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Interaction of both charge density waves in $NbSe_3$ from interlayer tunneling experiments

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Interlayer tunneling technique has been used for spectroscopy of charge density wave (CDW) energy gaps, $\Delta_{1,2}$, in $NbSe_3$ subsequently opened at the Fermi surface on decreasing temperature at $T_{p1} = 145\,K$ (CDW1) and at $T_{p2} = 60\,K$ (CDW2). We found that the CDW2 formation is accompanied by an increase of the CDW1 gap below T_{p2}. The maximum enhancement of Δ_1, $\delta\Delta_1$, is about 10%. The effect observed has been predicted theoretically as resulting from the joint phase locking of both CDWs with the underlying crystalline lattice below T_{p2}.

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$NbSe_3$ is a chain charge density wave (CDW) compound which undergoes two seemingly independent charge density wave transitions at $T = T_{p1}$, T_{p2}. Below T_{p2} both CDWs in $NbSe_3$ coexist. The point whether they are independent or interacting with each other has been widely debated in literature. The first observations showed that the formation of the CDW2 has a negligibly small effect on the first one. That was based on the facts that no observable change occurs below T_{p2} in either the position [1] or intensity [2] of the diffraction spots associated with the first CDW. However, another experimental fact pointed out the possible joint commensurability effect between the two CDWs and the lattice. Namely, the wave vectors characterizing the two CDWs in the temperature range of their coexistence [1 – 3]: $q_1 = (0, 0.241,\ 0)$ and $q_2 = (0.5,\ 0.260,\ 0.5)$, in units of the reciprocal unit lengths, satisfy the approximate relation:

$$2(q_1 + q_2) \cong (1,\ 1,\ 1), \tag{1}$$

i.e. their sum is nearly a half of the reciprocal lattice vector. Some authors [4, 5] considered this point as an evidence for the phase coupling between both CDW's below T_{p2}. The analysis based on the simple Ginzburg–Landau theory [5] pointed out that the phase-locking effect can be accompanied by a small enhancement of the CDW1 energy gap below T_{p2}.

Recent studies revealed some features of the interaction between both CDWs in their dynamical state when one or both CDWs are sliding. In particular, it was found that the threshold field for depinning the CDW1 decreases below T_{p2} [6]. It was also found that the sliding of both CDWs below T_{p2} causes a correlated and opposite shift of the superlattice positions, whereas the projection of $q_1 + q_2$ along the chain remains unchanged [3]. This was interpreted as a dynamical decoupling of both CDWs.

However, the static phase-locking effect and the corresponding enhancement of the CDW1 gap below T_{p2} still has not been experimentally verified. That was partly related to the absence of a reliable and sensitive technique for the CDW gap spectroscopy. Recently a novel interlayer tunneling technique has been adapted for studies of the CDW energy gap [7] in $NbSe_3$. The high sensitivity of this method allowed to resolve intragap CDW states related to the CDW amplitude and phase excitations [8, 9]. Using this technique we undertook the search of the interaction between both CDWs in NbSe3 below T_{p2}.

Experiments have been carried out on three $NbSe_3$ stacked junctions oriented along the a^*-axis with sizes $L_b \times L_c \times L_{a^*} = 1\,\mu m \times 1\,\mu m \times (0.05{-}0.5)\mu m$. The junctions have been fabricated by double-sided processing of $NbSe_3$ thin single crystals by a focused ion beam [10]. SEM pictures of a typical stacked junction are shown in Fig.1. Microscopically, the stacked junction represents a vertical stack of elementary junctions formed by the layered crystalline structure of this material, the number of which can be varied from several tens to several hundreds [7]. The spacing between elementary junctions is about 1 nm. With an increase of the bias voltage across the stack the voltage drops on the weakest elementary junction [11], thus providing the possibility of interlayer tunneling spectroscopy on a single elementary tunnel junction.

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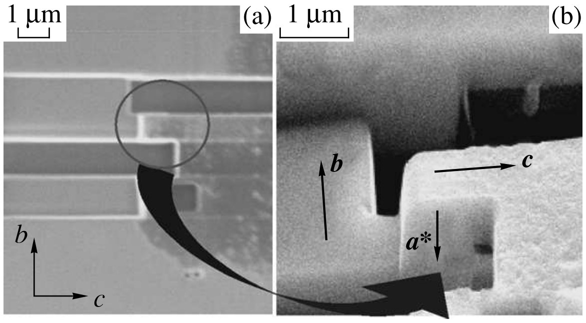

Fig.1. SEM pictures of a NbSe$_3$ stacked structure: (a) along the a^*-axis, (b) at the angle of 60° to the bc-plane

