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The role of tunnel junction resistances and defects on electron transport mechanism in networks of two-dimensional disordered conductors

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HIGHLIGHTS

- The effect of tunnel junction resistances on the electronic property of FLGS networks was studied.
- At low TJRs below the quantum of resistance WL dominates and defects in FLGSs are neglected.
- WL effect is caused by self-interference of the electron wave function inside of FLGSs.
- At high TJRs Coulomb blockade effect results in SL and defects become important.
- Networks show negative MR in WL limit and positive MR in SL limit caused by Zeeman effect.

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ABSTRACT

The effect of tunnel junction resistances on the electronic property and the magneto-resistance of few-layer graphene sheet networks is investigated. By decreasing the tunnel junction resistances, transition from strong localization to weak localization occurs and magneto-resistance changes from positive to negative. It is shown that the positive magneto-resistance is due to Zeeman splitting of the electronic states at the Fermi level as it changes with the bias voltage. As the tunnel junction resistances decrease, the network resistance is well described by 2D weak localization model. Sensitivity of the magneto-resistance to the bias voltage becomes negligible and diminishes with increasing temperature. It is shown 2D weak localization effect mainly occurs inside of the few-layer graphene sheets and the minimum temperature of 5 K in our experiments is not sufficiently low to allow us to observe 2D weak localization effect of the networks as it occurs in 2D disordered metal films. Furthermore, defects inside the few-layer graphene sheets have negligible effect on the resistance of the networks which have small tunnel junction resistances between few-layer graphene sheets.

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1. Introduction

Tunnel junctions (TJs) are ubiquitous to nanoscale systems and can significantly affect the electronic property of the nanostructure assemblies. In a network of nanoparticles (NPs) if the tunnel junction resistances (TJRs) become smaller than the quantum of resistance ($R_Q=h/(2e^2)\sim 12.9\ \mathrm{k}\Omega$, where h is the Plank constant and e is the electron charge), the electron–electron (e–e) interaction results in a logarithmic correction to the conductivity irrespective of the network's dimensionality and the conduction will be insensitive to the magnetic field [1,2]. At higher TJRs, the network resistance increases exponentially with decreasing temperature (strong localization (SL) limit) [1,3]. But, in two-dimensional (2D) networks of NPs with low TJRs, and in 2D metals with a random distribution of impurities, the electron motion is diffusive and the self-interference of the electron wave function also results in a logarithmic correction to the conductivity. This effect is called 2D weak localization (WL) that by applying a magnetic field the phase interference is destroyed and a negative magneto-resistance (MR) is observed [1,4].

The logarithmic behavior caused by e–e interaction in 3D networks of NPs [5,6] and the logarithmic behavior due to 2D WL effect in disordered metal films [7,8] have been observed previously. Although, the logarithmic behavior in 2D disordered metal films and in 2D networks of NPs looks quite similar, the electron phase relaxation in 2D networks of NPs that destroys 2D

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WL effect can be caused by inelastic e–e interaction at two different temperatures. Below a critical temperature (T_c) the phase interference occurs due to propagation of the electron over distances larger than the size of NPs whereas at higher temperatures the inelastic e–e interaction can occur in a shorter time scale and the phase interference occurs only inside NPs [9].

Therefore, observation of the 2D WL effect in 2D network of NPs is quite challenging. There are several reasons for the limited experimental evidence of the 2D WL effect in 2D networks of NPs. First, the mean free path for the self-interference of the electron wave function is usually larger than the size of NPs. Therefore, the effect of self-interference is strongly diminished by electron scattering from the grain boundaries which can cause dephasing. Second, inelastic cotunneling in networks of NPs at low temperatures where experiments are usually performed significantly decreases 2D WL effect [10]. Third, NPs have a very small tunnel junction cross-section area that makes it extremely difficult to decrease the TJRs. Finally, for a significant decrease in the TJRs, the gap between NPs must be in the sub-nanometer range.

In order to overcome these challenges, we have used networks of few-layer graphene sheets (FLGSs). Due to metallic property of FLGSs they can be treated as 2D metals. Tunnel junction resistances are low because FLGSs are of the order of 4 μm and the tunnel junction cross-section areas can be very large.

Defects such as voids, lattice dislocations, and impurities in each FLGS can create a random potential distribution which can cause electron localization. Electrons are localized, because they experience different potentials and are reflected from the boundaries determined by defects. These regions where electrons are localized in are separated by tunnel barriers and have size distributions which can be treated as potential wells. If the TJRs between potential wells to be smaller than R_Q then electrons will be weakly localized otherwise SL can occur. The potential wells can have different Coulomb blockade (CB) energies (E_c is the electrostatic energy that an electron needs to tunnel between two neighboring NPs) as they have different sizes. Each FLGS can be also considered as a potential well because they are separated by TJs. Therefore, the electronic property of FLGS networks can be significantly affected by the TJRs inside FLGSs and the TJRs between FLGSs themselves.

