Subgap Collective Tunneling and Its Staircase Structure in Charge Density Waves

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Tunneling spectra of chain materials $NbSe_3$ and TaS_3 were studied in nanoscale mesa devices. Current-voltage I-V characteristics related to all charge density waves (CDWs) reveal universal spectra within the normally forbidden region of low V, below the electronic CDW gap 2Δ. The tunneling always demonstrates a threshold $V_t \approx 0.2\Delta$, followed, for both CDWs in $NbSe_3$, by a staircase fine structure. T dependencies of $V_t(T)$ and $\Delta(T)$ scale together for each CDW, while the low T values $V_t(0)$ correlate with the CDWs' transition temperatures T_p. Fine structures of CDWs perfectly coincide when scaled along V/Δ. The results evidence the sequential entering of CDW vortices (dislocations) in the junction area with the tunneling current concentrated in their cores. The subgap tunneling proceeds via the phase channel: coherent phase slips at neighboring chains.

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Electronic spectra of strongly correlated systems are formed self-consistently via interactions among electrons or between them and the host crystal. Commonly observed states with spontaneously broken symmetries are susceptible to external fields and are affected by even the measuring device. That can modify the ground state, being able to readjust to changes of the local concentration of electrons and even to added individual particles. We are concerned with so-called electronic crystals [1], especially charge density waves (CDWs) [2]. A usual temptation is to treat CDWs as a kind of semiconducting superlattices with a gap at the Fermi level. But contrary to usual crystals, here the number of unit cells (CDW periods) is not fixed and can be readjusted to absorb added or removed electrons to the extended ground state. Particularly intriguing is that this process goes on via topologically nontrivial defects: discommensurations, dislocations, solitons. Dynamically, they are created as instantons of field theories, i.e., phase slips in the language of incommensurate CDWs (ICDWs). In this Letter, together with the earlier publication [3], we show that all these effects appear in the experiments.

We report measurements of the intrinsic interlayer tunneling in two quasi-one-dimensional (1D) materials, $NbSe_3$ and o-TaS_3, below their Peierls transition temperatures T_p to CDW states. $NbSe_3$ has two ICDWs which develop below their corresponding T_p: $T_1 = 145$ K (CDW1) and $T_2 = 59$ K (CDW2) opening the gaps Δ_1 and Δ_2. In o-TaS_3, a single ICDW is formed below $T_p = 215$ K. In general, the intrinsic tunneling is observed [4] in layered or chain crystalline materials with well decoupled elementary conducting planes. Experimentally, the interlayer transport is studied by the method [5,6] of nanoscale devices called stacked structures, or mesas, or overlap junctions (Fig. 1). We fabricated the devices by a focused ion beam (FIB) technique developed previously for a high-T_c superconductor [7] and extended to the layered CDW material $NbSe_3$ [8]. The essence of the method is that all elements of the device are parts of the same single crystal. The typical size of the junctions is $1\ \mu m \times 1\ \mu m \times 0.05\ \mu m$, so the depth includes only 20–30 atomic layers. We used two modifications of the FIB processing: (A) the double-sided one [7] (used here for $NbSe_3$) and (B) the lateral one [5] (used here for TaS_3). Technique A allowed us to fabricate stacked structures with connecting electrodes oriented across the chains—Fig. 1, which prevents an interference between the interlayer tunneling and a possible CDW sliding within connecting channels.

Figure 2 shows the tunneling spectra dI/dV of $NbSe_3$ for both (a) upper and (b) lower CDW states and of (c) TaS_3. We performed experiments at higher T, $T_p/2 \lesssim T \lesssim T_p$, because at lower 4 K $\lesssim T \lesssim$ 20 K, the low V spectrum of $NbSe_3$ is dominated by a zero bias conducting peak (ZBCP), originated by specific carriers from remnant pockets [8]. Two features are common for both materials, including both CDW states in $NbSe_3$: the CDW gap peak at $V_g(T) = 2\Delta$ and the much lower sharp threshold voltage $V_t \approx 0.2\Delta$ for the tunneling onset. The peak at V_g is particularly pronounced at lower T, where $V_g(0)$ reaches the

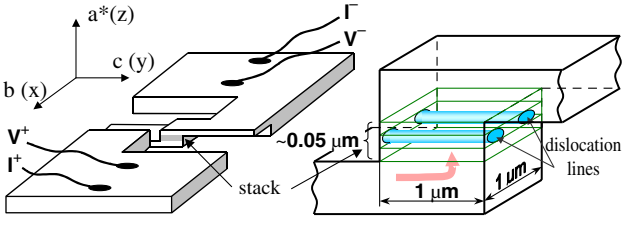

FIG. 1 (color online). Scheme of the tunneling device of type A. Crystallographic axes of $NbSe_3$ a^*, b, c (b is the chain direction) correspond to coordinates z, x, y. Elliptic cylinders show the cores of dislocation lines; the curved arrow shows the current path.

