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Nonlinear interlayer transport in the aligned carbon nanotube films and graphite

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Abstract. Interlayer tunneling spectroscopy on graphite stacked junctions and on aligned carbon nanotube (ACN) films shows universal zero bias anomaly (dip) for both type of objects. For graphite this anomaly disappears above 30K while for aligned nanotube films that persists up to 350K. We consider this anomaly as a pseudogap that appears due to a presence of interlayer correlated state. In a presence of magnetic field of 1-10T oriented across the layers we found characteristic peaks on interlayer tunneling spectra of graphite mesas. Their voltage position and square root dependence on magnetic field let us to identify the origin of those peaks to be related with interlayer tunneling between Landau levels (LLs) in graphite typical for Dirac fermions in graphene.

1. Introduction

A discovery of graphene [1,2] stimulated returned interest to graphite itself. The presence of Dirac fermions has been found in graphite as well using ARPES technique [3] and has been confirmed later by STM [4] and IR transmission [5] experiments. Recently anomalous behavior of graphite at low temperatures under magnetic field was found and interpreted as a presence of fluctuations of superconductivity [6]. Also some evidence of non-homogeneous superconductivity with T_c ~ 25K [7] has been recently reported on thin samples of highly oriented pyrolytic graphite. Existence of superconductivity has been revealed in single wall and multi wall nanotubes [8] at temperatures up to 12 K. Also a sharp decrease of resistance of ACN films has been reported above 250K under a pressure [9].

All these findings, however, has not been supported yet by tunneling measurements. Recently, the interlayer tunneling technique has been developed for spectroscopy of layered superconductors and charge density wave materials [10]. We adapt this technique for studies of graphite and ACN films. Using interlayer tunneling technique we found a presence of a pseudogap states in both type of objects. In graphite that state exists below 25-30K, whereas in ACN films that was found at higher temperatures up to 350K. In a presence of magnetic field oriented along the *c*-axis we also identified transitions between LLs in graphite typical for Dirac fermions in graphene.

2. Experimental

The dense nanotube films with nanotube axis oriented perpendicular to the substrate ($LiNbO_3$, SiO_2, Si) has been grown by electron beam evaporation technique [11]. STM image of one of the film is shown in Fig. 1a. Mesa type graphite nanostructures (Fig. 1b) have been obtained by double-sided etching of thin single crystal of natural graphite in focused ion beam [12]. In structures of both types transport is realized across the elementary carbon layers. In ACN films interlayer transport happens across the carbon layers curved into the nanotubes. A method of interlayer tunneling implies that tunneling across the layers occurs via tunneling between elementary layers. The appropriate I-V characteristics have been measured by computer control system using the programmable current source and a nanovoltmeter. For measurements in magnetic fields up to 10T we used pulsed fields and a system of measurements of the I-V characteristics at few kHz described in [13].

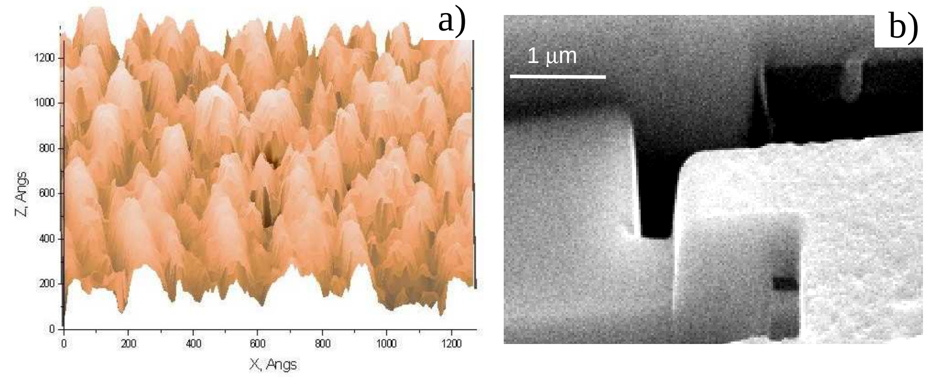

Figure 1. STM picture of aligned carbon nanotubes on YAG substrate (a), SEM picture of graphite mesa (b).

3. Results and discussion

3.1. Graphite mesas.

Fig. 2a shows temperature dependence of the resistance of graphite mesa #1 in comparison with in-plane resistance, measured on thin single crystal from the same batch. Room temperature resistivity anisotropy of natural graphite single crystals we used was about $4{\cdot}10^3$ and increases by factor 7 at low temperatures. The out of plane resistance is characterized by a slow growth with a temperature decrease with the following more steep growth part below 30K, while the in-plane resistance drops down sharply at low temperatures. High quality of single crystals we used has been also confirmed by IR transmission experiments [14]. Fig. 2b shows a series of the interlayer tunneling spectra dI/dV(V) at different temperatures. At T< 25K they exhibit a sharp zero bias dip that becomes more pronounced at low temperatures. Amazingly, the parts of spectra below and above some bias voltage V_0 behaves with temperature in a different way. Low bias dynamic conductivity increases with temperature (semiconducting type of T-dependence) while at $V>V_0$ that decreases with temperature (metallic type T-dependence), see Fig. 2b. That is a signature of the opening a gap (or a pseudogap) in electron spectra with energy value close to V_0.

