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Long-Wavelength Local Density of States Oscillations Near Graphene Step Edges

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Using scanning tunneling microscopy and spectroscopy, we have studied the local density of states (LDOS) of graphene over step edges in boron nitride. Long-wavelength oscillations in the LDOS are observed with maxima parallel to the step edge. Their wavelength and amplitude are controlled by the energy of the quasiparticles allowing a direct probe of the graphene dispersion relation. We also observe a faster decay of the LDOS oscillations away from the step edge than in conventional metals. This is due to the chiral nature of the Dirac fermions in graphene.

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Defects, impurities and edges have a large effect on the density of states in metals and semiconductors. Their spatial extent is usually limited by screening which suppresses the long-range behavior of the potential. However, at the Fermi wave vector \( k_F \) the electron gas has a singularity leading to strong and long-range oscillations in the electron density known as Friedel oscillations. Friedel oscillations have been observed near step edges on the surface of many metals such as Cu [1] and Au [2]. These materials are described by Fermi liquid theory and the oscillations have a characteristic wavelength given by twice the Fermi wave vector and a slow power law decay with distance. Graphene is a two-dimensional material where the quasiparticles obey the Dirac equation and therefore behave as massless Dirac fermions [3]. Many unusual effects ranging from a different quantization of the quantum Hall effect [4,5] to the suppression of backscattering [3] have been observed in graphene. These effects are due to the chiral nature of the quasiparticles in graphene as well as the linear dispersion relation. These two properties also affect the Friedel oscillations and are expected to cause a faster decay with distance [6].

There have been several scanning tunneling spectroscopy studies of graphene on SiC [7,8] and SiO\(_2\) [9,10] which have shown interference patterns due to quasiparticle scattering. These studies have observed standing wave patterns due to scattering from defects in both monolayer and bilayer graphene and verified that the chiral nature of the quasiparticles leads to the suppression of backscattering for monolayer graphene [8]. There has also been a topographic study of the effect of an armchair edge on the density of states in graphene on SiC which shows an oscillation along the C-C bonds that does not depend on energy [11]. However, none of these studies has observed a long-wavelength oscillation in the density of states controlled by the energy of the quasiparticles along with its spatial decay. In this Letter, we use scanning tunneling spectroscopy to directly probe the local density of states (LDOS) near step edges in monolayer graphene. We observe a long-wavelength oscillation of the LDOS due to scattering from the step edge. The wavelength of the oscillation is controlled by the energy of the quasiparticles. We also observe a faster decay \( (y^{-3/2}) \) of these oscillations as compared to noble metals \( (y^{-1/2}) \) due to the chiral nature of the quasiparticles in graphene.

The measurements were done in an ultrahigh vacuum, low-temperature STM (Omicron LT-STM) that was cooled to 4.5 K. In order to observe the long-wavelength standing waves in the density of states, a very flat surface with few scattering centers other than the step edges was needed. Exfoliated graphene on SiO\(_2\) has about a 1 nm roughness due to the roughness of the underlying oxide [12,13]. In order to have exfoliated graphene laying flat on a substrate we used hexagonal boron nitride (hBN) as a substrate. The sample was fabricated in a similar manner to previous graphene on hBN work [14,15]. Step edges in hBN occur naturally during the sample preparation procedure. The graphene lies flat on the hBN causing steps in the hBN to be transferred to the graphene.

Figure 1(a) shows a schematic of the experimental setup. The graphene flake is held at ground and the voltage on the tip is changed with respect to it. The tunnel current is measured from the tip to the sample. Figure 1(b) shows a 100 \( \times \) 100 nm\(^2\) topography image with a step edge running almost horizontally. The step height is about 0.5 nm. On the higher portion [upper portion of Fig. 1(b)] of the step edge, a 10 nm moiré pattern can be seen. This is due to the lattice mismatch and rotation between the graphene and the underlying hBN as was reported earlier [15,16]. Figure 1(c) shows a zoom-in on the lower portion [bottom of Fig. 1(b)] also showing a moiré pattern, but with a shorter \( \sim \)2 nm period. The different moiré patterns are a clear signature of a relative rotation between the graphene and hBN lattices. An atomically resolved image [Fig. 1(d)] across the step edge shows that the graphene is continuous and therefore the step edge is formed in the hBN layer.
To study the influence of the step edge on the electronic properties of graphene, we have performed scanning tunneling spectroscopy measurements over the area near the step edge. In these measurements, we recorded \( dI/dV \), which is proportional to the LDOS, as a function of position and energy. The tip height was first stabilized with a tip voltage of \(-0.3\) V and 100 pA. Then the feedback circuit was turned off and a 5 mV ac voltage at 563 Hz was added to the tip voltage. \( dI/dV \) as a function of tip voltage was recorded using lock-in detection in order to create images of the LDOS. Figures 2(a)–2(f) are maps of \( dI/dV \) for different tip voltages acquired in the region shown in Fig. 1(b). Two interesting features are noticed. First, LDOS oscillations in the lower portion are present and they decay moving away from the step edge. There are also LDOS oscillations in the upper portion but the signal is significantly weaker. Second, the wavelength of the oscillations increases as the tip voltage decreases approaching the Fermi energy and Dirac point.

