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Interlayer tunneling spectroscopy of layered CDW materials

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Abstract

We demonstrated high efficiency of interlayer tunneling technique for spectroscopy of charge density wave (CDW) gap and intragap states in $NbSe_3$ and o-TaS_3 including experiments in high magnetic fields up to 28T.

1 INTRODUCTION

The method of interlayer tunneling is based on the layered crystalline structure of some highly anisotropic materials where elementary, atomically thin conducting layers are separated by elementary isolating layers. The transport along the layers in these materials is provided by metallic intralayer conductivity while the transport across the layers occurs via interlayer tunneling. If metallic layer undergoes at low temperatures a phase transition into electron condensed state (e.g. superconducting or CDW state) the interlayer tunneling may be used as an effective tool to study that state. In many cases as in layered superconductors the amplitude of the order parameter (OP) becomes vertically modulated, while phases in neighbour layers remain coupled. That results in effects of both types in interlayer tunneling, one related with phase interference (intrinsic Josephson effect) or phase decoupling (appearance of Josephson vortices at $H > H_{c1}$). Another type, as quasiparticle tunneling over a superconducting gap is related with the amplitude of the OP. Many fundamental properties of high temperature superconductors related with the symmetry of the OP [1], qap/pseudogap spectroscopy [2], dynamics of Josephson vortex lattices [3] have been explored using interlayer tunneling technique. One of the attractive features of the method to compare with some others widely used spectroscopic techniques as IRS, ARPES, STM is the possibility to get information from the bulk of the material.

Recently, the method has been extended to other types of layered materials like layered manganites [4] and layered CDW materials [5]. For MX_3 type CDW materials the conducting chains are assembled in elementary conducting layers isolated from each other by a double barrier of insulating prism bases [5]. The CDW gap and zero bias conductance peak (ZBCP) have been identified in $NbSe_3$ using this method [5]. Here we report on the observation and studies of new collective states with the energy lying inside the CDW gap.

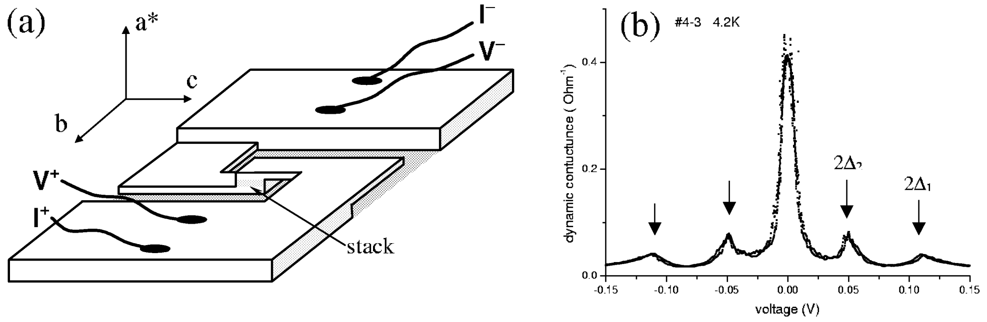

Fig. 1. Experimental set up (a) and low temperature interlayer tunneling spectrum of $NbSe_3$ stacked junction (b).

2 STACKED STRUCTURE FABRICATION AND CHARACTERIZATION

The stacked structures used for interlayer tunneling spectroscopy in HTS materials usually have micron scale lateral sizes and contain several tens of elementary junctions. The small sizes of the stacks provide phase coherence of interlayer tunneling [1] and highly reduced self-heating effects [6]. Recently the focused ion beam (FIB) technique has been developed for fabrication those structures [7]. We reproduced this technique for two CDW materials $NbSe_3$ and o-TaS_3. Experimental set up is shown in Fig.1a. The low temperature interlayer tunneling spectrum for $NbSe_3$ (Fig. 1b) clearly demonstrates both CDW gaps at bias voltage 50-60 mV and 130-150 mV, and ZBCP. The gap values found are consistent with STM [8], optics [9] and low temperature ARPES data [10] as well as with point contact spectra $NbSe_3$-$NbSe_3$ along the a^*-axis [5]. The results show that only one elementary tunnel junction in the stack is working at high bias voltage, and we consider this single junction as the weakest elementary junction, where phase decoupling of CDW occurs with increase of V.

We interpreted the ZBCP anomaly as a result of coherent interlayer tunneling of non-condensed carriers localized at small pockets on Fermi-surface [5]. Both, the height and the width of ZBCP characterize the stack quality. For the best samples the ratio of ZBCP height to the value of background at $V > 2\Delta$, r, reaches 30 and the ZBCP width is as small as 10 mV. To the contrary, for poor quality stacked junctions that ratio r drops to 5-10 and for the a^*- axis oriented point contacts $NbSe_3$-$NbSe_3$ that is even less, $r \sim 2$.

