論文内容の要旨

論文題目: Studies of Two-Dimensional Electron System at Graphite Surfaces by Scanning Tunneling Spectroscopy

(走査トンネル分光法によるグラファイト表面の２次元電子系の研究)

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Two-dimensional electron systems (2DESs) exhibit fascinating quantum phenomena at low temperatures. The quantum Hall (QH) effect is a well known example. The essence of the integer QH effect is alternating extension and localization of a two-dimensional electron gas (2DEG), which is realized when the Fermi energy (E_F) is tuned to one of the Landau levels (LLs) and in between the adjacent LLs, respectively. In the localized states, it is commonly believed that the 2DEG is trapped around impurities or sample edges running along the equipotential lines with an approximate width of the magnetic length l_B ($= \sqrt{\hbar/eB}$ where B is magnetic field). However, it is generally difficult to observe such localized states in real space at the nanometer scale with local probes, as the 2DESs are usually formed at heterojunctions one hundred nanometers below the semiconductor surface.

Graphite is another candidate to study 2DESs with a small effective mass and a relatively high electron mobility. Unlike the semiconductor heterojunctions, it has a quasi 2DEG even at the surface. This feature is suitable for scanning tunneling spectroscopy (STS) which is a powerful tool to investigate the local density of states (LDOS) at sample surfaces microscopically and spectroscopically. Recent STS [1] and transport [2] measurements revealed that the two-dimensional (2D) nature of the electronic properties for higly oriented pyrolytic graphite (HOPG) is much stronger than that for bulk (single crystal) graphite because of its much higher stacking-fault density. The observed Hall resistance plateau [2] indicates the probable QH state in this material. More recently, the distinct QH plateaus have been observed in the 2DESs at a single layer of graphite (so-called "graphene") as well as bilayer graphene [3]. Therefore, the localized and extended states of the QH effect can be investigated in the thin-layer graphite systems including HOPG with the STS techniques.

In this thesis, we have studied the LDOS of the quasi 2DES near defects and step edges at graphite surfaces using an ultra-low temperature scanning tunneling microscope (STM). We have succeeded in the first clear visualization of the alternating localized and extended states near defects in the 2DES at the

HOPG surface in magnetic fileds, which indicates the possible QH state at the surface. In the vicinity of the zigzag step edges, theoretically predicted graphite edge state have been observed for the first time. We have also investigated the LDOS of the 2DES formed at surfaces of epitaxially grown InAs thin films.

In Figs. 1(a)-(c), we show differential tunnel conductance (dI/dV) curves and images obtained around point defects at the HOPG surface at $B = 6$ T. The dI/dV images, i.e., LDOS mappings, reveal a clear contrast between localized and extended spatial distributions of the LDOS at the valley and peak energies of the LL spectrum, respectively. The localized electronic state has a single circular distribution around the defects with a radius comparable to l_B. These results indicate that the QH state is realized at the HOPG surface.

