Extended Abstracts of the 2010 International Conference on Solid State Devices and Materials, Tokyo, 2010, pp83-84

F-1-1(Invited)

STS Observations of Topological Dirac Fermion on Graphite Surfaces

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The unique characters of Dirac fermion (DF) start to attract a big interest, since the experimental success to fabricate Graphene, a monolayer sheet of graphite.[1,2] Different from the conventional electron and hole, to say Schrödinger fermion (SF), which can be described with Schrödinger equation, DF can be described with Dirac equation including relativistic properties. Especially in the case of graphene, the effective mass of DF is zero and it has a linear energy dispersion, called Dirac corn, crossing at Dirac point. Recently, some other materials are also paid many attentions because of their topologically appeared DFs.[3]

Graphite is a well known quasi two-dimensional (2D) material and one of the good candidates which have the topological Dirac fermion (TDF). The Fermi surface of graphite is located along the Brillouin zone edge (*H*-*K*-*H*), and it shows linear band dispersion around *H* point like massless DFs, while it is parabolic around *K* point like SFs. Such band structure of graphite had long been discussed theoretically [4] and confirmed by angle resolved photo emission spectroscopy (ARPES) rather recently.[5] Since the wave function along k_z direction has a node at *H* point, the carriers at *H* point can be localized at surfaces, while the ones at *K* point cannot. Consequently, the TDF at *H* point can be expected to appear on the surface of graphite.

In magnetic field, the electrons are quantized in a plane perpendicular to the field. Therefore, 2D electrons would have quantized energy levels known as Landau levels (LLs). Typical field (B) and Landau index (n) dependencies of LLs are calculated for SF, massless DF and massive DF of bi-layer graphene as $E\mathrm{n} = (n+1/2)\hbar eB/m \propto nB$, $E_{\mathrm{n}} = \sqrt{2e\hbar v_{\mathrm{F}} nB} \propto \sqrt{nB}$, $E_{\mathrm{n}} = \sqrt{n(n+1)}\hbar eB/m \propto \sqrt{n(n+1)}B$, respectively. Previously, we have shown that the Landau quantisation, a character of 2D electrons, can be observed on the surface of graphite with scanning tunneling spectroscopy (STS).[6] At that time, we have measured the dI/dV spectra for two kinds of graphite, i.e., Kish graphite and HOPG (highly oriented pyrolytic graphite). The former is a single crystalline graphite grown as precipitation from molten iron, while the latter is a poly-crystalline graphite with ordered *c*-axis orientation. Therefore, HOPG contains more stacking fault which decouples the interlayer interaction of graphite. In the dI/dV spectra, many peaks are appeared in magnetic field, while it shows a V-shape patern around Fermi energy (E_{F}) in zero magnetic field. There are more peaks and the structure is more complicated in HOPG than in Kish graphite. Such peak structure can be reproduced very well with a theoretical calculation by considering the DOS of the surface layer of graphite with finite thickness, and some surface potential φ corresponding to an electro-static potential of the STM tip. It clearly shows that the dI/dV spectra measured on Kish graphite and HOPG can be explained with surface DOS of graphite with infinite thickness and 40 layer thick, respectively. It is worth noting that this study also succeeded to show that the dI/dV spectrum with STS is comparable to the DOS on surfaces, and that each peak in spectra corresponds to the Landau level on the surface.

The field dependence of the surface LLs had been analyzed in detail and found three levels with different magnetic field dependence in ref. 7. They measured the surface LLs on HOPG and suggested that one kind has $\sqrt{n(n+1)}B$ dependence like bilayer graphene, one has $\sqrt{nB}$ dependence like graphene, and the other is hard to define any field dependencies. At first glance, this result is consistent to the previous conclusion in ref. 6 that HOPG is effectively thinner than Kish graphite and has stronger 2D tendencies. However, they showed only one set of the field dependence of the surface DOS and it was not clear enough if it is a character of HOPG or of graphite in general. To clarify this point, we have analyzed the surface LLs for various graphite surfaces and discussed their behavior theoretically.

In addition to the previously studied HOPG and Kish graphite, we also studied about ZYX graphite in this study. It is an exfoliated graphite made by careful exfoliation of HOPG and subsequent recompression. It can be expected to contain more stacking faults and, therefore, has thinner effective thickness among the graphite studied here. Actually, we can find more peaks in dI/dV spectra on ZYX graphite than on HOPG and Kish graphite. The dI/dV spectra for three HOPG samples and one ZYX graphite and one Kish graphite are measured in magnetic fields up to 6 T at very low temperatures.[8]

The similar B and n dependencies as were reported in ref. 7 are observed for one of the HOPG samples as shown

in figure 1. However, it was the only sample which apparently showed these three characters and no clear field dependencies could be found for other HOPG samples and for ZYX graphite. Even though it may possible to say some field dependencies for some of the peaks, it is hard to find any Landau index dependencies.

