Graphene, a single layer of graphite, has attracted both scientific and technological interests as a new two-dimensional (2D) electron gas system with extremely high mobility of ~10,000 cm² V⁻¹ s⁻¹ at room temperature (RT). The density of electrons and holes can be modulated by the gate electric field due to a low density of states of the graphene. Although there are many reports on electrical properties of the graphene on SiO₂, one problem is that the mobility varies widely from 2,000 to 20,000 cm² V⁻¹ s⁻¹, even though two-probe measurement results were excluded. Another problem is that the layer number dependence is still unclear, since current research focuses mainly on mono- and bi-layer graphene films. Monolayer and multilayer graphene films possess a linear dispersion and parabolic ones with the band overlapping, respectively. Monolayer graphene film is clearly distinguished from multilayer films by two-dimensional (2D) band around 2700 cm⁻¹ in the Raman spectrum. The layer number dependence of the spectra for multilayer graphene films, however, is not decisive, and the spectrum shape and position are also strongly affected by the doping. Atomic force microscopy is not suitable to determine the layer number due to an uncertain dead space between multilayer graphene films and SiO₂. Therefore, in order to systematically investigate electrical properties as a function of the layer number, a simple but reliable layer number determination method is required.

In the present study, we first show that the combination of optical microscopy and Raman spectroscopy can determine the layer number, and then electrical transport properties, especially, the mobility and Dirac point shift, are reported as a function of the layer number.

Monolayer and multilayer graphene films were transferred onto 90-nm SiO₂/p-Si substrates via the micromechanical cleavage of Kish graphite. After a suitable graphene film had been selected under the optical microscope, electron-beam lithography was utilized in order to pattern electrical contacts on the graphene film. Thermal evaporation of Cr/Au (5/25 nm) was followed by lift-off in warm acetone. To remove the resist residual, graphene devices were annealed in a H₂–Ar mixture at 300 °C for 1 h and then electrical measurements were performed in vacuum. Using a Hall-bar type electrode configuration (see the inset in Fig. 4(a)), Hall measurements were also performed.

Figure 1 shows the contrast of graphene as a function of SiO₂ thickness calculated using the Fresnel equation, assuming a trilayer model of graphene, SiO₂ and Si. This is basically the same as the calculation by Blake et al., who showed that the visibility of graphene is determined by the wavelength and SiO₂ thickness from the difference in reflected light intensities, R, between with and without graphene as

\[
\text{contrast} = \frac{R(\omega - \text{g}) - R(\omega \text{g})}{R(\omega - \text{g})}.
\]

In the present calculation, contrasts for the wavelengths between 450 and 750 nm are superimposed to represent the total contrast for visible light, including the dependence of the light intensity on the wavelength for the halogen lamp and the neutral colour balance filter. The inset shows both \(R(\omega - \text{g})\) and \(R(\omega \text{g})\) superimposed for visible light wavelengths. A small but finite difference between \(R(\omega - \text{g})\) and \(R(\omega \text{g})\) makes the contrast observable.

The electric properties of mono- and multi-layer graphene films were systematically studied. The current modulation increased monotonically with a decrease in the layer number due to the reduction of the interlayer scattering. The carrier mobility in the monolayer was greater than that in the multilayer due to the linear dispersion relation. On the other hand, in the monolayer, the carrier transport was significantly sensitive to the charged impurity density due to the reduction in the screening effect, which caused larger mobility variation. The reduction of the charged impurity density is thus key for high mobility.
preferable. The insulator thickness can be further reduced to 
\( \sim 35 \) nm by replacing SiO\(_2\) with TiO\(_2\), which has a larger refractive index of 2.7.

The inset of Fig. 2(a) shows an optical micrograph of the monolayer graphene observed on a 90 nm SiO\(_2\)/Si substrate. Many mono- and multi-layer graphene films whose contrasts depend on the layer number can be easily found on a SiO\(_2\)/Si wafer. After converting the color to gray scale, the intensity ratio of graphene to SiO\(_2\) was analyzed based on a line profile. In order to determine the reference for the intensity ratio data, only the monolayer was confirmed by a 488 nm Raman spectrum of the 2D band, as shown in Fig. 2(b), since a single peak in the Raman spectrum is a direct evidence for monolayer graphene.\(^6\) The intensity ratio is plotted as a function of the layer number in Fig. 2(a). Even for monolayer graphene, the intensity ratios are scattered and some of them are close to those observed for the bilayer graphene film. However, when the intensity-ratio data for mono- and multi-layer graphene films on the same SiO\(_2\)/Si wafer are selected as shown by red and blue squares with lines (two examples), the intensity ratio is clearly separated and is linearly dependent on the layer number. Therefore, this simple intensity-ratio analysis using the gray-scale images can be used to determine the layer number. Although Ni et al. suggested the optical layer counting without the Raman spectrum,\(^10\) the present experiment has shown that the reference of the monolayer graphene determined by the Raman spectrum is required even for 90 nm SiO\(_2\) with a higher contrast than 300 nm SiO\(_2\) because of the variation of the intensity-ratio data possibly due to the uncertain dead space between graphene and SiO\(_2\). This optical method is applicable for graphene multilayer films thinner than \( \sim 4 \) nm (\( \sim 10 \) layers) because the intensity ratio increases with the thickness on the contrary.

Figure 3 shows (a) sheet resistivity and (b) conductivity as a function of the gate voltage \( V_g \) for mono- and multi-layer graphene films with different layer number. All data were obtained by four-probe measurement, since graphene was not degraded significantly by the deposition of metal electrodes unlike the carbon nanotube (CNT) case.\(^11\) It should be noted that a back gate voltage of 30 V for 90 nm SiO\(_2\) is almost equivalent to 100 V for 300 nm SiO\(_2\). Sheet resistivity monotonically increases with a decrease in layer number, then, for the monolayer, the resistivity curve drastically changes and has the smallest full-width at half-maximum but almost the same resistivity at the Dirac point as that of the bilayer graphene film.