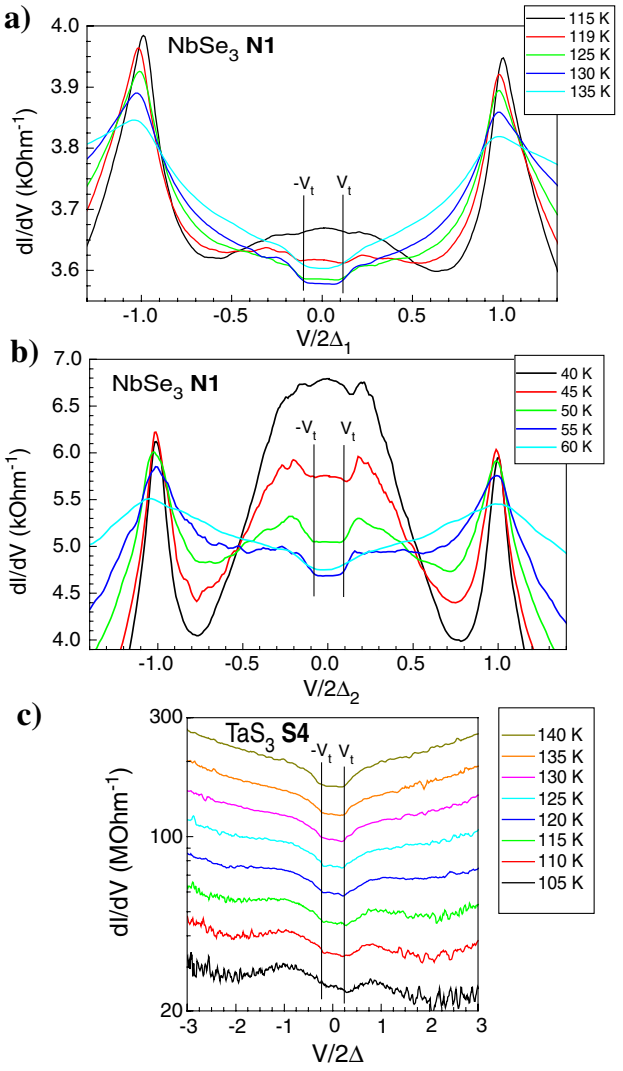

FIG. 2 (color online). Tunneling spectra dI/dV as a function of the voltage V normalized to the CDW gap 2Δ at different T: (a) CDW1 in $NbSe_3$, (b) CDW2 in $NbSe_3$, (c) o-TaS_3.

expected values $2\Delta_0$ of the CDW gaps ($V_g \approx 50$ mV, $\approx$150 mV for CDWs in $NbSe_3$ and $V_g \approx 180$ mV for TaS_3). These values are in reasonable agreement with those measured for $NbSe_3$ in optics [9], angle-resolved photoemission spectroscopy [10], tunneling, and STM [11], point contact spectroscopy [12], and for TaS_3 in optics [13].

The T dependence $V_g(T)$ in $NbSe_3$ is well fitted by the BCS law [8,14]; hence, at any T this peak can be attributed to the tunneling across the CDW gap $2\Delta(T)$. The gap peak in o-TaS_3 is much broader. To define its position at $T >$ 120 K, we subtracted from the original spectra the parabolic background fitted at high biases. That type of background has been confirmed by measurements above T_p. Figure 2 demonstrates that the ratio $V_t(T)/\Delta(T) \approx 0.2$ is temperature independent for all cases; hence, the origin of V_t is linked to the CDW gap. The next enlightening step is to compare V_t with the 3D ordering scale kT_p among the different CDWs. We find a surprisingly good linear relation $V_t(0) \approx 1.3kT_p$, in a wide range of T_p from 60 to 215 K. It proves that the origin of the threshold voltage is related also with the phase decoupling of CDWs in adjacent layers.