In the case of Kish graphite, on the other hand, the situation is different. Besides the n = 0, -1 level of remarkable peak, two kinds of peaks can be identified even though they are rather faint. The peaks in higher energies are energetically well isolated, while they are well packed in lower and only in negative energies. They are found to have $\sqrt{B}$ and B dependencies, respectively. Moreover, former peaks are proportional to $\sqrt{nB}$ with coefficient with v_F of 1.03 x 10^6 m/s, which is comparable to the massless DF in graphene. Concerning the latter levels, however, they seem to have $\sqrt{n \cdot B}$ dependence, but there are no reasonable understanding for such behavior, yet.

These experimental results seem to conflict to the previous results in ref. 6 which suggests that HOPG is thinner and, therefore, closer to graphene, and the Kish graphite is infinitely thick. However, this is not the case. This is happening because we measured the surface state which reflects the underneath electronic properties.

To understand this behavior, the surface DOS on graphite with infinite thickness was calculated by the same way used in ref. 6. In this case, magnetic fields up to 10 T are took into account and the surface potential φ is not included to consider the pure surface state of graphite. In the calculated surface DOS, the peaks are appeared similar to the experimentally observed dI/dV spectra, namely there are n = 0, -1 level, levels in higher energies, and in lower and negative energies. This structure of LLs can be roughly understood from the Landau sub-band of bulk graphite in magnetic fields. The LLs at K point is energetically equi-separated with smaller separation. On the other hand, the LLs at H point are grown away from E_F and exist in higher energies. In addition to K and H point, the band is bended in between K and H point, and the maximums make peaks in negative energies in DOS. From such bulk band structure, we can expect that the peaks in higher energies come from H point and the ones in lower negative energies from either K point or a band bending state. Actually, and the same as experiments, the peaks in higher energies clearly show $\sqrt{nB}$ dependence. In this case, v_F is estimated to be 1.01 x 10^6 m/s. Therefore, we can again refer them as levels at H point which behave as massless DF on graphene. However, different from the experiment, no clear B and n dependencies could be found for the peaks in lower negative energies. Since the levels close to E_F are strongly affected by the surface potential φ, the B dependence could be observed after the effect of φ. The origin of these peaks is not clearly understood yet at this stage.

The fact that the massless DF is observed on the surface of graphite and that it is originally the property of H point in Brillouin zone can be further confirmed by calculating DOS on each layer and the LLs at certain k_z.

First, DOS on 1st to 4th layer from the surface are calculated. It shows that the levels of massless DF appear on 1st and 3rd layers. Considering the two layer periodicity of graphite, these peaks of massless DF can be confirmed to be originated from the wave functions with $2c_0$ periodicity. This behavior clearly shows that the massless DF is appeared every second layers and especially isolated on the surface and that it comes from the H point. On the layers deeper than 2nd, peaks with small separation appear, and the amplitude of these peaks becomes larger in deeper layers. Afterwards, the former peaks are almost buried under the latter. In this way, the DOS structure approaches to that for the bulk state.

Second, the LLs of bulk state at certain k_z are calculated. It clearly shows that the LLs at K point changes linearly against magnetic field, and the energy separation of levels are rather small. When k_z approaches to H point, Such linear field dependence starts to be destroyed in higher energies and in higher magnetic fields. The level separation is getting larger, and the levels show $\sqrt{B}$ dependence finally at H point. This fact also suggests that the $\sqrt{B}$ dependent levels come from H point.

In summary, we have measured the dI/dV spectra on different kinds of graphite and showed that the feature of massless DF can be appeared topologically on the surface of graphite. This behavior can be well understood by theoretical calculation. It shows that graphite has TDF originated from H point and it appears on the surface.

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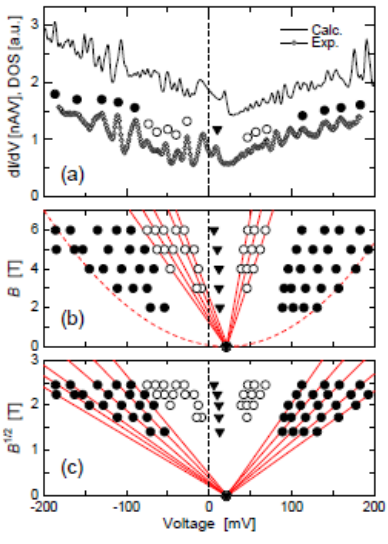

Fig.1 (a) The dI/dV spectrum observed on one of the HOPG samples, and the surface DOS calculated for a graphite with 40 layers thick. A peak with filled triangle corresponds to n = 0, -1 level, and peaks with open/closed circles show linear and square-root dependence against magnetic field (B) as shown in (b) and (c). Peaks which are not marked show no clear B dependence.