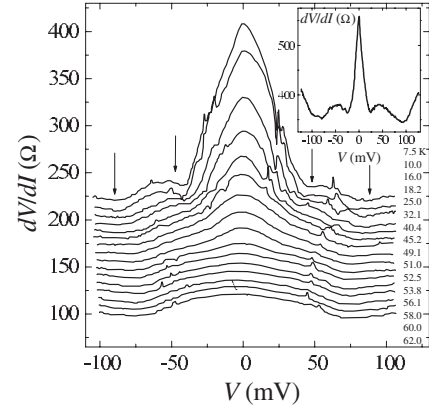
smeared out, but it can be seen that its voltage position goes to zero when $T \to T_{p2}$. At the same time, the voltage position of the second minimum of $R_d(V)$, corresponding to the energy gap of the high-temperature CDW, changes from 90 mV at T=7.5 K to 75 mV at T=62 K. For $0 < T$ $< 0.5T_{p1}$, a BCS-type variation would yield a nearly constant variation. The non-BCS behavior of the Peierls energy gap in NbSe$_3$ was also observed in optical investigation.[33]

The normalized temperature dependences of R_{d0} for both types of junctions are shown in Fig. 4 demonstrating a striking qualitative difference; while $R_{d0}(T)$ decreases monotonically for a contact demonstrating ZBCP, the junctions with ZBRP exhibit the opposite temperature variation. Such a behavior correlates well with the results obtained for stacked mesa junctions[19] and for longitudinal nanoconstrictions.[23] The amplitude of the ZBCP increases when the temperature
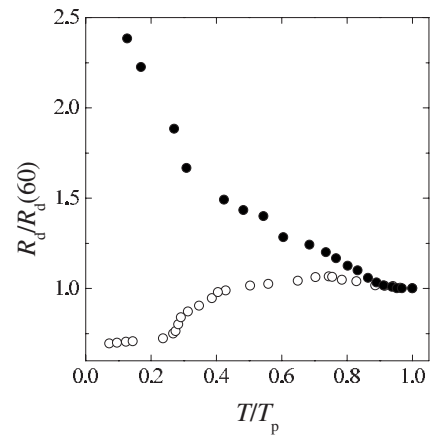

FIG. 4. Temperature dependences of the zero bias differential resistance R_{d0} normalized to the resistance at low CDW transition temperature T_{p2}=59 K for NbSe$_3$-NbSe$_3$ contacts revealing either a zero bias resistance peak (ZBRP, closed circles) or a zero bias conductance peak (ZBCP, open circles).

is decreased and saturates at $T < 10-12$ K; this agrees with the results obtained in Refs. 9 and 19. Note that, at the same temperature, the slope of the $R_{d_0}(T)$ curve for the ZBRP contact changes, and below this temperature, the amplitude of the ZBRP increases more rapidly.

IV. DISCUSSION

Comparing the point-contact spectra obtained in the present work with those of mesa-type junctions,[9,18,19] we can conclude that low resistance point contacts reveal features corresponding to coherent interlayer tunneling. Indeed, the characteristic features of such junctions are visible ZBCP observed at low temperatures, characteristic minima of $R_d(V)$ corresponding to the energy gap positions, and plateau of $R_d(V)$ near $V=0$ appearing at high temperatures (when the ZBCP is suppressed, see Fig. 2) ascribed as a threshold voltage V_t for tunneling[21] with V_t proportional to the CDW gap, such as $V_t \approx 0.2\Delta_p$. A theoretical model was proposed[21] which describes the phase decoupling between layers at V_t via the formation in the weakest junction of an array of dislocation lines (the CDW phase dislocation defects). V_t is only detectable when the ZBCP is suppressed. In can be seen from Fig. 2 that just the same behavior is observed in the present work.

