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STM/STS measurements of two-dimensional electronic states trapped around surface defects in magnetic fields

Y. Niimi, T. Matsui, H. Kambara, Hiroshi Fukuyama*

Department of Physics, University of Tokyo, 7-3-1 Hongo Bunkyo-ku, Tokyo 113-0033, Japan

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Abstract

The local density of states (LDOS) near point defects on a surface of highly oriented pyrolytic graphite (HOPG) was studied at very low temperatures in magnetic fields up to 6 T. We observed localized electronic states over a distance of the magnetic length around the defects in differential tunnel conductance images at the valley energies of the Landau levels (LLs) as well as relatively extended states at the peak ones of LLs. These states appear mainly at energies above the Fermi energy corresponding to the electron LL bands. The data suggest that the quantum Hall state is realized in the quasi two dimensional electron system in HOPG. At the peak energy associated with the n = 0 (electron) and -1 (hole) LLs characteristic of the graphite structure, a reduced LDOS around the defects is observed. The spatial distribution is almost field independent, which indicates that it represents the potential shape produced by the defects. $\bigcirc 2006$ Elsevier B.V. All rights reserved.

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Two-dimensional electron systems (2DESs) exhibit fascinating quantum phenomena at low temperatures and in high magnetic fields. The quantum Hall effect (QHE) is a well known example. Usually, these phenomena have been studied by means of transport measurements. Recently, it was demonstrated that the low temperature scanning tunneling microscopy and spectroscopy (LT-STM/STS) is a powerful tool to investigate 2DESs spectroscopically and microscopically. Morgenstern et al. [1] showed clear Landau quantization of the adsorbate-induced 2DES at the surface of InAs(110) with evaporated Fe submonolayers as well as complicated networks of the local density of states (LDOS) depending on bias voltage with LT-STM/ STS.

More recently, similarly clear Landau quantization of the quasi two-dimensional (2D) electrons and holes at a surface of highly oriented pyrolytic graphite (HOPG) was observed by STS at temperatures below 100 mK [2]. The

fax: +81 03 5800 6791.

E-mail address: hiroshi@phys.s.u-tokyo.ac.jp (H. Fukuyama).

measured tunnel spectra were quantitatively consistent with the calculated surface LDOS for graphite with a finite thickness of several tens of layers. This indicates that the electronic state in HOPG has a much stronger 2D nature compared to bulk graphite, likely due to a higher concentration of stacking faults. In transport measurements [3,4], a plateau structure was observed in the Hall resistance for HOPG samples in the quasi quantum limit. These experimental results altogether suggest the quantum Hall state (QHS) in HOPG, which is predicted for a simple 2D graphite sheet [5].

In this report, we show visualization of the possible localized (drift) and extended states of the QHE at a surface of HOPG by the ultra-low temperature STM/STS (ULT-STM/STS). At energies above the Fermi energy $(E_{\rm F})$, a series of differential tunnel conductance (dI/dV) images taken at the valleys and peaks of the Landau level (LL) spectra show distinct alternation of the localized and extended distributions of LDOS, respectively. At the valleys of LLs, the LDOS is trapped circularly around surface defects with a radius comparable to the magnetic length and this radius increases with decreasing field as

^{*}Corresponding author. Tel.: +81338122111x4184;

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expected. The results seem to support the above mentioned hypothesis of the QHE in HOPG and give insight into the microscopic nature of electronic states in the QHE. On the other hand, at the valleys and peaks of LLs in the negative bias range such behavior is not observed. This suggests that the QHS in HOPG originates from the quasi 2D electron LL bands. The dI/dV images at the peak of LLs (LL0, -1) characteristic of the graphite lattice structure show the spatial extent of the electrostatic potential produced by the defects.

The experiments were carried out using a recently constructed ULT-STM at temperatures below 30 mK and in magnetic fields up to 6 T [6]. The HOPG sample [7] was cleaved in air and then loaded into an ultra high vacuum chamber ($P < 2 \times 10^{-8}$ Pa) of the ULT-STM. The dI/dV curves and images were measured by the lock-in technique (f = 412 Hz, $V_{mod} = 1$ mV).

Fig. 1 shows an STM image obtained near point defects at the HOPG surface. A $\sqrt{3} \times \sqrt{3}$ superstructure in the vicinity of the defects and a three-fold symmetric electron scattering from the defects can be seen. The former is generally observed near defects on graphite, such as step edges, deposited metal adatoms, and grain boundaries [8]. The latter was predicted for a 2D graphite sheet with a small number of point defects [9]. Although we cannot extract the detailed defect structure (number, kind, position, etc.) from the image, the similarity between the STM image and the theoretical prediction indicates that only a few point defects are involved in the three-fold scattering and the superstructure.

The closed symbols in Fig. 2 show Landau quantization of the quasi 2D electrons and holes far away (>30 nm) from the defects. At B = 6 T, several pronounced peaks appear both at the positive and negative bias voltages. The amplitude of the peaks decreases with decreasing field and the peaks vanish as the magnetic field goes to zero. The oscillation in these LL spectra is more pronounced with finer structures than that observed for single crystal graphite [2]. This suggests that HOPG has a much stronger 2D nature due to much higher concentration of stacking faults [2,10]. Actually, this is confirmed by the calculated



Fig. 1. STM image in the vicinity of point defects ($6 \times 6 \text{ nm}^2$, I = 0.2 nA, V = 180 mV, T = 30 mK).



Fig. 2. Tunnel spectra taken near the defects (open symbol) and far away (>30 nm) from them (closed symbol) at B = 6, 3, and 0 T (I = 0.2 nA, V = 180 mV, T = 30 mK). Each spectrum is vertically shifted for clarity. The data are averaged over $10 \times 10 \text{ nm}^2$.

LDOS for graphite with various finite numbers of layers [2]. In addition to the field dependent peaks, we also observed a nearly field independent peak slightly above E_F by 20 meV. This peak originates from the n = 0 (electron) and -1 (hole) LLs which are characteristic of graphite. Other usual LLs are labeled as LL1, LL2, LL3, ..., with increasing energy. In zero magnetic field, a peak structure is seen at a bias voltage of -10 mV. It is attributable to the electrostatic potential caused by the STM tip, as was already reported in STM/STS works on semiconductor [1,11] and semimetal surfaces [2].

In the vicinity of the point defects, we obtained qualitatively different tunnel spectra depending on bias voltage. At the peak energies of the LLs, there is no difference between the LDOS near and far from the defects. On the other hand, at the valleys of the LLs in the positive voltage range the LDOS near the defects is higher than that far from the defects. The difference of the LDOS vanishes with increasing energy. This is presumably because the electrons with higher energies than the defect potential are no longer confined. At negative bias voltages, such a clear difference was not observed except for the tail of the LL at an energy of V = -30 mV at B = 6 T.

The dI/dV images near the point defects at the three distinct energies are shown in Figs. 3(a)–(f) for B = 6 and 3 T. At the valley energies in between the LL2 and LL3, circular distributions of the LDOS are observed around the

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