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Low-temperature hysteresis in the field effect of bilayer graphene

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Abstract. Hysteresis in the field effect of bilayer graphene is observed at a low temperature. We attribute this effect to charge traps in the substrate. When the sweep rate of the back-gate voltage is increased to higher values, the hysteresis becomes more pronounced. By measuring the hysteresis in the field effect, the lifetime of the charge traps is estimated as 16.9 min. It is shown that the influence of charge traps on graphene is strongly affected by a magnetic field. Above 5 T the hysteresis remains constant.

Since the discovery of free-standing atomically thin graphite—the so-called graphene—such monolayers of carbon have been intensively studied [1]–[3]. This system is especially interesting because graphene exhibits a field effect, i.e. by applying a gate voltage it is possible to change the majority charge carriers from holes to electrons [1]. In some samples, hysteresis effects were observed by measuring the field effect of graphene at room temperature [4]. The observed shift in the charge neutrality point depends on the sweep direction of the back-gate voltage and is attributed to dipolar adsorbates, e.g. water that act as charge traps [5]. To suppress such hysteresis under ambient conditions the substrate can be coated with a thin layer of a hydrophobic substance [6]. Using microsoldered graphene, it was shown that polymethylmethacrylate (PMMA) has a doping effect on graphene but does not change the voltage-induced hysteresis [7]. At low temperatures, Yoo *et al* [8] observed a hysteresis effect in graphene nanorings, which they attribute to available trap states in the rough edges of the graphene nanoring structure. Furthermore, hysteresis effects were observed on some systems similar to graphene, e.g. carbon nanotubes. Lee *et al* [9] argue that their observed hysteresis in carbon nanotubes at $T = 56$ K is due to silanol groups (SiOH) at the surface of the substrate.

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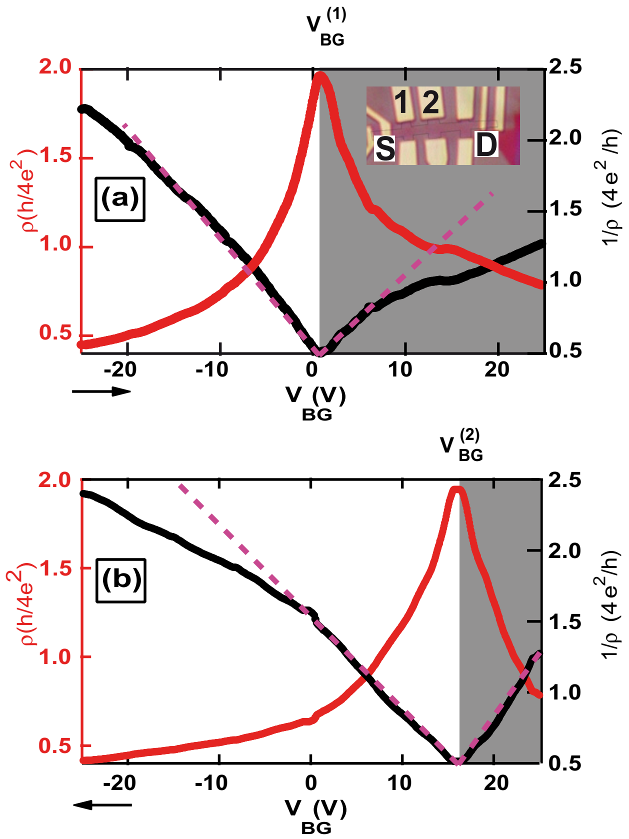

Figure 1. The field effect of graphene when sweeping the back-gate voltage (a) forward from −25 to +25 V and (b) backwards. The left axes show the specific resistance ρ corresponding to the red line; the conductivity σ is demonstrated on the right axes and as the black curve ($B = 0$ T). $V_{BG}^{(1)}$ is the charge neutrality point in the forward direction and $V_{BG}^{(2)}$ for the backward direction. The sweep rate in both cases is $0.15\,V\,s^{-1}$. A clear hysteresis is visible. Inset: optical images of the sample, source (S) and drain (D) contacts are marked; contacts 1 and 2 are used for measurements.

