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Effect of hydrogen annealing on electronic transport properties of quasi-free-standing monolayer graphene

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Abstract

We report that annealing temperature during hydrogen intercalation affects the electronic transport properties of quasi-free-standing monolayer graphene. We found that charged-impurity density changes with varying the annealing temperature. From transport measurements, annealing temperatures between 700℃ and 800℃ were found to be optimum for making high-quality quasi-free-standing monolayer graphene with the lowest density of charged impurities.

1. Introduction

Quasi-free-standing monolayer graphene (QFMLG) is one type of graphene that can be formed on SiC over a large area. It is generally obtained by annealing a buffer layer on SiC(0001) in hydrogen atmosphere (hydrogen intercalation) [1]. The buffer layer is an electrically insulating layer consisting of only C atoms, and some of the C atoms are covalently bonded to the Si atoms of the substrate. During hydrogen intercalation, hydrogen atoms break the bonds between the SiC and buffer layer, terminate the SiC by forming Si-H bonds, and convert the decoupled buffer into graphene. The mobility of QFMLG is about 3000 cm^2 V^{-1} s^{-1} at room temperature and in all reported carrier densities [2-4]. Channels for high-frequency devices and integrated circuits are possible applications of QFMLG. If the mobility of QFMLG is improved, its performance in such applications may exceed that of other types of graphene grown on SiC [5, 6].

Higher mobility in QFMLG may be achievable by optimizing its fabrication process. Generally, hydrogen intercalation is carried out at around atmospheric pressure and in a wide range of annealing temperatures (550℃ to 1000℃) [1-4]. Analysis on the effect of the annealing temperature on the transport properties and determination of the optimum annealing temperature for obtaining high-quality QFMLG are the scope of this research.

2. Experimental

Top-gated Hall bars with QFMLG as their channels were used in this study. Methods for fabricating the buffer layer and the devices are similar to those in Ref. 4. We used QFMLGs that were obtained by annealing the buffer layer at 600, 700, 800, and 950℃ in molecular hydrogen atmosphere. After the hydrogen intercalation, we carried out Raman spectroscopy to confirm the formation of QFMLG in all annealing conditions. Electrical measurements in four-terminal method was conducted with a constant current of 1 μA. Measurement temperature was varied from 1.8 to 300 K. Magnetic field B was applied perpendicular to the QFMLG plane.

3. Results and Discussion

Figure 1 shows conductivity σ as a function of carrier density n at low temperature (1.8 or 2 K) for all annealing conditions. n was obtained from measurements of Hall resistance as a function of gate voltage at $B = \pm 0.5$ T or ± 1.0 T. For the annealing temperatures of 600, 700, and 800℃, conductivity has a linear relationship with n in both the hole and electron transport regions. At annealing temperature of 950℃, conductivity is sublinearly dependent on n in the hole transport region. In order to compare the results for the four annealing temperatures, we limit the discussion to the hole transport region.

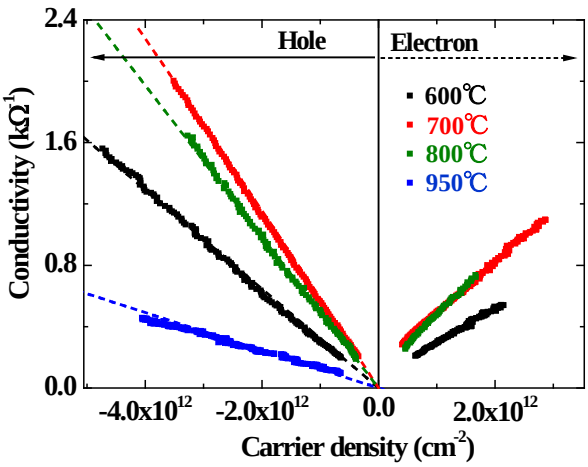

Fig. 1 Carrier-density dependence of conductivity of QFMLG obtained by annealing at 600 (measured at 1.8 K), 700 (measured at 1.8 K), 800 (measured at 1.8 K), and 950℃ (measured at 2 K). The dashed lines show the linear fits to Eq. 1.

The measurement-temperature dependence of mobility for all annealing conditions is next investigated. For the annealing temperature of 700℃, the highest mobility of about 4000 cm^2 V^{-1} s^{-1} was obtained from 1.8 to 300 K at hole density of 3×10^{12} cm^{-2}. Mobility decreases as annealing temperature becomes 800, 600, and 950℃. Our results indicate that an annealing temperature be-

tween 700 and 800℃ is the optimum for obtaining QFMLG with better quality when the buffer layer is annealed by molecular hydrogen at atmospheric pressure.

