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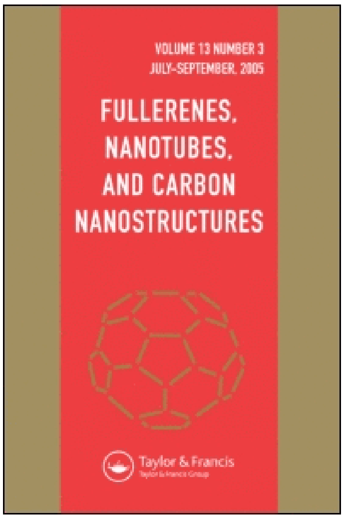

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Nonlinear Interlayer Transport in the Aligned Carbon Nanotube Films and Graphite

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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 it persists up to 350K. We consider this anomaly as a pseudogap that appears due to the presence of interlayer correlated state.

Keywords: Aligned carbon nanotube films, Graphite, Interlayer tunneling

INTRODUCTION

A discovery of graphene (1, 2) stimulated returned interest to graphite itself. The presence of Dirac fermions was found in graphite as well using the ARPES technique (3). Recently, anomalous behavior of graphite at low temperatures under magnetic field was found and interpreted as a presence of fluctuations of superconductivity (4). The existence of superconductivity has been revealed in single-walled and multi-walled nanotubes (5) at temperatures up to 12K. Also, a sharp decrease of resistance of ACN films has been reported above 250K under pressure (6). All these findings, however, have not been supported by tunneling measurements. Recently, an interlayer tunneling technique has been developed for spectroscopy of layered superconductors and charge density wave materials (7). We adapt this technique for studies of

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graphite and ACN films. Using interlayer tunneling technique we found a presence of a pseudogap states in both type of objects. In graphite this state exists below 25–30K, whereas in ACN films it was found at higher temperatures up to 350K. The STM studies of graphite show linear density of states typical for 2D case. That is consistent with ARPES measurements (3) and points to a very weak interlayer interaction in graphite at room temperature.

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 (8). STM image of one of the films shown in Figure 1a. Mesa type graphite nanostructures (Figure 1b) have been obtained by the double-sided etching of thin single crystals of natural graphite in a focused ion beam (9). In the structures of both types, transport is realized across the elementary carbon layers. In ACN films, interlayer transport happens across the carbon layers curved into nanotubes. A method of interlayer tunneling implies that a transport 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.

RESULTS AND DISCUSSION

Graphite Mesas

Figure 2a shows temperature dependence of the resistance of graphite mesa #1 in comparison with in-plane resistance, measured on a thin

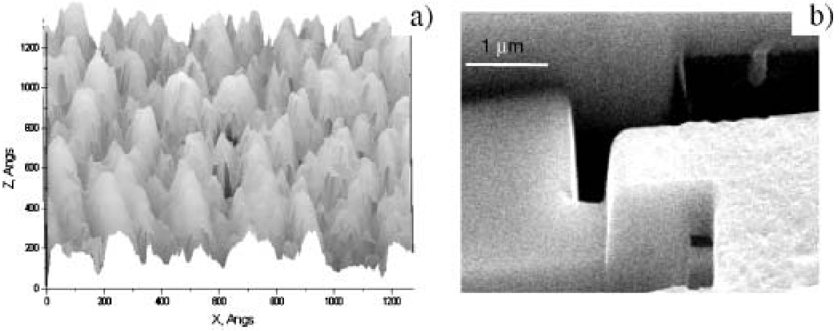

Figure 1. STM picture of aligned carbon nanotubes on $LiNbO_3$ substrate **(a)** and SEM picture of graphite mesa **(b)**.