3 INTRAGAP CDW STATES

The order parameter Δ_0 in the ground state of the incommensurate CDW (ICDW) can be expressed as $\Delta_0 = A \cdot cos(Qx + \phi)$ with Q the CDW wave vector $Q = 2k_F$ and ϕ the arbitrary phase in the ICDW state A =const. That means that the ground state is degenerated with respect to $A \leftrightarrow -A$. That leads to the possibility of configuration with accepting of one electron from a free band and formation of new ground state with $A = tanh(x/\xi_0)$ called the amplitude soliton (AS) [11]. AS is a self-localized state with an energy $E_\xi = 2\Delta_0/\pi$ [11]. This state is preferable since its energy is smaller than the lowest energy Δ_0 of the free band electron by $\Delta_0/3$. The existence of ASs has been well documented for dimeric CDW materials (polyacetilene or $CuGeO_3$). However, for ICDW materials of higher order incommensurability as MX_3 existence of ASs has not been reliably demonstrated yet. We undertook an attempt to search the AS states using interlayer tunneling first in $NbSe_3$. To eliminate the masking

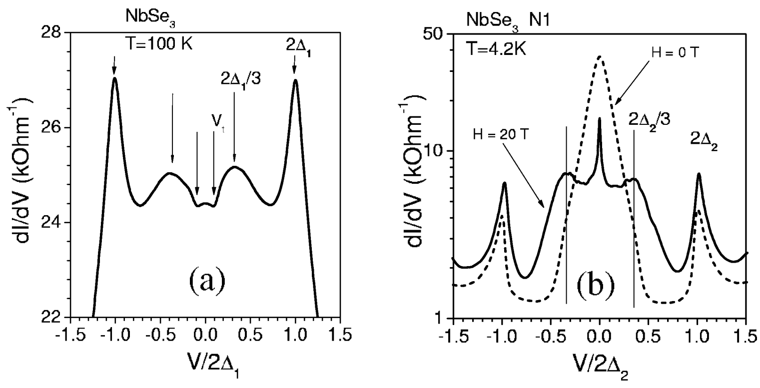

Fig. 2. The intragap peak on NbSe3 interlayer tunneling spectra at $V = 2\Delta/3$: (a) upper CDW, (b) lower CDW under magnetic field $H = 20$ T, $H \parallel c$.

effect of ZBCP we used either high temperatures regime when ZBCP is highly suppressed or low temperature regime in the presence of high magnetic field parallel to the layers, $H \parallel c$, when ZBCP is significantly narrowed under field [5].

In both cases we succeeded to observe a peak of interlayer tunneling conductivity at $V = V_s = 2\Delta/3$ [12](Figs. 2a, 2b). That has been observed for both upper and lower CDW in NbSe3. At the next step we have studied the interlayer tunneling spectra for o-TaS$_3$. That compound undergoes fully gapped Peierls transition at 215K and also experiences transition from ICDW to commensurate CDW (CCDW) state at 130K. The CDW gap feature is also clearly seen on the stacked junction of that compound (Fig. 3a). That is much more broaden to compare with NbSe$_3$. The ZBCP is absent in o-TaS$_3$. That confirms that its origin related with existence of the ungapped pockets.

The temperature dependence of the gap has two steps [13](Fig. 3b): first sharp increase below T_p, saturation to 130 mV approaching 130 K and then a new increase by about 1/3 below the transition to the CCDW state. We explain the unexpected spectral features for both

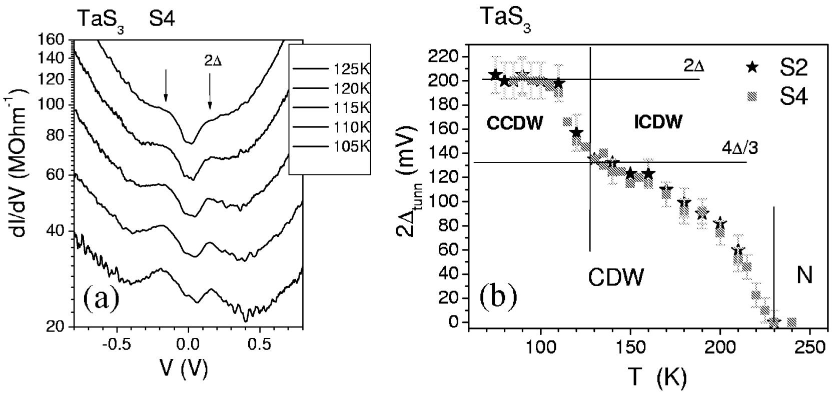

Fig. 3. The interlayer tunneling spectra of o-TaS3 stacked junction (a) and the extracted temperature dependence of tunneling gap (b).