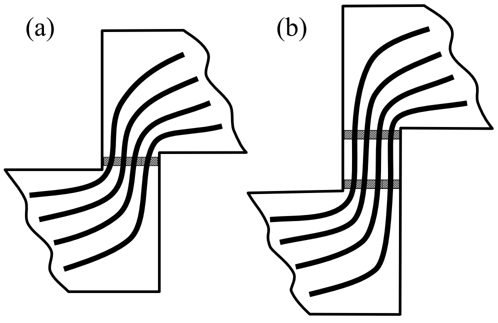

Fig.2. Schematic view of formation of a weak junction in thin stack (a) and a couple of weak junctions in a "thick" stack (b) in regions of the highest current density

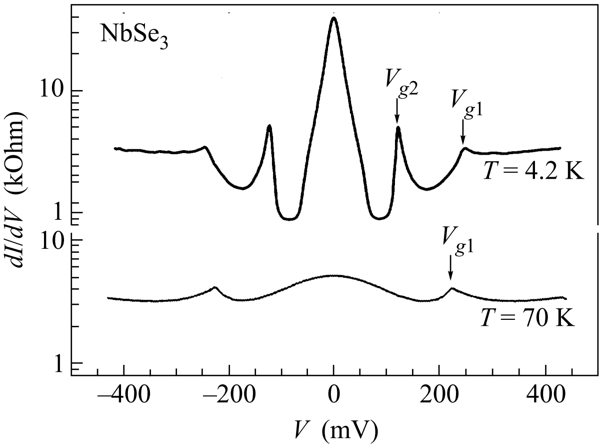

Fig.3. Interlayer tunneling spectra of a NbSe$_3$ stacked structure with sizes $L_b \times L_c \times L_{a^*} = 1\,\mu\text{m} \times 1\,\mu\text{m} \times 0.3\,\mu\text{m}$. The peaks at $V = V_{g1}, V_{g2}$ correspond to the CDW energy gaps 2Δ such as $V_g = 2\,(2\Delta)$

As developed by Brazovskii [9] in his theoretical model, this process is characterized by the successive entering of phase CDW dislocations or dislocation lines (DLs) in the weakest junction at voltages exceeding some threshold value, $V_t \approx 0.1\Delta$ [9]. The DL has a charge $2e$ localized mostly in its core. The core of a dislocation line has a very short size in the transverse direction across the layers, which is about the spacing between elementary conducting layers: $s = 1{-}2$ nm, while its in-plane size l is 20-50 times bigger, $l \approx 2s\omega_p/T_p$ [9], where ω_p is the plasma frequency of the layered material. At low bias voltage, $V < V_t$, the potential is uniformly distributed over the stack. The entering of charged DLs lead to a redistribution of the potential where most of the voltage applied to the stack drops on a set of dislocation lines located along the weakest junction [9]. That means that the voltage mostly drops on this junction.

From geometrical considerations this junction should be located at the place where the maximum current density is achieved. For thin enough stacks, containing few tens of elementary junctions, that is likely to be the central junction. For thick stacks of few hundred junctions the more likely configuration includes two weak junctions located near the vertical ends of the stack where current concentration occurs (Fig.2). Experimentally we regularly observed that for thin mesas of thickness ≈ 50 nm the voltage position for the main peak in interlayer tunneling spectra corresponds to the overgap tunneling of a single junction, while for "thick" stacks of thickness more than 200 nm this peak is located at a twice higher bias voltage indicating that the voltage drops on two junctions connected in series. That observation supports our geometrical considerations.

Fig.3 shows typical interlayer tunneling spectra $dI/dV(V)$ of a "thick" stack at two temperatures, below T_{p1} (spectrum b) and T_{p2} (spectrum a). In spectrum b we see peaks corresponding to the high temperature CDW energy gap at $V = V_{g1}$, while at low temperatures there are two peaks, corresponding to both – upper at $V = V_{g1}$ and lower at $V = V_{g2}$ – CDW gaps. As mentioned above, $V_{g1,2} = 2(2\Delta_{1,2})$. The extracted CDW gap values at low temperatures are as follows: $2\Delta_{1,2} = 140$ mV, 60 mV. These values are consistent with the data obtained from STM [12], ARPES [13], infrared [14] and point contact [15] spectroscopies.

Besides, at low temperatures, there is also a zero bias conductance peak associated with coherent interlayer tunneling of the carriers located on the ungapped "pockets" of the Fermi surface [16]. Preliminary studies of the temperature dependences of both gap peaks pointed out the quite good scaling with the BCS-type dependence [7]. In the present paper we report on more detailed measurements of the temperature dependence of Δ_1 and especially in the vicinity of T_{p2}.