LDOS oscillations have been previously observed on Cu [1] and Au [2] surfaces. However, there is an obvious difference from our case. The wavelength of standing waves on noble metals is an order of magnitude shorter compared with that of graphene (1 nm compared to 10 nm). We will see later that this difference comes from the different band structures. Long-wavelength LDOS oscillations have been observed in other systems where the Fermi surface is not near the \( \Gamma \) point [17,18]. However, the chiral nature of the quasiparticles and suppression of backscattering in graphene leads to a different decay behavior of the oscillations as compared to these other materials. Recently, similar effects were also reported on topological insulators [19,20] and explained by theoretical calculations [21]. Since both graphene and topological insulators are Dirac systems, they share a similar linear dispersion relation and chirality of electrons.
We adapt the theory for topological insulators [21] to our case of graphene. A step edge in graphene due to curvature can produce scalar and vector potentials which act as scatterers for electron waves [22]. We define the direction along the step edge as the $x$ axis [See Fig. 2(a)]. Consider an incident electron with wave vector $\vec{k} = \vec{k}_{\text{Dirac}} + \vec{k}'$ where $\vec{k}_{\text{Dirac}}$ is the crystal momentum at the Dirac point and $|\vec{k}'| = k$ is the radius of the constant energy circle in the Dirac cone. This electron will be partially reflected to a wave vector $\vec{k}' = \vec{k}_{\text{Dirac}} + \vec{k}''$. The electron will remain in the same Dirac cone as long as the step edge is not atomically sharp [the height of the graphene in Fig. 1(d) changes over a distance of about 2 nm] and intervalley scattering is forbidden. A scattering barrier running in the $x$ direction conserves the $x$ component of $\vec{k}$ in the scattering process. So if $\vec{k} = \vec{k}_x + \vec{k}_y$ then $\vec{k}' = \vec{k}_x - \vec{k}_y$ in order to conserve energy and momentum [see Fig. 3(a) for illustration]. The incident and scattered wave interfere with each other, and give rise to a spatially modulated electron density. So far, we have only considered a specific pair of wave vectors, $\vec{k}$ and $\vec{k}'$. In the actual scattering process, there are a range of wave vectors which are possible. All of the possible incoming and scattering wave vectors $\vec{k}$ lie on a constant energy circle (CEC) determined by the tip voltage. In order to find the total change in the electron density, we must integrate the contributions from all of them, which gives,

$$\rho(E, y) = \int_{\text{CEC}} |\Psi(k_x, k_y)|^2 dk,$$

where $\Psi(k_x, k_y) = \frac{\psi(k_x + r, k_y) + \psi(k_x - r, k_y)}{\sqrt{1 + |r|^2}}$ and $r$ is the reflection coefficient. Since the electrons in graphene are Dirac fermions they obey the 2D Dirac equation, and the wave function in momentum space near the Dirac point is given by

$$\psi(k_x, k_y) = \frac{1}{\sqrt{2}} \left( e^{i\theta_0/2} e^{i(k_x - k_y)} \right),$$

where $\theta_0 = \arctan(\frac{k_y}{k_x})$. After reflecting off the barrier, the phase factor changes to $\theta' = \pi - \theta_0$. Then the spatially varying portion of the density of states is given by

$$\delta \rho(E, y) = \frac{4r}{|r|^2} \cos(2k_y, y) \sin(\theta_0) dk.$$

For large distances, the integrand varies rapidly and the only points that contribute are when $k_y$ is approximately constant. This occurs if $k_y$ is near its maximum value and hence the scattering is at nearly normal incidence [see Fig. 3(a)]. If the step edge is modeled as a potential barrier, then the reflection coefficient is given by $r \propto \sin(\theta_0)$ for small $\theta_0$ [21]. Using the method of stationary phase, the integral can be evaluated. Making the substitution, $E = h\nu_F k$ where $\nu_F$ is the Fermi velocity, we obtain an expression for the variation in the LDOS as

$$\delta \rho(E, y) \propto \frac{\cos(2\nu_F y - 3\pi/4)}{(E^y)^{3/2}}.$$  \hspace{1cm} (1)

This expression describes the LDOS as a decaying oscillation with minima spaced by $\pi \nu_F / E$. Since we only probed energies near the Dirac point, which means the radius of the CEC and hence $E$ is small, we get long-wavelength LDOS oscillations compared with those from noble metals, where $E$ is usually much larger. Another interesting feature of this expression is the decay rate. In noble metals, the LDOS oscillations decay with distance from the step edge as $1/\sqrt{y}$ [1]. However, due to the pseudospin degree of freedom in graphene, backscattering is forbidden. This gives a faster, $1/y^{3/2}$ decay rate to the LDOS oscillation.

Now we compare the experimental data with the theoretical prediction. Figure 3(b) plots $dI/dV$ as a function of tip voltage and distance away from a step edge. The data are acquired by averaging, in the $x$ direction, all of the data in the two-dimensional images in Fig. 2, leaving a one
In conclusion, we have demonstrated LDOS oscillations near step edges in graphene on hBN. These oscillations are long wavelength and are controlled by the energy of the quasiparticles. The decay of the oscillations is much faster than in Fermi liquids and is a direct consequence of the chiral nature of the quasiparticles in graphene.

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