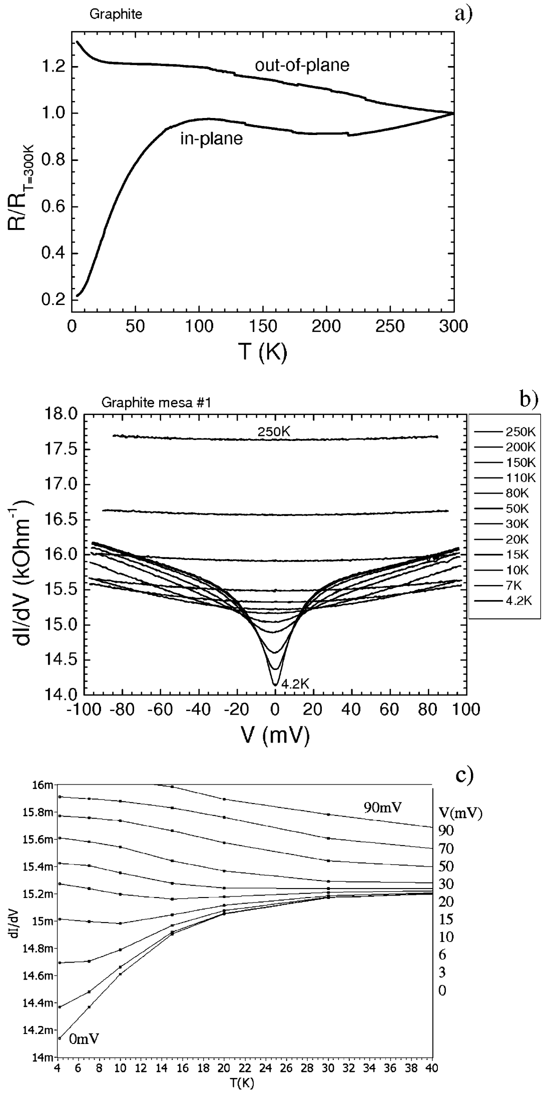

Figure 2. Temperature dependences of the out of plane and the in-plane resistance of graphite single crystals **(a)**, interlayer tunneling spectra of graphite mesa **(b)** and temperature behavior of dynamic conductance at different bias voltages **(c)**, extracted from **(b)**.

single crystal from the same batch. Room temperature resistivity anisotropy of the graphite single crystals was about 10^3. The out of plane resistance is characterized by a slow growth with a temperature decrease with the following more steep growth below 30K, while the in-plane resistance drops down sharply at low temperatures. Figure 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 Figure 2c). That is a signature of the opening a gap (or a pseudogap) in electron spectra with energy value close to V_0.

ACN Films

Figure 3a shows a temperature dependence of the resistance of ACN film on $LiNbO_3$ substrate. A thermoactivated behavior 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 (Figure 3b) 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 (Figure 3c).

STM Measurements

As a reference measurement, we also studied STM spectra of the same graphite single crystals we used for mesa fabrication. Figure 4a shows STM spectrum dI/dV(V) measured using tunneling microscope Beetle (Karl Zeiss Co). The measurements have been carried out at room temperature and tunnel currents below 1 nA. First a sample has been scanned under bias voltage 300 mV and current 600 pA to select atomically smooth area. This area then has been used for measurements of the tunneling I–V characteristics.

DISCUSSION

Basically, the interlayer tunneling technique probes the interaction between elementary layers due to the 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 have been observed only at low temperatures below 30K. In the ACN films they exist at high temperatures and become even more stronger at T> 100K and T>250K