We next discuss the annealing-temperature dependence seen in Fig. 1. At low measurement temperature, where scattering from phonons can be neglected, charged-impurity scattering (Coulomb scattering) and defect-induced scattering (short-range scattering) are dominant [7]. A distinction between these two types of scattering can be made from the n dependence of σ since σ governed by Coulomb scattering shows a linear dependence with n, while σ becomes sublinear with n when a factor in σ originating from the short-range scattering comes to play a role. The linear relationships between σ and n (Fig. 1) indicate that Coulomb scattering has dominant effect over short-range scattering at low measurement temperature.

We analyze the effect of Coulomb scattering, the major scattering factor, in more detail. σ_c, which is conductivity originating from Coulomb scattering, can be expressed as [8]

$$\sigma_{\mathrm{C}} = C\frac{e^2}{h}\frac{n}{n_{\mathrm{imp}}}, \qquad (1)$$

where C is a constant and n_{imp} is the density of charged impurities. C is about 52 for our devices, and it is calculated by using the dielectric constant of SiC (9.7) and the top-gate insulator (about 6.7) [8]. We estimate n_{imp} by fitting the linearly dependent part of σ–n shown in Fig. 1 at the hole transport region with Eq. (1). Figure 2 shows the relation between n_{imp} and the annealing temperature. n_{imp} is above 6×10^{12} cm^{-2} at the annealing temperature of 600℃, and it decreases to around 3×10^{12} cm^{-2} for 700 and 800℃. At 950℃, n_{imp} increases to above 16×10^{12} cm^{-2}.

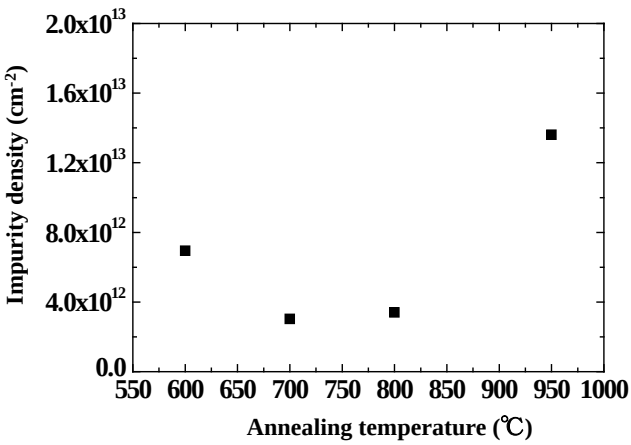

Fig. 2 Density of charged impurities as a function of annealing temperature for hydrogen intercalation.

The high density of charged impurities and the variation of n_{imp} with different annealing temperatures may be originated from the interface states present at the QFMLG/top-gate insulator and QFMLG/substrate. The density of charged impurities originating from the interface states of QFMLG/top-gate insulator should be constant as a function of the annealing temperature and depends on the fabrication process of the insulator. It should be noted that the process for making the insulator is the same for all QFMLG devices.

The structure of QFMLG/substrate changes with the annealing temperature, and thus, the density of interface states can have annealing temperature dependence. For example, Riedl *et al*. reported that annealing QFMLG in vacuum at around 700℃ starts hydrogen desorption at QFMLG/substrate. At above 900℃, most hydrogen atoms are desorbed and the QFMLG returns to the buffer layer [1]. Although we did not perform vacuum annealing after obtaining QFMLG, our transport measurements infer that at the annealing temperature of 950℃, intercalation and desorption of hydrogen atoms come into equilibrium. As a result, a large number of Si atoms at the substrate surface do not form Si-H bonds and become dangling bonds compared to the case in the 700-800℃ range. The dangling bond states then can become origin of charged impurities that affect the carrier scattering. At the annealing temperature of 600℃, it is unlikely for hydrogen desorption to occur. Nevertheless, the density of charged impurities is large. This could be due to incomplete hydrogen intercalation, which would also leave a certain number of dangling bonds. At the annealing temperatures of 700 and 800℃, more hydrogen atoms are intercalated at the QFMLG/substrate compared to 600℃, and they are not desorbed as much as they are at 950℃. Thus, the number of dangling bonds is suppressed, and this temperature range becomes the optimum condition for making QFMLG with fewer interface states.

4. Conclusions

We prepared QFMLG by annealing the buffer layer on SiC(0001) in molecular hydrogen at atmospheric pressure at various annealing temperatures from 600 to 950℃. From electronic transport measurements of top-gated QFMLG devices, we found that annealing temperatures from 700 to 800℃ show lowest number of charged impurities, which contributes to higher QFMLG mobility.

References

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