## Determination of mobility in top and bottom surfaces of multilayer graphene placed on SiO<sub>2</sub>/Si substrate

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We experimentally demonstrate that in a thick multilayer graphene sheet placed on a  $SiO_2/Si$  substrate, the mobility of the top surface, facing the vacuum, is about five times larger than that of the bottom surface, attaching to  $SiO_2$ , at room temperature. From the temperature dependence of the mobility, we argue that the dominant mechanism which deteriorates the mobility of multilayer and even single layer graphene is the Coulomb scattering originating from the charged impurities in the substrate.

The samples are dual-gated graphene devices with a suspended contactless top gate fabricated with a method reported in [1]. A highly-doped Si substrate covered with a SiO<sub>2</sub> layer was used as the back gate. We examined seven multilayer graphene (MLG) films with thickness t ranging from 0.9 to 5.3 nm, which were obtained by using mechanical exfoliation of kish graphite. We measured the top-gate voltage ( $V_{tg}$ ) dependence of the conductance with the back gate grounded ( $V_{bg} = 0$ ) and the  $V_{bg}$  dependence of the conductance with the top gate grounded at room temperature, 77 K and 4 K.

We estimate the mobility of the top surface and that of the bottom surface of MLG based on a simple model in which a MLG film is divided into three layers: the layers near the top (bottom) surface with thickness  $\lambda$  (layer A (C)) and the remaining central part (layer B), and we make two assumptions: 1) the mobility of layers A, B and C ( $\mu_t$ ,  $\mu_0$ , and  $\mu_b$ , respectively) are uniform in each layer, and 2) there is no change in the carrier density in each layer in the direction perpendicular to the surfaces. Under these assumptions,  $\mu_t$  ( $\mu_b$ ) is calculated from the slope of the conductance,  $dG/dV_{tg}$  ( $dG/dV_{bg}$ ), and sample dimensions.

We find that the ratio  $r = \mu_b/\mu_t$  as a function of t steeply decreases around  $t \sim 1$  nm and stays at a constant value of  $\sim 0.2$  in thick MLG. The ratio slightly increases with decreasing temperature, but is around 0.25 for thick films at 4.2 K, indicating that the major electron scattering of graphene occurs at the bottom surface, and it survives even at low temperatures. This means that the mobility of graphene placed on a substrate can be improved by removing the charged impurities in the substrate.

[1] G. Liu, J. Velasco, Jr., W. Bao, and C. N. Lau, Appl. Phys. Lett. 92, 203103 (2008).