The interlayer tunneling spectra of graphite mesas under magnetic field oriented across the layers (B = 0.4 - 6 T) show the peaks of differential conductivity dI/dV(V), which can be identified as transitions between Landau levels (LLs) with numbers N = 1, 2, 3 in valence band and $N' = N$ in conduction band. The most pronounced are peaks correspondent to transitions (1,1) (Fig.3a). The higher order peaks become resolved at fields above 2T. The energy of the peaks and their square root dependence on NB corresponds to relativistic LLs observed in graphene by magneto-transmission experiment [15] (see Fig. 3b). Our results are consistent with recent observations of Dirac fermion features in graphite by ARPES [3], STM [4] and magneto-transmission [5,14] methods.

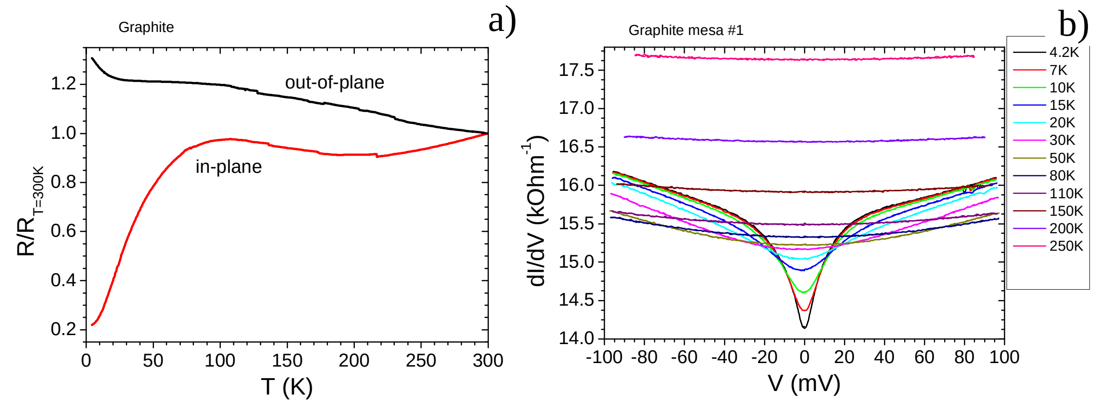

Figure 2. Temperature dependences of the out of plane and the in-plane resistance of graphite single crystals (a), interlayer tunneling spectra of graphite mesa (b).

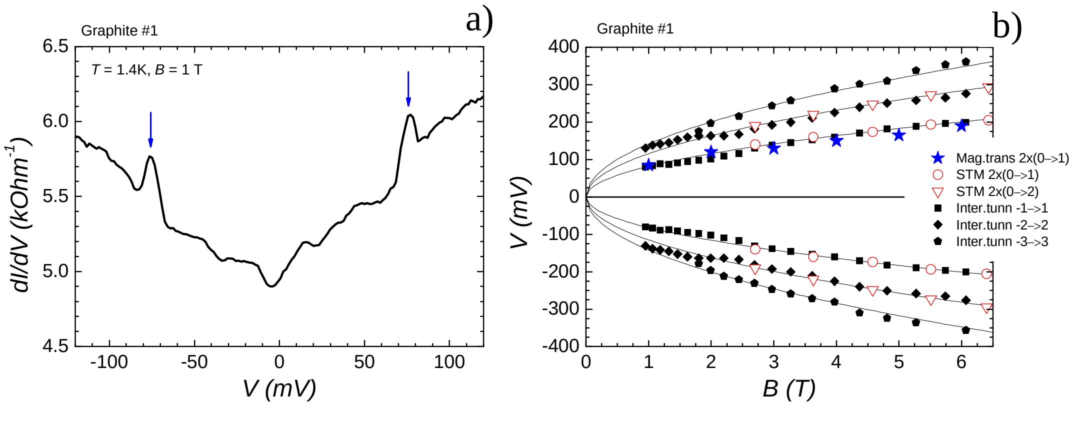

Figure 3. The interlayer tunneling spectrum dI/dV(V) of the graphite structure #1 at 1.4 K and B= 1T, B//c (a). The peaks correspond to transition (-1,1) are marked by arrows. Dependences of the doubled energy of the first three Landau levels extracted from interlayer tunneling data (full symbols) on magnetic field in comparison with STM data [6] (open symbols) and magneto-transmission data [7] (asterisks). Solid lines correspond to theoretical fit for Dirac fermions with Fermi velocity $v_F=10^8$cm/s (b).

3.2. ACN films.

Fig. 4a shows a temperature dependence of the resistance of ACN film on $LiNbO_3$ substrate. A thermoactivated behaviour is observed below 100K. Resistivity grows by 3 orders with a temperature decrease from 300K down to 40K. However, dI/dV(V) spectra normalized by its value at zero bias are not different so much (Fig. 4b) and resemble spectra obtained on graphite mesas at low temperatures. Again, we observe the same character of dI/dV (V) dependences below and above some characteristic voltage V_0 with temperature variation (Fig. 4c).