Here, we show hysteresis effects in the field effect of graphene at a low temperature (1.5 K), which we attribute not to the edges of the sample but to charge traps in the substrate. Additionally, the influence of the sweep rate and an applied magnetic field is discussed.

The graphene samples shown here have been prepared by micromechanical cleavage [10] on a variety of silicon substrates with an insulating silicon dioxide layer on top; hence, a back-gate voltage can be applied. Standard e-beam lithography and plasma etching are used to structure the graphene flakes into Hall bars.

The inset of figure 1(a) shows an optical image of the investigated sample. The setup allows us to perform four-point measurements. A direct current $I_{SD} = 500$ nA is driven through the contacts marked source (S) and drain (D). The longitudinal voltage is measured between the contacts labeled 1 and 2. The etched sample is 1 μm wide and the length between the two according contacts is 3.6 μm. During the measurements the device is placed in a helium

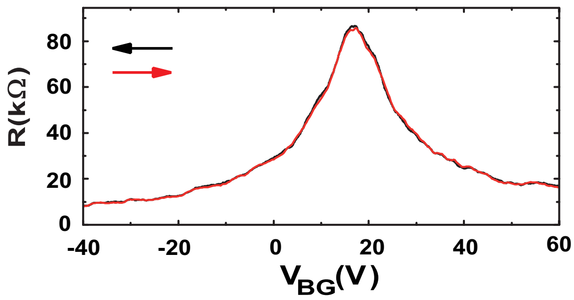

Figure 2. The field effect of a single-crystal bilayer sample on substrate B does not show any hysteresis; that is the black and red curves (back and forth sweep of the back-gate voltage as indicated by the arrows) show no shift.

bathcryostat, so that the temperature is controllable down to 1.5 K. At the same time, a magnetic field B can be applied perpendicular to the sample surface.

In figure 1, the measured field effect at $B = 0$ T and $T = 1.5$ K is shown. In figure 1(a), the back-gate voltage is swept forward from −25 to +25 V, whereas it is swept backward in figure 1(b); in both cases the sweep rate is 0.15 V s^{-1}. The left axes show the resistivity ρ in units of $h/4e^2$, with h being Planck's constant and e the elementary charge. The right axes show the conductivity σ. The dashed lines are a guide to the eye to stress the partially linear behavior of the conductivity. In figure 1(a), the charge neutrality point for the forward sweep $V_{BG}^{(1)}$ is reached at 0.8 V. The conductivity shows a linear behavior in the region from −25 to +9 V. For higher positive back-gate voltages the conductivity shows a kink and stays below the dotted line in figure 1(a). In figure 1(b), a linear behavior is observed in the region from +25 to −4 V. The neutrality point for the backward sweep $V_{BG}^{(2)}$ is found at 16 V. The splitting ($V_{BG}^{(2)}-V_{BG}^{(1)}$) is reproducible as verified for several cycles.

A linear behavior in the conductivity is expected for undoped graphene. It is theoretically understood that molecular adsorbates cause an asymmetry in the field effect of graphene and a nonlinear behavior as observed here in our experiments [11].

To reduce the number of adsorbates, we annealed the sample. Before and after this annealing process the hysteresis was observed in a similar way although the charge neutrality point is shifted by 27 V, i.e. a reduction of the number of adsorbates does not change the hysteresis[2].

To further clarify the origin of the observed hysteresis effect, different samples were produced. Whereas a clear hysteresis was observed in samples produced on substrate A with 265 nm SiO_2 on top, no hysteresis was found in samples on substrate B with 330 nm SiO_2. Figure 2 depicts the field effect of a bilayer sample on substrate B at $T = 1.2$ K. For both the shown sweep directions of the back-gate voltage, the charge neutrality points are found at a finite voltage $V_{BG} = 17$ V, due to residual adsorbates on the sample. There is another piece of evidence that adsorbates do not cause this hysteresis.

These two substrates were provided by different manufacturers. The supplier revealed that charge traps are present in substrate A. The concentration of mobile charges in the oxide

[2] The sample discussed here was annealed for 1.4 h at 150 °C in an atmosphere of 80% N_2 and 20% H_2. The charge neutrality point in the field effect is moved from 40 to 13 V back-gate voltage, indicating residual adsorbates on the sample. Additional samples were annealed within a helium atmosphere; as they were manufactured on the substrate A used here they show the hysteresis effect as well.