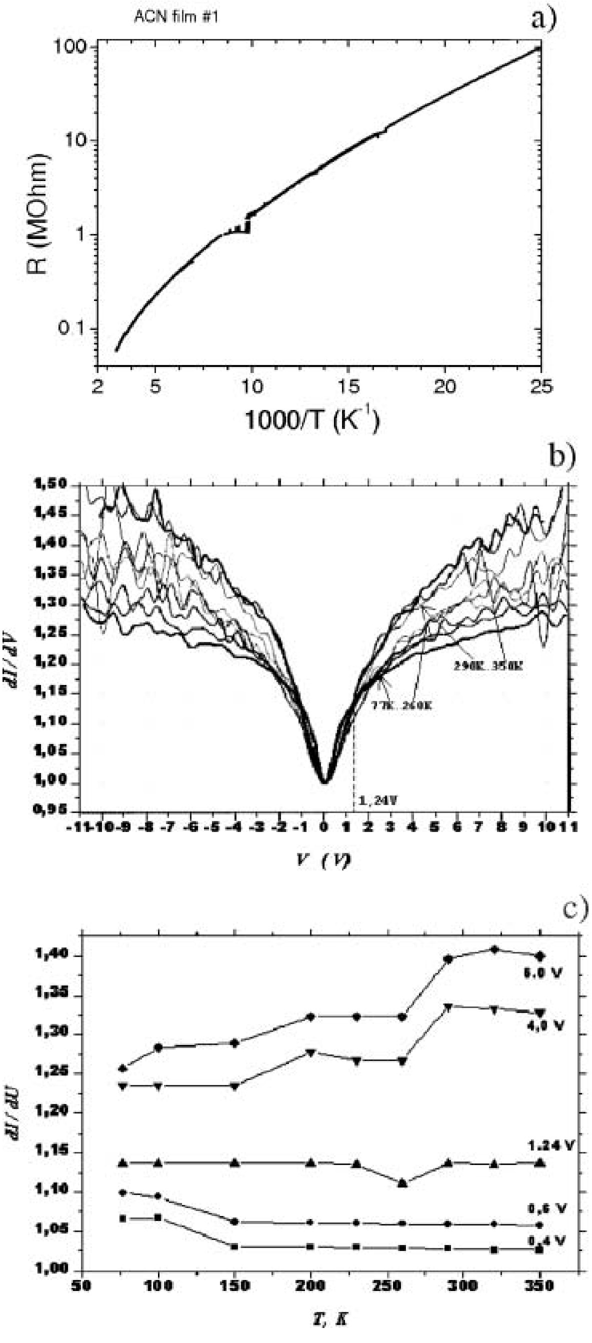

Figure 3. Temperature variation of resistance **(a)** and interlayer tunneling spectra **(b,c)** of ACN films on $LiNbO_3$ substrate.

(Figures 3b,c). 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 (Figure 4b) points to its fundamental and common origin.

As one can see from graphite interlayer tunneling spectra (Figure 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

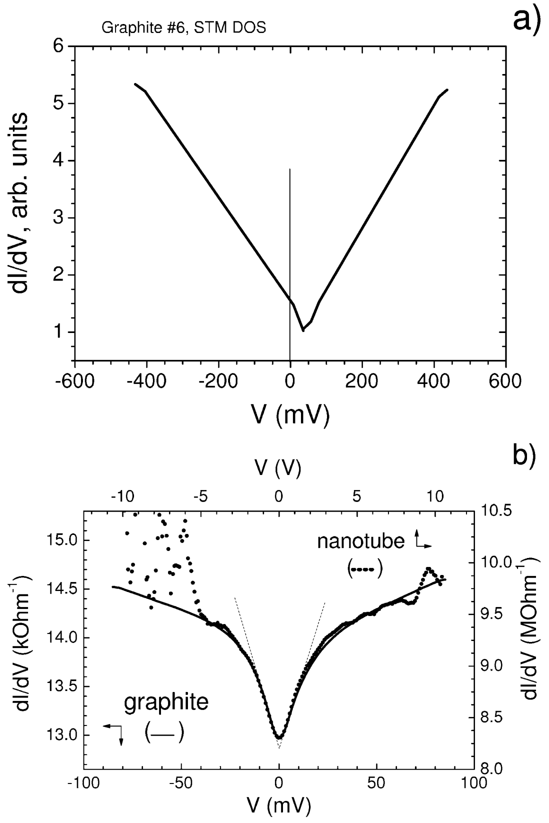

Figure 4. **(a)** STM spectra of graphite showing a linear behavior of density of states with energy typical for 2D-case. **(b)** Scaling behavior of zero bias anomaly for graphite mesa and ACN film.

(Figure 4a) is the same as found from the ARPES data (3) and corresponds to the known shift of the Fermi level from the Dirac point in graphene (3).

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