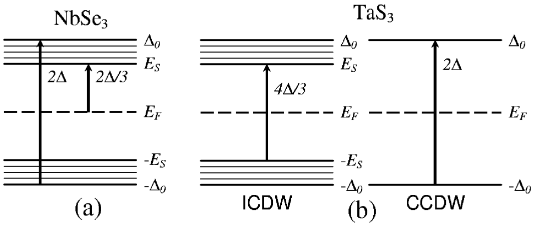

Fig. 4. Schematic view of soliton levels and tunneling transitions (marked by arrows) for $NbSe_3$ (a) and o-TaS_3 (b) for both ICDW and CCDW states.

materials by the presence of soliton states in the ICDW state schematically shown in Fig.4. In $NbSe_3$ that explains additional peak at $V = 2\Delta/3$ as related with transitions of pocket carriers to the AS levels. For o-TaS_3 we consider the observed broad gap in the ICDW state to be associated with transitions between soliton levels over a gap. In the CCDW state the phase of the OP is coupled with lattice periodicity and the amplitude solitons with acceptance of one electron are forbidden. As a result, the interband transitions in the CCDW state can be realized only between the band edges. At the experiment that looks as an effective increase of interband gap at the CCDW state. The expected increase of the gap is $(2\Delta - 4\Delta_0/3)/2\Delta = 1/3$. That is consistent with the value 0.3-0.35 observed experimentally.

4 THRESHOLD FOR INTERLAYER TUNNELING

Another remarkable feature we found at low energies within CDW gap is a sharp threshold for onset of tunneling conductivity. This threshold behaviour looks universal for both CDWs in $NbSe_3$ and for o-TaS_3 [14]. To exclude the possible explanation of this feature by simple CDW depinning in the connecting electrodes we fabricated special mesas where connecting electrodes were oriented across the chains (Fig. 1a). The threshold behaviour was nicely reproduced on those mesas (Fig. 5a). The value of threshold voltage V_t was found to be very low. For all the cases we studied the ratio $V_t/2\Delta$ was about 0.1 and was temperature independent demonstrating the scaling of V_t with CDW gap. We also found scaling relation between low temperature value of V_t value and Peierls transition temperature $eV_t \approx 1.3kT_p$ for wide variation of T_p from 60 K of lower CDW in $NbSe_3$ to 215 K in TaS_3. The energy $\sim kT_p$ is known as an energy of 3D CDW ordering. As known from structural measurements, above T_p transversal phase coherence of the CDW becomes lost. Therefore, V_t may be interpreted as the energy of phase decoupling between neighbour elementary layers.

S. Brazovskii suggested [14] that this decoupling occurs via successive entering in the "weakest" junction a set of dislocation lines (DLs). DLs appear as a result of share stress induced by electric field across the layers. Each DL is oriented across the chains in elementary junction and corresponds to the charge $2e$ per chain or corresponding to one unit of CDW period. In some sense DL can be considered as phase CDW vortex since circulation around DL gives phase variation 2π. That looks very similar to the Josephson vortex. There is also some similarity between V_t and H_{c1} in the layered superconductors.

It was shown [14] that electric field concentrates within dislocation core having vertical size $d_z \sim 10$ Å, i.e. that drops within one junction. The lateral size of the DL core, L, is much larger $L = d_z\omega_p/T_p$, where ω_p is a plasma frequency of material. We can estimate the ratio $w_p/T_p \sim 20$. That gives for $L \sim 200$ Å. For the junction of 1 μm lateral size one needs to

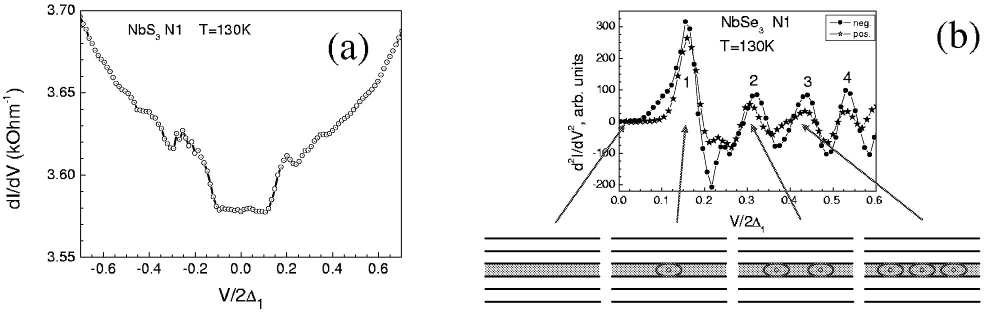

Fig. 5. Threshold for interlayer tunneling conductivity in NbSe$_3$ stacked junction (a) and its fine structure (b). The insert schematically shows subsequent entrance of dislocation lines in the weakest elementary junction.