Coherent tunneling involves tunneling with conservation of momentum.[9] Such type of tunneling is possible if the energy parameter for scattering γ is less than the Peierls energy gap and the bandwidth of the pocket electrons; $\gamma = \gamma_{sc} + \gamma_{inc}$, where $\gamma_{sc} = \hbar\nu$ is the scattering in a layer, ν is the collision frequency, and γ_{inc} determines the change in momentum due to tunneling. Coherent tunneling appears in the IV curves as a zero bias conductance peak with a width $\sim \gamma$. At $eV > 2\gamma$, tunneling is impossible up to the voltage $V = 2\Delta_p/e$. In this case, the carriers condensed into the CDW ground state begin to contribute to the interlayer current in the form of a conventional quasiparticle tunneling through the double CDW energy gap. For the stacked junctions investigated in Ref. 9, the estimation of the scattering parameter gives $\gamma \approx 0.3$ meV. In NbSe$_3$-NbSe$_3$ artificial contacts, interface scattering is obviously a random factor, and the γ_{inc} value is thus expected to be much larger than for intrinsic tunneling. The variation in the amplitude of the parameter γ can be attributed to an imperfect connection between the touching wiskers in the contact region such as space disorientation, impurities, and others. So, the different curves represented in Fig. 1 can be associated with contacts with a different parameter γ. The width of the ZBCP for better contacts, $\delta V \approx 5$ mV, is smaller than the single particle gap Δ_{p2}/e, and the conditions of coherent tunneling are thus satisfied. If the scattering due to tunneling for a given contact becomes larger, the contribution from the ordinary tunneling becomes larger too; as a result, a characteristic maximum of the dynamical resistance appears on the IV curves. So, the curves in Fig. 1 can be considered as demonstrating the crossover from coherent to noncoherent tunneling.

This crossover was also observed with increasing temperature. It is well known that the mobilities of the pocket electrons and holes decrease rapidly when the temperature is increased.[31] That means that γ_{sc} should increase, and then the ZBCP decreases. According to the results obtained in Refs. 9,18,19, the ZBCP disappears at $T \simeq 40$ K. As can be seen from Fig. 2, a similar behavior is also observed in the present work. It is interesting to note that for contacts with a ZBRP, the contribution from coherent tunneling probably exists too. This follows from the analysis of the temperature dependences of the dynamical resistances at zero bias. It is seen in Fig. 4 that the upper curve, which corresponds to noncoherent tunneling, has an inflection point at the same temperature at which the amplitude of the ZBCP saturates. It means that the probability of coherent tunneling is nonzero for such type of contacts and that, in real point contacts, both types of tunneling take place simultaneously.

Let us now analyze if self-heating in our experiments may explain the observed IV singularities in the tunneling spectra as it was proposed in Ref. 30. First, the temperature dependence, $R_{d0}(T)$, is not universal and is qualitatively different for different types of junctions (Fig. 4). This indicates that the observed IV singularities are not due to the temperature dependence of the bulk resistance. Second, the variation in contact resistance in our junctions achieves 3 orders of magnitude. In such conditions, one expect important changes in the heating conditions, and consequently, significant changes in the bias voltage peculiarities should be observed too. However, the voltage positions of minima of $R_d(V)$ in our experiments remain unchanged. Third, we have not observed indications of a noticeable heating in our contacts that should appear as a strong parabolic $R_d(V)$ dependence at high bias voltages. So, we have all the reasons to associate these singularities with the Peierls energy gaps and not with self-heating up to the Peierls transition temperatures. Thus, the similarity of the spectra observed in stacked junctions[18–20] and in the present work allows us to conclude that effects of self-heating in NbSe$_3$-based stacked junctions are not relevant. There are, however, some uncertainty in the absolute values of both energy gaps. For measurements based on tunneling (through Pb contacts, point contacts, or mesas), the different low temperature Δ_{p2}/e ranges from 52 mV,[23] 24 mV (present work), 25 mV,[18] 35 mV,[26,34] and 37 mV,[24,25] while Δ_{p1}/e ranges from 120 mV,[18] 101 mV,[34] 100 mV,[24,25] 95 mV,[23] and 89 mV (present work). In spite of their dispersion, these values are in good agreement with those obtained from photoemission:[35,36] 45 mV for Δ_{p2}/e and 110 mV for Δ_{p1}/e. Anisotropy of the CDW gap magnitude has also to be considered in Ref. 28. Note, however, that the energy gap values estimated from infrared spectra[33] are larger than the gaps obtained from tunneling. Taking into account the anisotropic gapping for the diagonal nesting,[28,33] this difference can be explained by the fact that optical measurements average over the k space.