Figure 3(a) shows the fine structure in $NbSe_3$ at $T =$ 130 K. We see clear steps in dI/dV and corresponding sharp peaks in d^2I/dV^2. Figure 3(b) proves that this is not an artifact: For each CDW, the structure is well reproduced for both bias polarities. Moreover, Fig. 3(b) shows that for the normalized $V/2\Delta_{1,2}$ the peak positions coincide with a remarkable accuracy for both CDWs at so different $T =$ 130 K, 120 K, 50 K. It proves the universality of the fine structure and its common origin with V_t, viewed as the first step position. Similarly to the threshold feature, the fine structure was well resolved only at high enough temperatures where the ZBCP was suppressed.

The model presented below aims to explain these unexpected observations. Let (see Fig. 1) x be the chain direction along the junction; z is the transverse direction across the junction; y is the direction along the junction but perpendicular to chains. d_x, d_y, d_z are the corresponding unit cell sizes. The Fermi scales of the parent metal are the energy E_F, the momentum k_F, and the velocity v_F; then the screening radius $r_0 < d_z$ and the plasma frequency $\omega_p = \hbar v_F/r_0 \gg \Delta$. The ICDW is the modulation $\sim\Delta\cos(2p_Fx + \varphi)$ of the electronic density. Considering it as a rigid crystal, we would arrive at the conventional picture of a semiconducting superlattice where electrons e or holes h with the activation energy Δ would be responsible for all effects. But the ICDW is a self-adjusting system where the number of particles $n = \bar{n} + \delta n$ in the ground state is not fixed: $p_F = \bar{p}_F + \delta p_F$, $E_F = \bar{E}_F + \delta E_F$ can float with the gap 2Δ being attached to them. Local shifts $\delta E_F = v_F\delta p_F = v_F\delta n\pi/2$ correspond to the density (per unit length, per chain) $\delta n = \varphi'/\pi$ ($\varphi' = \partial\varphi/\partial x$) of particles added to the condensate. Contrary to usual conductors, the excess screening charge can come directly from the condensate density δn, if it is allowed to change across the layers (numbered by an integer m): $\delta n \Rightarrow \delta n_m$. But it requires a difference in $\varphi'_m - \varphi'_{m\pm1}$, which means a mismatching of their CDW periodicities corresponding to wave numbers $2p_{Fm}$. Hence, to onset the collective screening the interplane structural correlation must be broken, while normally the phases are correlated at $T < T_p$. As a feasibility argument, recall the x-ray studies [15], which demonstrate the interchain decoupling (actually between coexisting CDWs in $NbSe_3$); also, the possibility of local variations along the chain is well confirmed by space resolved x-ray studies [16].

Consider first the limit of a vanishing interplane coupling $J_z = 0$ which strictly is realized above the 3D ordering at T_p. When neighboring CDWs are well decoupled, they react to the electric potential Φ in the same way as the parent metal without the gap would do: The gap can float together with E_F. The potentials will drop across the stack of layers as $\Phi_m \sim (r_0/d_z)^m$, which is fast at small $r_0/d_z \sim$ 0.2; hence, the voltage would be almost screened across