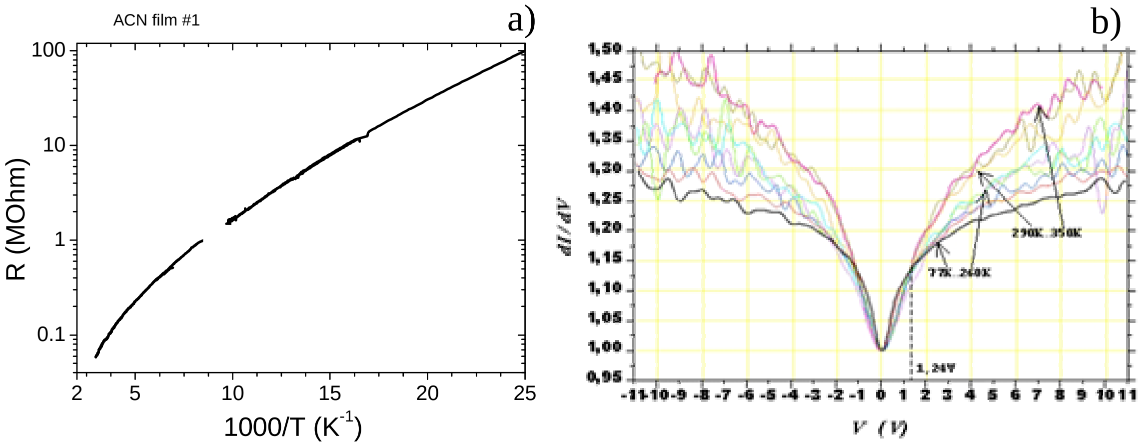

Figure 4. Temperature variation of resistance (a) and interlayer tunneling spectra (b) of ACN films on $LiNbO_3$ substrate.

3.3. Discussion.

Basically, the interlayer tunneling technique probes the interaction between elementary layers due to existence of some interlayer correlated states. We found a presence of interlayer correlations as a pseudogap features on interlayer tunneling spectra of both type objects. In graphite these correlations are weak and they has been observed only at low temperatures below 30K. In the ACN films they exist at high temperatures and become even stronger at T> 100K and T>250K (Fig. 4b). The nature of these pseudogap correlated states is still not clear. That may be mediated by interlayer Coulomb interaction or be a precursor to more ordered gap state, like superconductivity or charge density wave, that possibly may develop under pressure, magnetic field or at lower temperatures.

However, the universal shape of the pseudogap spectra for both objects (Fig. 5b) points out to its fundamental and common origin.

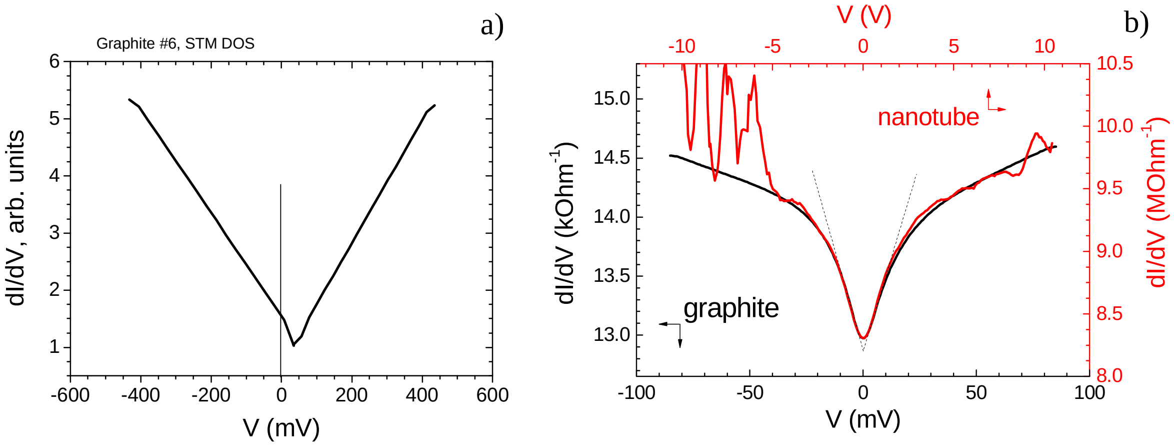

Figure 5. (a) STM spectra of graphite showing a linear behaviour of density of states with energy typical for 2D-case. (b) Scaling behaviour of zero bias anomaly for graphite mesa and ACN film.

As one can see from graphite interlayer tunneling spectra (Fig. 2b), the interlayer correlations in graphite become negligibly small at room temperature. Therefore, the observed linear V-shaped STM spectra most likely related with two-dimensional density of states of individual carbon nanolayer, graphene. The observed STM spectra are well consistent with recent density of state data extracted from ARPES experiments on graphite [3]. Moreover, the observed 50 mV shift in STM density of states (Fig. 5a) is the same as found from the ARPES data [3] and corresponds to the known shift of Fermi level from Dirac point in graphite [3].

Acknowledgments

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