An important finding is that the Dirac point shifts to a negative voltage as the layer number increases. The Dirac point is very sensitive to charged impurities such as resist residue, atmospheric H\(_2\)O and so on.\(^1\) Therefore, the resist residue was carefully removed by H\(_2\)/Ar annealing\(^8^\) and the effect of atmospheric H\(_2\)O was avoided by performing electric measurements in vacuum. The Dirac point in the present experiments moved to the same negative voltages after H\(_2\)/Ar annealing. These results strongly suggest that the Dirac point shifts with the layer number is intrinsic to graphene films. Here, it is worth mentioning that if the work function changes gradually from monolayer graphene to graphite with a increase in the layer number, the charge transfer may take place between the metal electrodes and the graphene film and result in the Fermi level shift due to the relatively low density of states of mono- and multi-layer graphene films. In fact, it has been reported that the work function increased relatively from mono- to bi-layer graphene films in the case of graphene films on SiC.\(^12\) However, qualitative study is required to elucidate the origin of the Dirac point shift.

Figure 3(b) shows the linear relationship between conductivity and gate voltage. The conductivity--\( V_g \) curve for the monolayer graphene is sharper than that of others. This fact is attributable to the much smaller density of states for the monolayer graphene compared to the multilayer graphene films due to the difference in the dispersion relationship. Moreover, only for the monolayer graphene, it was observed that the conductivity curve changed from a linear to a sublinear curve with an increase in the mobility (not shown in the figure), as reported.\(^13\)

Figure 4(a) shows the mobility for mono- and multi-layer graphene films. Since it was found that the carrier density, as determined by the Hall measurement, approximately agreed with the surface charge density induced capacitively.

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**References:**

1. Ni et al.
2. HOPG
3. TiO\(_2\)
4. C\(_24\)
5. SiC
6. monolayer graphene
7. multilayer graphene
8. H\(_2\)/Ar annealing
9. SiO\(_2\)
10. 488 nm Raman spectrum
11. CNT
12. SiC
13. Mobility
by the back gate, the mobility was evaluated by $\mu = 1/\eta n \rho$ where the carrier density $n$ was obtained from $n = \varepsilon_{\text{ox}}/d_{\text{ox}}(V_g - V_{\text{Dirac}})/e$, and $\varepsilon_{\text{ox}}$ and $d_{\text{ox}}$ are the permittivity and thickness of SiO$_2$, respectively. The mobilities of multilayer graphene films with 2, 3, and 4 layers are all roughly 3000 cm$^2$/V·s, while 8200 cm$^2$/V·s at 20 K was calculated to be the highest mobility has been achieved for the monolayer graphene at RT even though there is a wide scattering of the data. When the temperature was reduced to 20K, the mobility increased to $\sim$12,000 cm$^2$/V·s$^{-1}$ for the monolayer graphene, while it did not change for multilayer graphene films. The mean free path for the highest mobility of the monolayer graphene at 20 K was calculated to be $\lambda = v_F \tau$ where $v_F$ is the Fermi velocity of $\sim 10^6$ m/s and $\tau = \hbar \sigma (\pi/\eta n)^{1/2}/(e^2 v_F)$. It was much shorter than the present device size ($\sim 8\mu$m), which suggests that the transport in the monolayer graphene is not ballistic, but is still diffusive.

It has been reported that the conductivity of the monolayer graphene is limited by charged impurity scattering in the case of the linear relationship between conductivity and carrier density. The mobility can be evaluated as a function of the charged impurity density $n_{\text{imp}}$ as shown in Fig. 4(b), where experimental results in Fig. 4(a) are also plotted. A large variation of the charged impurity density in the range of $(5.8-15) \times 10^{11}$ cm$^{-2}$ may result in wide scattering of the mobility obtained experimentally.

With decreasing the layer number, the current modulation is enhanced possibly due to the reduction of the interlayer scattering. On the other hand, when the layer number is decreased from the bi- to mono-layer graphene, the mobility drastically increased due to the inherent change from the quadratic to linear dispersion relationships. The screening length in multilayer graphene films is reported, theoretically, to be 0.5 nm and 0.7 nm, and experimentally to be 1.2 nm. The charges induced by back gate voltage are mainly located within one or two layers considering the inter-graphene layer distance of 0.335 nm. Electric transport of the monolayer graphene should be quite sensitive to charged impurities as the scattering origin due to the screening effect reduction. It is strongly inferred that the large mobility variation for the monolayer graphene is caused by variations in the charged impurity density. If the charged impurity density can be reduced to below $4.8 \times 10^{11}$ cm$^{-2}$, the mobility will exceed 10,000 cm$^2$/V·s$^{-1}$ at RT.

In summary, the number of layers in multilayer graphene films was optically determined by a simple and reliable method using a reference of the monolayer graphene verified by the Raman spectroscopy. Electric characterization for graphene films with well-determined layer numbers revealed that the electric field effectively modulated the carrier transport for the monolayer graphene due to the linear $E-k$ dispersion relation. The mobility became significantly sensitive to charged impurities as a scattering origin due to a decrease in the screening effect, which resulted in large mobility variations. Reduction of the charged impurity density at the graphene/SiO$_2$ interface is key to achieve monolayer graphene with a mobility greater than 10,000 cm$^2$/V·s$^{-1}$ at room temperature.

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