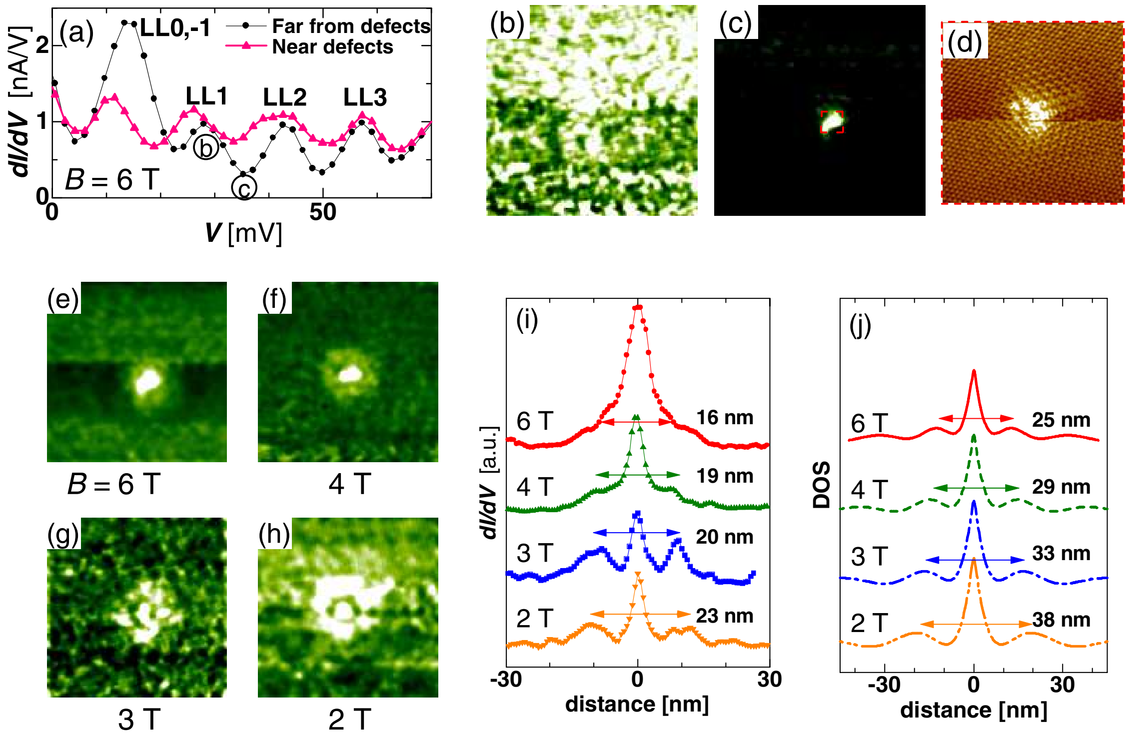

Figure 1: (a) Tunnel spectra averaged over 20 × 20 nm^2, centered on point defects (triangle) and far away (> 30 nm) from them (circle) at $B = 6$ T and $T = 30$ mK. (b), (c) dI/dV images (80 × 80 nm^2) around the defects at (b) the peak and (c) valley energies of the LLs. (d) Higher resolution STM image of the point defects (8 × 8 nm^2). (e)-(h) The magnetic field dependence of dI/dV images (80 × 80 nm^2) near the defects at the valley energies in between LL1 and LL2; (e) 6 T, (f) 4 T, (g) 3 T, (h) 2 T. (i) Typical cross sections of the dI/dV images in the different fields. (j) The field dependence of calculated LDOS at the valley energies in between LL1 and LL2 in the $1/r$ potential.

Figures 1(e)-(h) show the LDOS distributions near the defects at the valley energies in between LL1 and LL2 in several different magnetic fields. The localized LDOS has a maximum just on the defects and a satellite around them with a radius comparable to l_B (Fig. 1(i)). The observed LDOS distributions are in good agreement with the calculated LDOS for a single electron in 2D in a Coulomb potential in magnetic fields (Fig. 1(j)). These results reveal that the functional form of potential plays an important role in the formation of the localized states.

In the vicinity of graphite edges, a peculiar localized state was predicted even at zero magnetic field from the tight-binding band calculations [4]. This localized state is known as the "graphite edge state". It stems from the topology of the π electron networks at the zigzag edge and does not appear at the armchair edge. We carried out the LDOS measurements near monoatomic step edges of both the zigzag and armchair types on the graphite surface at zero field. With STS, a clear LDOS peak at several tens meV below E_F is observed on terraces with the zigzag edges but not on those with the armchair ones (Figs. 2(a) and (b)). This is the first clear spectroscopic observation of the "graphite edge state" theoretically predicted from the tight-binding calculations on graphene ribbons [4]. The existence of the LDOS peak near the zigzag edge is also confirmed by first-principles calculations.

In the STM measurements, two types of superstructures, i.e., $(\sqrt{3} \times \sqrt{3})R30°$ and honeycomb ones, are observed over 3$-$4 nm from both the zigzag and armchair edges (Figs. 2(c) and (d)). Calculations based on a density-functional derived nonorthogonal tight binding model show that the coexistence is due to a slight admixing of the two types of edges at the graphite surface.