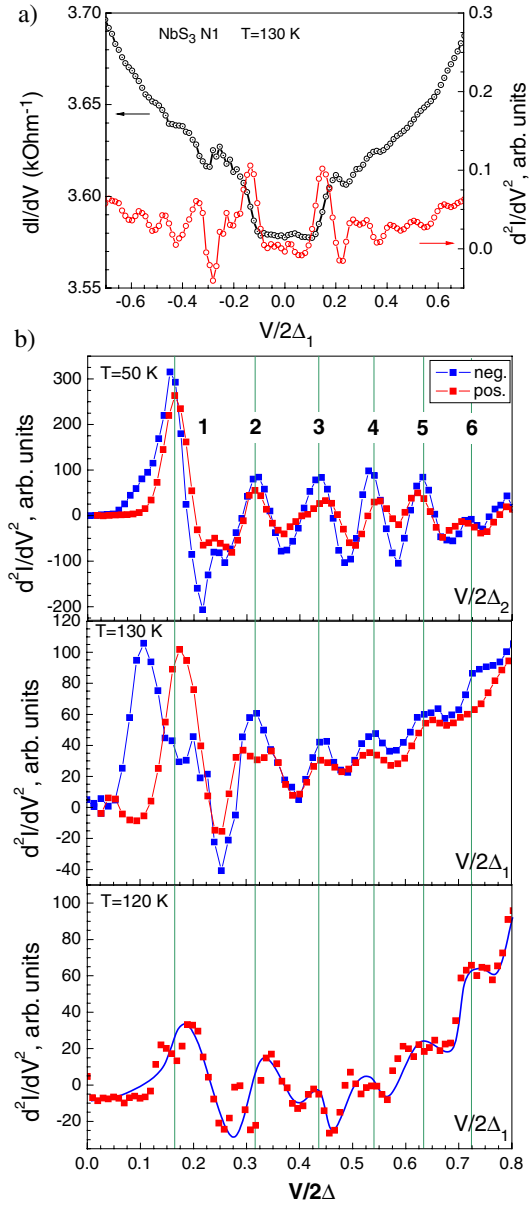

FIG. 3 (color online). Fine structure of the tunneling spectra in $NbSe_3$ within the magnified threshold region. (a) dI/dV and its derivative d^2I/dV^2 as a function of the voltage V normalized to the CDW gap, at $T = 130$ K. (b) Comparison of d^2I/dV^2 for the two voltage polarities for both CDWs, at $T = 130$ and 50 K and the positive polarity at $T = 120$ K (lines are guides for the eye). The peaks are interpreted as a sequential entering of dislocation lines into the junction area.

only one spacing. The minimal model treats the interlayer decoupling as an internal incommensurability transition. It takes into account only two layers 1 and 2 kept at potentials $\pm V/2$ as given by the energy functional (cf. [15])

$$\int dx\left(\frac{\hbar v_F}{4\pi}[\varphi_1'^2+\varphi_2'^2]+\frac{V}{2\pi}[\varphi_1'-\varphi_2']-J_z\cos(\varphi_1-\varphi_2)\right).$$

Here the three terms are the CDWs' elastic energy, the interchain charge transfer gain, and the interchain coupling. Its minimization gives an array of interplane discommensurations which develops starting from the isolated line (in the y direction) of 2π solitons in the phase difference $\varphi_1 - \varphi_2$. Then V_t is identified as the energy necessary to create the first soliton: $E_s = 4(\hbar v_F J_z/\pi)^{1/2}$ [17]. At higher $V > E_s$, the solitons overlap [their width is $l_s = 4(\hbar v_F/J_z)^{1/2}$ [17]], and at high $V \gg E_s$ the phase difference changes almost linearly, $\varphi_1 - \varphi_2 \approx 2Vx/\hbar v_F$, which means complete decoupling.

The above model is only a transparent illustration: Actually, the decoupled planes are the boundaries for two semivolumes $z \gtrless 0$ with the voltage $\pm V/2$ applied at the distant outer boundaries of the junction, while the decoupling happens in between. The former lattice of discommensurations must be generalized to a sequence of dislocation lines (DLs—the CDW vortices [18]). V_t is to be identified as the DL entry energy, in analogy to the H_{c1} field for entering the Josephson vortex in layered superconductors. The DLs are lying along the y direction, presumably in the median junction plane $(x, y, 0)$ [19]. Close to the plane $z = 0$, the sequence of DLs will look almost like a solitonic lattice with $\pm e$ charged phase increments of $\pm\pi$ ($\pm$ for passing above or below the DL). The distribution becomes more diffuse at distant planes and eventually overlaps, but invariantly the total phase difference 2π is accumulated between the lower and the upper halves of the junction, hence the charge transfer $2e$ per chain per DL. The two-plane interaction is generalized as a distributed shear stress energy $\sim\{(\partial_x\varphi)^2 + (\beta\partial_y\varphi)^2\}$ characterized by the dimensionless parameter of anisotropy $\beta \sim d_z(J_z/\hbar v_F)^{1/2} \ll 1$.