In this work, first we show that the electronic property of FLGS networks are strongly affected by the TJRs and transition from SL to WL can be observed. In the SL limit, the TJRs between FLGSs and contact electrodes are high, the CB effect within FLGSs dominates and the network's resistance increases exponentially with decreasing temperature. In the WL limit, the TJRs between FLGSs are small and the CB effect does not play a major role and 2D WL effect is observed. The 2D WL effect mainly occurs inside of the FLGSs due to self-interference of the electron wave function and 2D WL effect of the networks similar to 2D disordered metal films cannot be observed as the minimum temperature of 5 K in our experiments significantly decreases the phase interference effect at larger distances because of the inelastic e–e interaction. By measuring the MR of the networks at different temperatures over a wide bias voltage range, we show that the positive MR which is observed in the SL limit arises from Zeeman splitting of the electronic states at the Fermi level of FLGSs and the negative MR that is observed in the WL limit is indeed due to phase interference effect inside of the FLGSs.

2. Experimental setup and measurement procedure

Fabrication of the FLGSs was carried out in an inductively coupled plasma-assisted chemical vapor deposition reactor (ICP CVD, 13.56 MHz, 1.0 kW) depicted in Fig. 1(a) and (b). For

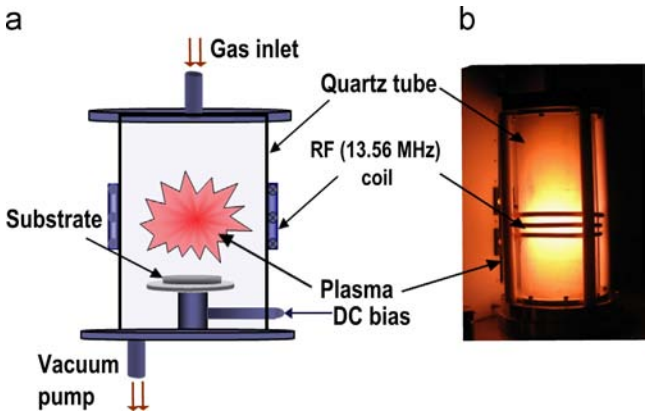

Fig. 1. (a) Schematic shows the substrate-plasma configuration in the inductively-coupled plasma CVD reactor for deposition of the FLGS networks. (b) An optical photo shows the plasma discharge inside the reactor during the deposition process.

sample S1, a polycrystalline Cu foil (99.99% purity) was used as a substrate where it was loaded into the reactor chamber and the chamber was pumped down to 9.2×10^{-4} Pa. Then, H_2 gas was injected into the chamber, the substrate was also negatively biased at −25 V DC and the plasma was generated at a chamber pressure of 2.5 Pa using 400 W RF power. After 5 min of H-plasma treatment, a mixture of CH_4, Ar and H_2 gas was injected at a ratio of 1.5:1:1 and RF power was increased up to 800 W while keeping the chamber pressure constant. The substrate temperature increased up to 300 °C due to the plasma-surface interactions that lead to the additional surface heating. The process was carried out for 6 min.

To investigate the effect of TJRs on the conduction mechanism of the network, as-deposited FLGSs film was transferred from a native Cu substrate to a polyethylene terephthalate (PET) substrate in a de-ionized water-assisted process (sample S2). The film detached from Cu substrate surface and floated on water when the substrate was dipped into water. Thus, the chemical-free process eliminates any contamination-effects on the measurements. The details and mechanisms of this transfer process will be reported elsewhere.

Sample S3 was prepared by growing FLGSs on a thermally oxidized (500 nm thick oxide-layer) silicon wafer. Two different experimental plasma processes were conducted to deposit FLGSs on SiO_2 and Cu substrates. Nonetheless, in both processes, the plasma-substrate configuration was the same. For sample S3, the SiO_2 substrate was subjected to 5 min of Ar-plasma treatment at chamber pressure of 2.5 Pa using 500 W RF power. Then, a mixture of CH_4, Ar and H_2 gas was injected at a ratio of 4:4:1 into the chamber, the substrate was negatively biased at −25 V DC and the plasma was generated at a chamber pressure of 2.5 Pa using 500 W RF power, the process took 6 min to complete.

Fig. 2(a)–(d) shows the scanning electron microscope (SEM, Zeiss Ultra plus, 30 kV) images of FLGSs. The FLGSs are few microns long and are very thin (of the order of nm) and grow vertically above the substrate which their height is ~4 μm. Transmission electron microscope (TEM, JEOL 3000F, 300 kV accelerating voltage) images in Fig. 2(e)–(f) shows FLGSs consist of typically 3–8 graphene layers. More details regarding the fabrication and characterization of FLGSs can be found somewhere else [11].

Fig. 3 shows the Raman spectra of samples (Reinshaw *inVia*, 633 nm laser excitation). The graphitic G-peak is originated due to first-order Raman scattering by doubly degenerate E_{2g} phonon mode at Brillion zone centre, while the 2D-peak is associated to the second-order phonon scattering at the zone

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