As can be seen from Figs. 1 and 3, contacts with a high zero bias resistance R_{d0} demonstrate a typical quasiparticle tunneling behavior.[26] However, we cannot explain the observed contact characteristics in the frame of an ordinary tunneling assuming only the existence of normal electrons and holes at the Fermi level, noncondensed into the CDWs. In this case, two singularities, one at $V = \pm\Delta_p/e$ corresponding to a N-CDW-type tunneling and the other at $V = \pm2\Delta_p/e$, should be observed as predicted in Ref. 29. This
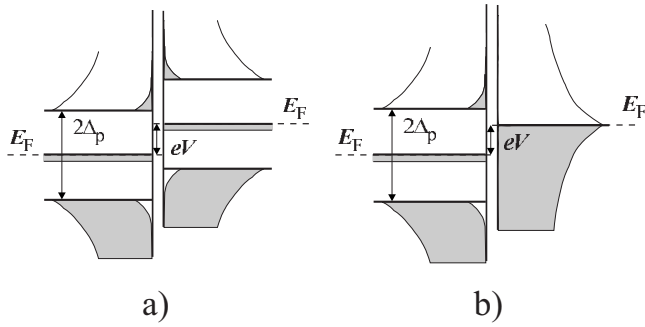
FIG. 5. Diagram of (a) CDW-insulator-CDW and (b) CDW–insulator–normal–metal tunnel junctions in the case of partly gapped electronic spectrum in the Peierls state.

result is illustrated in Fig. 5(a) where the energy diagram of a CDW-I-CDW junction is shown schematically for the case of a partly gapped electronic spectrum. So, the singularities at $V=\pm\Delta_{p2}/e$ (24–28 meV) and $V=\pm 2\Delta_{p2}/e$ as well as at $V=\pm\Delta_{p1}/e$ (80–100 meV) and $V=\pm 2\Delta_{p1}/e$ should be observed in NbSe$_3$-I-NbSe$_3$ junctions. Unfortunately, at high bias voltages ($V>120-140$ mV), the noise increases significantly, the characteristics of point contacts become very unstable, and correct measurements were impossible. So, it was not possible to observe the singularity corresponding to double Peierls gap corresponding the first CDW.

However, in the present experiments, we have not observed either the singularity corresponding to Δ_{p2}/e. At the same time, the energy gap singularity at $|V|\approx 80$ mV (Δ_{p1}/e) corresponding to a N-CDW-type tunneling for the upper temperature CDW is clearly detectable. As can be seen in Fig. 5(b) in the case of CDW-I-N tunnel junction, only the singularity at $V=\pm\Delta_p/e$ should be observed. Thus, the N-CDW-type tunneling for the low temperature CDW in NbSe$_3$ has been observed in the characteristics of normal-metal–NbSe$_3$ point contacts.[28] As an example, the differential IV curve of a Au-NbSe$_3$ point contact measured at $T=4.2$ K is shown in inset of Fig. 3. It can be seen that both energy gap singularities at $|V|=85$ mV, corresponding to Δ_{p1}/e, and at $|V|=25$ mV, corresponding to Δ_{p2}/e, are visible as characteristic minima in $R_d(V)$.

The absence of a singularity at Δ_{p2}/e in the case of symmetrical CDW-I-CDW junctions indicates that, in NbSe$_3$, the ordinary tunneling of normal carriers is only possible for the high-temperature CDW but not for the lower one. The same results were obtained in the case of interlayer tunneling[18–20] and for longitudinal nanoconstrictions.[23] Formally, that means that the carriers remaining in pockets cannot be involved as quasiparticle excitations for the low-temperature CDW. Such a situation is possible, for example, if the CDW order parameter has an unusual symmetry.[37,38] By analogy with oxide HTSC systems, one may consider the remaining pocket carriers in NbSe$_3$ as a kind of nodal carriers. However, this hypothesis needs to be further tested by new experiments.

V. CONCLUSION

In summary, we have shown that the differential current-voltage characteristics of NbSe$_3$-NbSe$_3$ artificial contacts (point contacts) formed along the a^* axis exhibit a tunneling behavior in the charge-density-wave (CDW) state. Depending of the contact resistance, a crossover from coherent to noncoherent tunneling was observed below the second Peierls transition ($T_{p2}=59$ K). In the case of noncoherent tunnel junctions, we have observed typical one-particle tunnel IV characteristics with energy gaps singularities corresponding to Δ_{p1} and $2\Delta_{p2}$. The contacts demonstrating coherent tunneling reveal all the features observed in stacked mesa structures: large zero bias conductance peak observed at low temperature, characteristic minima of $R_d(V)$ corresponding to the energy gaps singularities for the upper CDW at $V=\pm\Delta_{p1}/e$ and for the lower the CDW at $V=\pm 2\Delta_{p2}/e$, and plateau of $R_d(V)$ near $V=0$. In both cases, there is no singularity corresponding to Δ_{p2}/e that indicates that the normal carriers noncondensed in the CDW remaining at low temperature cannot be quasiparticle excitations for the low-temperature CDW.

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