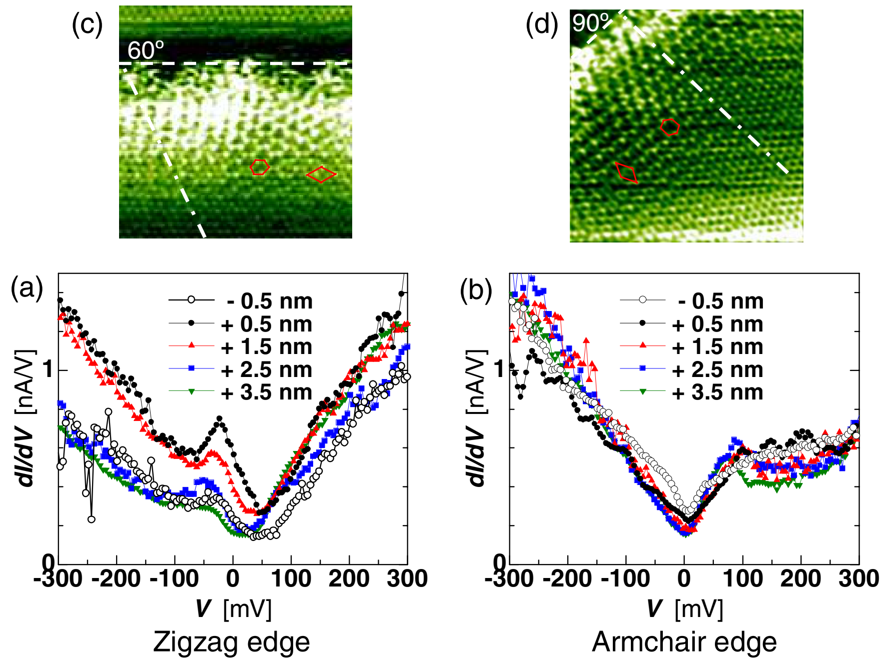

Figure 2: (a), (b) dI/dV curves measured at $|V| \leq 300$ meV near (a) the zigzag and (b) armchair edges at the graphite surface. The numbers denoted are distances from the edge ($d = 0$). (c), (d) STM images in the vicinity of (c) the zigzag and (d) armchair edges (6×6 nm^2). The dashed and dot-dashed lines show the edge and the atomic row of B-site atoms, respectively. The diamond and hexagon represent the $(\sqrt{3} \times \sqrt{3})R30°$ superstructure and honeycomb one.

The 2DES is also formed at the surface of an InAs thin film epitaxially grown on a GaAs substrate (InAs/GaAs) [5]. We studied the LDOS of the 2DES at the InAs/GaAs(111)A surface in magnetic fields. Several peaks associated with the Landau quantization of the 2DEG are observed in the tunnel spectra at negative bias voltages just above the subband energy of the 2DES (Fig. 3). On the other hand, a field-independent oscillation with a larger energy separation than the LL is observed above E_F. From the tunnel spectra for the InAs/GaAs(111)A, InAs/GaAs/GaAs(111)A and InAs/InAs(111)A, we found that this oscillation is associated with the transmission probability of the tunneling current through the Schottky barrier at the interface between the InAs film and the GaAs substrate.

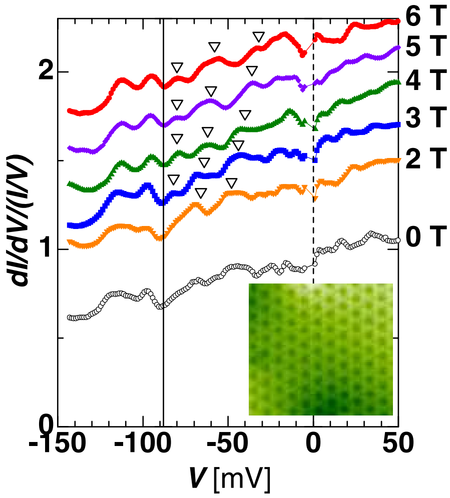

Figure 3: $(\mathrm{d}I/\mathrm{d}V)/(I/V)$ curves at the InAs/GaAs(111)A surface in several different magnetic fields. Each spectrum is vertically shifted for clarity. The vertical solid line represents the bottom of the 2D subband. The inset shows a typical STM image of the reconstructed InAs(111)A surface (7.5×6.8 nm^2).

In conclusion, we have measured the LDOS of the quasi 2DES near point defects and step edges at graphite surfaces with the STM/STS techniques. The LDOS mappings near the defects in magnetic fields show alternating localization and extension of the 2DEG depending on energy, which is consistent with the electronic states expected in the QH effect. The localized LDOS distributions at the valley energies of the LLs are semi-quantitatively explained by the calculated LDOS for the 2DEG in the $1/r$ potential in magnetic fields. In the vicinity of the zigzag edges, we have observed a clear peak in the tunnel spectra near E_F in zero field. This LDOS peak corresponds to the graphite edge state theoretically predicted from the tight-binding band calculations on graphene ribbons. The LDOS of the 2DES have also been investigated at surfaces of epitaxially grown InAs thin films, where the LLs of the 2DEG are observed in the tunnel spectra. The results obtained here can be achieved only by means of STM/STS, and play an important role in understanding the electron localization in 2D from the microscopic view point.

References

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