

Electronic structure of fully intercalated few-layer graphene probed by Raman spectroscopy

Da Zhan, Li Sun, Zhenhua Ni, Lei Liu, Xiaofeng Fan, Yanan Xu, Jiaxu Yan and Zexiang Shen*

Division of Physics and Applied Physics, Nanyang Technological University, 637371, Singapore
zexiang@ntu.edu.sg

Huge progress on graphene researches has been achieved in recent years including: Various methods for graphene fabrication; Discovery of its unique electronic, thermal and mechanical properties; Successful fabrication of prototype graphene-based devices. However, much of the unique properties of graphene are accorded to that of single layer graphene (SLG). It would be very desirable to modify few-layer graphene (FLG) samples so that they have similar properties as that of SLG. Graphite intercalation compounds (GICs) are complex materials that formed by insertion of atomic or molecular layers of different chemical species between graphite interlayer space.^[1] The interlayer distance of graphite is dramatically increased due to presence of intercalants, which would strongly affect the electronic coupling between graphene layers, hence changes its properties. Thus, graphene based intercalation compounds would be an efficient method to modify the properties of FLG.^[1-3] Until now, there are no experimental reports on few-layer graphene intercalation compounds (FLGIC) except the most recent report on Br₂ and I₂ intercalated FLG by Jung et al.,^[4] however, the FLG is still not fully intercalated according to their Raman spectra and the structural model.^[4]

In this presentation, we will report the fully intercalated FeCl₃-FLGIC which fabricated by two-zone vapor transport method. This is the first report on full intercalation for graphene samples. Fig. 1 shows the schematic crystal structure of FLGIC. The distance between adjacent graphene layers enlarged dramatically due to the insertion of FeCl₃. Furthermore, as FeCl₃ is an acceptor type intercalant, it gives rise to the strong charge transfer induced hole doping effect on graphene. The detailed information for FLGIC is investigated by Raman spectroscopy and mapping. For FLG, Raman G band properties are very sensitive to doping, while the 2D band properties quite dependent on the electronic band structure. Thus analyzing the evolution of G and 2D bands features from pristine FLG to FLGIC can give us a lot of information for probing the properties modification of FLGIC, such as Fermi level shift and band structure evolution. Fig. 2 shows the Raman spectra for graphenes (1 layer (1L) to 4 layers (4L)) after intercalation, which present obviously different Raman features compare with that of the pristine FLG. The features of the Raman G peak of such FLGIC are in good agreement with their full intercalation structures. The FLGIC presents single Lorentzian 2D peak, similar to that of single layer graphene, indicating the loss of electronic coupling between adjacent graphene layers. First principle calculations further reveals that the band structure of FLGIC is similar to single layer graphene but with strong doping effect due to the charge transfer from graphene to FeCl₃. Furthermore, the distribution of density states of FLGIC in k-space is strongly influenced because of the coupling with adjacent intercalant layer based on the observation of resonant Raman scattering for G peak.

FLGIC not only changes the physical structure from its precursor FLG, but also effectively modifies its properties, such as electronic structure and density of states. Therefore, the successful fabrication of FLGIC opens a new way to modify properties of FLG for fundamental studies and future applications.

References

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Figures

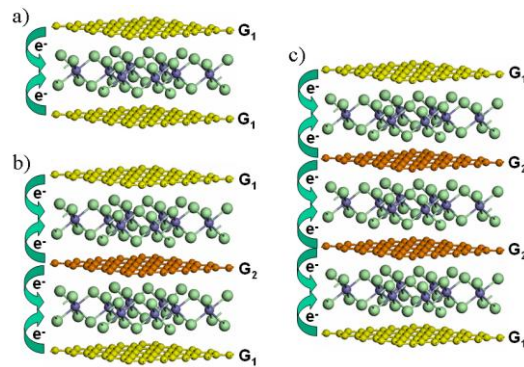


Figure 1

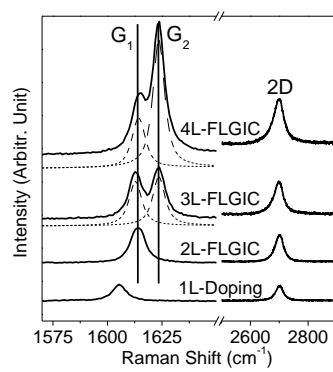


Figure 2

Figure caption

Figure 1. Schematic crystal structure of 2L- to 4L-FLGIC.

Figure 2. The Raman spectra of 1L doped graphene and 2L- to 4L-FLGIC.