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Study of correlation between electrochemical properties and density of states of graphene using field effect transistors

Graphene, a carbon material of a single atomic layer, has attracted attention for electrochemical applications such as batteries and sensors due to its high electrical conductivity, chemical and mechanical stability and high specific surface area. The electrochemical properties of graphene are determined by various factors. The density of states (DOS) nearby Fermi energy E_F has major effects on the electron transfer behaver in carbon-related materials that have lower DOS compared with metals.[1] However, general methods to change DOS of carbon materials influence other factors than DOS such as surface structures and impurity concentrations. We fabricated a field effect transistor (FET) with a graphene channel in order to investigate a correlation between electrochemical properties and DOS of graphene. Monolayer graphene films were grown by chemical vapor deposition (CVD) on copper foils and transferred onto SiO_2 (300 nm) /Si substrates. A back gate FET with Au electrodes was fabricated. The electrochemical properties of CVD monolayer-graphene films were evaluated by cyclic voltammetry (CV) measurements in 0.1 M potassium chloride solutions containing 1 mM hexa-ammine-ruthenium(III). The CV measurements were conducted under applying back gate voltages V_{bg} to modulate DOS ρ *(E)* (Figure 1). The reaction rate constants k^0 were estimated as a function of redox peak separations ΔE_p from cyclic voltammograms.[2] As shown in Figure 2, the k^0 values from the CV measurements increased when differences between V_{bg} and the Dirac point became larger. In other words, E_F located at energy levels where ρ *(E)* was higher. Thus, the heterogeneous electron transfer rate (HETR) at graphene surfaces became higher when ρ *(E)* nearby E_F became higher. This work, which combined research fields of electrochemistry and electronics, allowed us to reveal the correlation between HETR and DOS of graphene experimentally without changing parameters other than DOS. These results indicate that electrochemical properties of graphene strongly depend on DOS.

References

[1] R. L. McCreery, Chem. Rev., **108** (2008) 2646-2687
[2] R. J. Klinger and J. K. Kochi, J. Phys. Chem., **85** (1981) 1731-1741

Figures

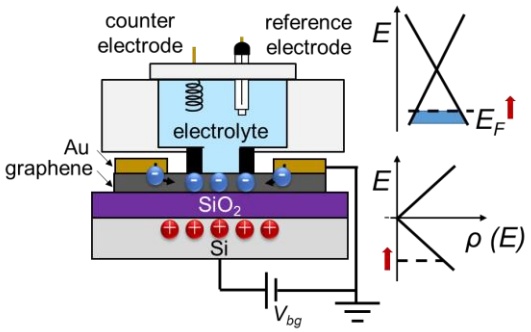

Figure 1: Schematics of cyclic voltammetry measurements under applying a back gate voltage and modulation of DOS.

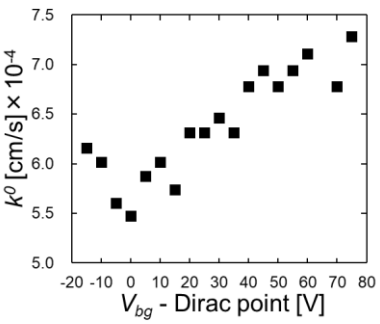

Figure 2: k^0 values as a function of V_{bg} – Dirac point.