The temperature dependence of $\Delta_1(T)$ is shown in Fig.4. When temperature is decreased below T_{p1}, the

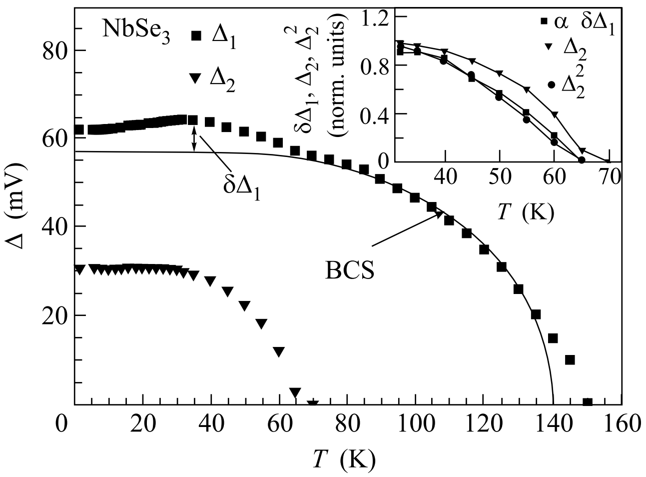

Fig.4. Temperature dependences of the CDW gaps Δ_1 and Δ_2 determined from interlayer tunneling spectra. The solid line is a fit by the BCS dependence for $T > 65$ K. The inset shows the temperature dependences of the enhancement in $\Delta_1(T)$, $\delta\Delta_1(T)$, below T_{p2} normalized to $\Delta_2(T)$ and $\Delta_2^2(T)$

gap of the CDW1 sharply increases, and then saturates; below T_{p2} one can see an additional increase of $\Delta_1(T)$. The increase in $\Delta_1(T)$ is more clearly seen from the comparison of the measured $\Delta_1(T)$ dependence with BCS-type $\Delta_{1\mathrm{BCS}}(T)$ dependence that is fitted to the measured $\Delta_1(T)$ at high temperatures $T > T_{p2}$ and is extrapolated to $T < T_{p2}$ (see solid curve in Fig.4). The enhancement in $\Delta_1(T)$ can be defined as $\delta\Delta_1(T) = \Delta_1(T) - \Delta_{1\mathrm{BCS}}(T)$. The temperature dependence of $\delta\Delta_1$ is plotted in the inset to Fig.4. The enhancement of Δ_1 below T_{p2} is evidently associated with the formation of Δ_2. A simple scaling of $\delta\Delta_1(T)$ with $\Delta_2(T)$ and $\Delta_2^2(T)$ (inset to Fig.4) shows a much better fit with $\Delta_2^2(T)$. This points out that the enhancement of Δ_1 is of the second order on Δ_2. The effect of enhancement of Δ_1 below T_{p2} has been reproduced on three stacked junctions.

Theoretical analysis of interactions on both CDWs in $NbSe_3$ based on the Ginzburg–Landau approach gives for the free energy F_2 at $T \le T_{p2}$ an additional term associated with the coupling of CDW phases φ_1 and φ_2 [5]:

$$F_2 = F_1(\Delta_1) + A_2\Delta_2^2 + B_2\Delta_2^4 + B_+\Delta_1^2\Delta_2^2\cos 2(\varphi_1 + \varphi_2). \tag{2}$$

$F_1(\Delta_1)$ in Eq. (2) corresponds to the free energy related with the formation of Δ_1, A and B are standard Ginzburg–Landau coefficients for expansion of the free energy. Considering $F_1(\Delta_1)$ to be nearly constant near T_{p2}, the minimization of F_2 on Δ_2^2 and $(\varphi_1 + \varphi_2)$ gives for Δ_2:

$$\Delta_2^2 = \frac{(A_2 - |B_+|\Delta_1^2)}{2B_2}. \tag{3}$$

Taking into account that A_2 becomes negative below T_{p2} one can see that the presence of Δ_1 increases Δ_2. The value of this enhancement is defined by the coefficient B_+ that is supposed to be small [4].

Similarly, one can consider that the presence of Δ_2 should increase Δ_1 as well since the interference term in the free energy is symmetric on both Δ_2 and Δ_1. However, a precise analysis for enhancement of Δ_1 using the Ginzburg–Landau approach has not been done since T_{p2} lies far below T_{p1}. Our experiment shows that the enhancement of Δ_1 is about 10%. Analysis of its temperature dependence points out that effect is proportional to Δ_2^2.

As follows from Eq. (3), the observed magnitude of Δ_2 should be enhanced by the presence of Δ_1. This even leads to the enhancement of T_{p2}, since the term $(A_2 - |B_+|\Delta_1^2)$ changes sign at temperature higher than the temperature where A_2 changes its sign. As it was mentioned in [5] the mutual influence of both gaps may be avoided in the sliding regime either of CDW2 or of both CDWs with different velocities. The phase coupling is expected to disappear in these cases. Recently, dynamical decoupling of both CDW wave vectors in the sliding regime has been observed [3]. We can suggest also a decrease of both CDW gaps in the sliding CDW case.

Note that the value of the enhancement of Δ_1 due to the phase coupling of CDW1 and CDW2 is very close to the energy eV_t associated with the phase decoupling of each CDW between adjacent layers forming the stack [9]. This energy was shown for both CDWs to be also about 0.1Δ.

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