There is also the Coulomb energy, which is particularly costly for charged phase variations φ' along the chain x; hence, they must be slow relative to other directions. Thus [20], the DL core has the atomic width d_z in z and a longer length $l \sim 100$ Å in the x direction: $l = d_z^2/\beta r_0 \sim d_z\omega_p/T_p \gg d_z$. The electric field E_z is concentrated close to the DL plane $(x, y, 0)$: $E_z \sim (\Phi_0/d_z)(l/|x|)^{1/2} \times \exp(-(z/d_z)^2(l/|x|))$ [20]. The potential drop across the junction width is $\Phi_0 = \pi\beta\omega_p$ per each entering DL, which determines the threshold voltage $V_t = \Phi_0$ and gives the same quantization for further steps, as is seen in $I(V)$ [Fig. 3(b)]. Coulomb interactions increase the energy cost to create DLs but also enlarge their efficiency in building the potential: Only a few DLs are sufficient to cover the whole gap interval of $V < 2\Delta(T)$. Recall that $\beta(T)$, hence $\Phi_0(T)$, and finally $V_t(T)$ must have the same T dependence governed by the factor $\Delta(T)$, which is in accordance with the experiment [21]. The tunneling takes place over the distance $2z(x) \sim \sqrt{x/l}$ between matching points $\pm z(x)$ of surfaces $\Phi(x, z) \equiv \pm\Delta$. The probability $\sim \exp(-2z(x)/d_z) \sim \exp(-2\sqrt{x/l})$ is exponentially enhanced towards smaller x where $z(x)$ is small; i.e., the tunneling is confined within the DL core. Only here are the potential changes fast enough to give a short path for tunneling.

The interchain coupling energy is proportional to the square of the CDW amplitude $J_z \sim \Delta(T)^2$; then $\beta(T)$, hence $\Phi_0(T)$, and finally $V_t(T)$ must have the same T dependence governed by the factor $\Delta(T)$. It explains the clear scaling of all spectral features with $\Delta(T)$. At low T, $J_z(0) \sim T_p^2/\hbar v_F$, then the saturated value of V_t scales with T_p, which is another observation [21].

The final step is to understand the nature of the microscopic dynamical processes of tunneling at $V > V_t$ [22]. Several plausible mechanisms can be excluded. (a) Our special construction of the device eliminates the interference with the threshold for CDW sliding. (b) The usual tunneling through creation of $e - h$ pairs cannot take place below $V_g = 2\Delta$. (c) Dressed single electron states, the "amplitude solitons," reduce the energy Δ by $2/3$, as confirmed by experiments [3], but the scale is still too high for $V_t/\Delta \approx 0.2$. (d) Contribution of normal carriers gives an opposite dependence $I(V)$: (i) In $NbSe_3$, V_t appears only when the ZBCP is suppressed; (ii) the concentration of the potential drop upon one layer can only reduce the normal current.

We are left with a fascinating, while firmly based, picture that the excess tunneling conductivity above V_t can be provided only by the low energy phase channel. Empirically, one can already recognize the necessary scale from very low activation energies E_a for the on-chain conductivity measured in gapful CDWs in contrast to high Δ_0 for the transverse one (e.g., in TaS_3 [23] $E_a \approx 200$ K, while $\Delta_0 \approx 800$ K). This conductivity is associated with $\pm 2\pi$ phase solitons, which correspond to stretching/squeezing of a chain by one period, $\delta\varphi = \pm 2\pi$, with respect to the surrounding ones. Contrary to their aggregated form of static DLs, the solitons exist as single chain items: elementary particles with the charge $\pm 2e$ and the energy $E_s \sim T_p$. Their dynamic creation might be very sensitive to the threshold proximity $\delta V = V - 2E_s$ and to the chain number $M = 2z/d_z$ to tunnel through: The tunneling rate drops as [24] $\sim(\delta V/E_s)^{M\alpha}$, where the index $\alpha \sim v_F/u \gg 1$ is big because of the low phase velocity $u \ll v_F$. Hence, the pair of 2π solitons can be created by tunneling almost exclusively within the DL core, which process can be interpreted as a quantum excitation of the DL string.

In conclusion, we have measured intrinsic interlayer tunneling in nanostructures of the quasi-1D materials $NbSe_3$ and TaS_3. Enhanced tunneling occurs above a threshold voltage V_t which scales with the CDW gap as $V_t \approx 0.2\Delta$. For $NbSe_3$, the tunneling spectrum exhibits a staircase structure. We have presented a theoretical model in which the tunneling occurs through a single layer in the core of dislocation lines in the junction plane. Sequential formation of a DL grid at $V > V_t$ gives rise to the observed steps. The resolved, for the first time, tunneling in the normally forbidden subgap ($2\Delta > V > V_t$) region recovers collective quantum processes such as coherent phase slips at adjacent chains.

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