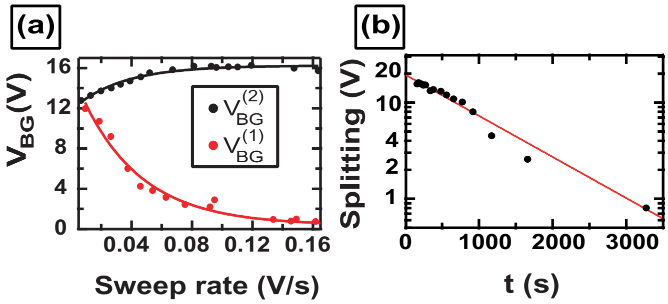

Figure 3. (a) Position of the peak in the field effect (at $B = 0\,\mathrm{T}$) versus the sweep rate of the back-gate voltage ($V_{BG}^{(1)}$ is the back-gate voltage of the charge neutrality point when the gate is swept from -25 to $25\,\mathrm{V}$ and $V_{BG}^{(2)}$ is for the backward direction). The lines are exponential fits to the data points. (b) Logarithm of the splitting ($V_{BG}^{(2)}-V_{BG}^{(1)}$) versus the time that is needed to sweep the back-gate voltage from the first peak $V_{BG}^{(1)}$ to return to the second peak $V_{BG}^{(2)}$.

layer is $9.04 \times 10^{14}\,\mathrm{cm}^{-2}$ and the concentration of interface trapped charges is given as $2.5 \times 10^{13}\,\mathrm{eV}^{-1}\mathrm{cm}^{-2}$ at room temperature. The concentration of traps in substrate B was measured in the same way as in substrate A. However, it was found to be below the detection limit. Hence, in contrast to Yoo *et al* [8], where the authors argue that rough edges of the graphene nanoring structure create available trapped states, we find that the cause of the hysteresis described here is charge traps within the substrate. Furthermore, additional measurements with samples being covered by PMMA showed the same hysteresis, indicating that PMMA does not cause the hysteresis effect [7] and changing the surface properties of the sample does not influence the hysteresis.

To investigate the charging mechanism of the traps with regard to a changing electric field in more detail, the sweep rate of the back-gate voltage is varied from 6.6 to $160\,\mathrm{mV\,s^{-1}}$. These measurements are carried out at $B = 0\,\mathrm{T}$ and $T = 1.5\,\mathrm{K}$. In figure 3(a), the voltage, when the charge neutrality point is reached, is plotted against the sweep rate. At a low sweep rate of $6.6\,\mathrm{mV\,s^{-1}}$ the charge neutrality points for the two sweep directions are almost the same, i.e. the splitting between them is almost 0 V. The charge neutrality point for both sweep directions is found at $V_{BG} = 13\,\mathrm{V}$ (see figure 3(a)). With rising sweep rates the charge neutrality point for the forward direction $V_{BG}^{(1)}$ moves to lower back-gate voltages, as depicted by the lower red curve in figure 3(a), whereas the charge neutrality point for the backward direction $V_{BG}^{(2)}$ moves to higher back-gate voltages for higher rates, as shown by the upper black curve in figure 3(a). In contrast to measurements under ambient conditions, which are explained by adsorbed water molecules acting as charge traps [4], we find the smaller that the splitting between the charge neutrality points in the field effect, the lower the sweep rate of the back-gate voltage.

To analyze the specific time dependence of the hysteresis, in figure 3(b), the splitting between the charge neutrality points for the two sweep directions is plotted against the time it takes to sweep the back-gate voltage from the charge neutrality point in the forward direction $V_{BG}^{(1)}$ to $V_{BG} = 25\,\mathrm{V}$ and back to the charge neutrality point for the backward direction $V_{BG}^{(2)}$ (gray-colored areas in figure 1). It is clearly seen that if a rather short time is needed to return to the charge neutrality point, a large splitting is observed. The more slowly the back-gate voltage returns to the charge neutrality point, the smaller the splitting between the two charge neutrality

