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Carrier scattering in graphene caused by electron-transfer molecules

Since graphene has extremely large specific surface area, its electronic property is significantly affected by the surface condition. The high sensitivity to surrounding environment is profitable for gas- and bio-sensors [1]. On the other hand, the carrier mobility is lowered by the adsorption of water [2] or oxygen [3] molecules, which degrades superior transport characteristics of graphene. Thus, for the application of graphene devices in atmospheric condition, it is important to understand the interaction between graphene and adsorbed molecules.

So far, we have studied the effects of molecular adsorption on carrier density in graphene [4,5]. Graphene devices were prepared on SiO₂/Si substrates for two- or four-terminal conductance measurement. Various kinds of electron acceptor (EA) molecules (7,7,8,8-tetracyanoquinodimethane and its derivatives, and p-benzoquinone derivatives) with different sizes, symmetries, and redox potentials were deposited on the graphene devices in vacuum, and the conductance, σ , was measured against gate voltage, V_{bg} , in situ (Fig. 1). The hole density, n_{hole} , accumulated by the molecules was estimated from the shift of the charge neutrality point, ΔV_n , and the doping efficiency was defined as number of holes accumulated by one EA molecule. The doping efficiency was enhanced when the LUMO level of molecules, E_{LUMO} , was lower than the Fermi energy of graphene, E_F , as shown in Fig. 2. Considering the small density of states of graphene and the shift of E_F , the doping efficiency in the electron transfer process was reasonably explained [4].

In this presentation, we report the effect of molecular adsorption on the carrier mobility. In general, the carrier mobility decreased with depositing EA molecules. The inverse of the mobility increased linearly with n_{hole} as shown in Fig. 3, indicating that EA molecules work as charged impurities and Coulomb scatterers [6]. However, one EA molecule (2,3,5,6-tetrabromo-1,4-benzoquinone, QBr₄, indicated with red symbols in Fig. 3) did not decrease the mobility despite of accumulating holes. To interpret the result, the effect of charged impurities in a SiO₂/Si substrate was taken into account. The inhomogeneity of potential energy due to charged impurities in the SiO₂/Si substrate can be eliminated by molecular doping, when the molecule has E_{LUMO} slightly higher than E_F . Our result is important not only for passivation but also for improvement of graphene devices prepared on SiO₂/Si substrates. In the conference, we also discuss different doping effects depending on the symmetry of molecules.

References

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Figures

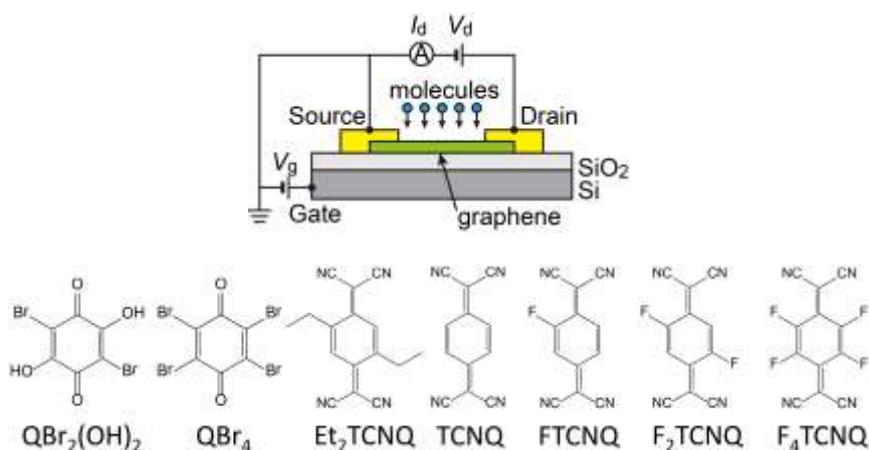


Figure 1: Schematic measurement system. Various EA molecules were deposited on monolayer graphene in vacuum, and two- or four-terminal conductivity was measured in situ. Structures and abbreviations of used EA molecules are indicated. The molecules are arranged in order of electron accepting strength, from the weakest $\text{QBr}_2(\text{OH})_2$ to the strongest F_4TCNQ .

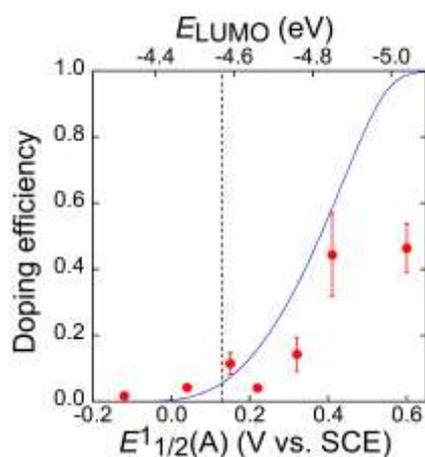


Figure 2: Doping efficiency as a function of redox potential, $E^{1/2(A)}$, and the LUMO level, E_{LUMO} , of molecules. A dashed line indicates E_F of graphene. A blue curve was doping efficiency calculated based on a simple electron-transfer model between graphene and molecules [4].

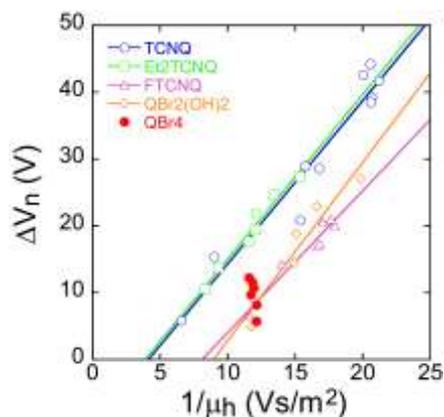


Figure 3: Relation between the inverse of mobility evaluated in the hole regime, $1/\mu_h$, and the shift of the charge neutrality point, ΔV_n , for various EA molecules. Note that the mobility of QBr_4 (red circles) does not change with increasing ΔV_n .