have 5-10 DLs to overlap all the junction area and to achieve complete decoupling of neigbour layers. When dislocation cores start to overlap at high voltage bias all the voltage drops on a single elementary junction. That can explain puzzling equivalence of the behaviour of the stacked junction and point contact containing one junction.

As a consequence of this model, one can expect multiple threshold structure corresponding to successive entering of DLs with growth of electric field across the layers. A careful measuraments of tunneling spectra above V_t reveal a staircase quasi-periodic structure of increases of dynamic conductance with bias voltage that was reproducible for both voltage polarities and for both CDWs in NbSe$_3$. That is more clearly seen at the second derivative picture (Fig5b). That observation strongly supported Brazovskii model.

Comparison of two systems of layered superconductors and layered CDW materials shows remarkable similarity: the interlayer phase decoupling in both cases occurs via formation of phase topological defects at the energies much lower then the energy of the gap.

5 FIELD INDUCED METAL-INSULATOR TRANSITION IN NBSE$_3$

A puzzling property of NbSe$_3$ that has been debated for a long time is an anomalously high magnetoresistance in this material [15]. One of the model atributed this behaviour to the field induced CDW transition [16] that happens due to the improvement of nesting conditions and the closure of the pockets at high magnetic fields. That contradicted to the observation of the Shubnikov-de Haas (ShdH) oscillations in this material up to the very high fields indicating still the existence of pockets at those fields. The picture could be clarified by tunneling measurements. However, until recently there were no tunneling measurements at high fields. Here we report on our preliminary measurements of interlayer tunneling in high fields perpendicular to the layers. The results are shown at Fig.6. One can see strong suppression of ZBCP and tunneling density of states within a CDW gap with field growth. As a result, conductivity at bias voltages below 2Δ can be decreased as much as by 2 orders at 28 T exhibiting colossal magnetoresistance. Simultaneously we found some increase of the lower CDW gap by about 20%. The picture, indeed, looks as field induced CDW transition. However, the ZBCP still survives and acquires step-like form indicating an existence of transverse coherent state at low biases even at very high fields. Probably, that state is responsible for observations of ShdH oscillations at very high fields.

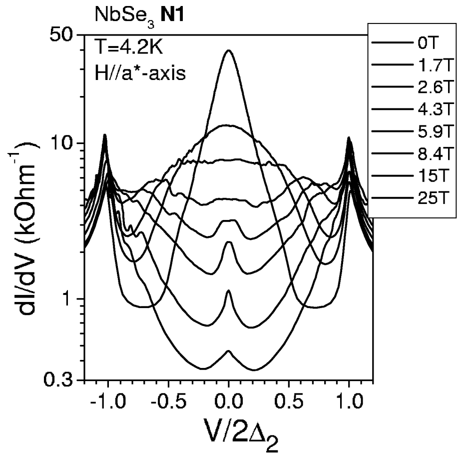

Fig. 6. Interlayer tunneling spectra of $\mathrm{NbSe_3}$ at high magnetic fields perpendicular to the layers, $H \parallel a^*$.

6 SUMMARY

Using focused ion beam technique we elaborated micron sized stacked junctions valid for interlayer tunneling spectroscopy of typical CDW materials of two types with fully gapped Fermi-surface(o-TaS3) and partially ungapped one ($\mathrm{NbSe_3}$). Along with CDW gap features we found strong evidence of the existence of self-localized states of amplitude soliton type in both materials with energy close to the expected value $2\Delta/\pi$. At even lower energy scale we found the threshold behaviour for the onset of interlayer tunneling accompanying by quasi-periodic staircase structure. That behaviour has been interpreted as the phase decoupling between neighbour layers of the weakest junction that happens via formation of a set of dislocation lines. We studied interlayer tunneling spectra in NbSe3 at high magnetic field perpendicular to the layers. We found strong suppression of tunneling density of states at bias voltages within CDW gap by magnetic field that leads to a colossal interlayer magneto-resistance. However, coherent interlayer state at low biases still survives at high fields.

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