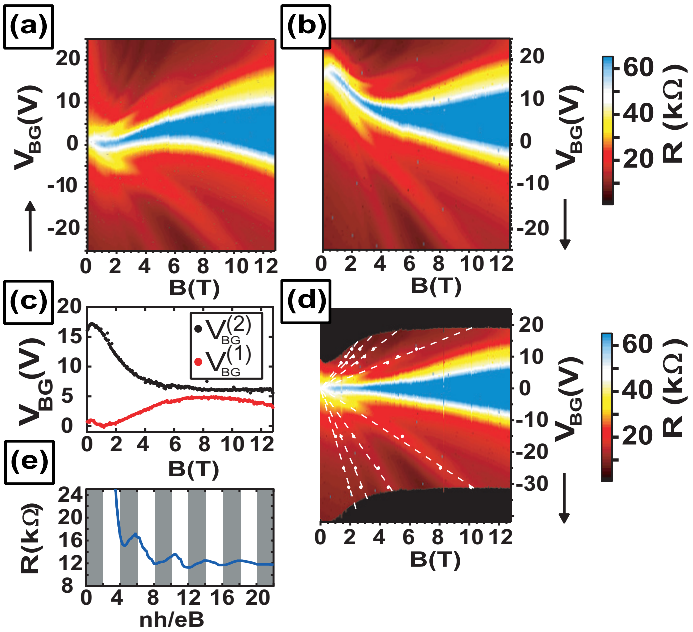

Figure 4. Color plot of the field effect as a function of the magnetic field B; (a) the back-gate voltage is swept from −25 up to +25 V; (b) the back-gate voltage is swept backwards from +25 to −25 V, (c) the position of the charge neutrality point versus the magnetic field in both sweep directions of the back-gate voltage; (d) columnwise shifted color plot of (b); and (e) cross-section through (d) at fixed back-gate voltage ($V_{BG} = 10$ V). The gray sections underline the rising slopes in the Subnikov–de Haas oscillations. The charge carrier concentration is $n = 7.5 \times 10^{15}\,\mathrm{m}^{-2}$.

points. When it takes about 3300 s (55 min), almost no splitting in the charge neutrality points is measured. When $t = 182$ s is needed to sweep the back-gate voltage from the neutrality point $V_{BG}^{(1)}$ to $V_{BG}^{(2)}$ with a rate of $160\,\mathrm{mV\,s^{-1}}$, the splitting is 16 V. As this observed behavior indicates a charge relaxation of the traps, the data points shown in figure 3(b) are fitted by the following expression:

$$(V_{BG}^{(2)} - V_{BG}^{(1)})(t) = A \cdot \exp(-C \cdot t). \quad (1)$$

$A = 19.47$ V and $C = 0.984 \times 10^{-3}\,\mathrm{Hz} = 1/\tau \approx 1/16.9\,\mathrm{min}^{-1}$ are found to be the fitting parameters. The fit in figure 3(b) shows that the characteristic time of the charge traps is $\tau = 16.9$ min for this system at the temperature $T = 1.5$ K. The lifetime of the charge traps at a low temperature was found to be in the same range (of several minutes) for different graphene samples on the same substrate A. By increasing the temperature to 20 K the splitting vanishes: hence, the hysteresis effect described here is not found at room temperature in contrast to the hysteresis discussed in [4, 5, 7].

So far, all the shown experiments were performed without a magnetic field applied ($B = 0$ T). To study the influence of a magnetic field on the hysteresis a magnetic field is applied perpendicular to the sample surface. The magnetic field is varied in steps of $\Delta B = 50$ mT. At each field the back-gate voltage is swept and the field effect is measured. The result is shown in figures 4(a) and (b) for each sweep direction of the back-gate voltage. In figure 4(a), the resistance of the sample is shown as a function of the magnetic field when sweeping the back-gate voltage forward with a sweep rate of $0.15\,\mathrm{V\,s^{-1}}$. It is clearly visible that additional maxima appear with rising magnetic field B. These maxima corresponding to Landau levels are expected to show a linear behavior with rising magnetic field. As can be seen in figure 4(a), those maxima show a nonlinear dependence on the magnetic field. By comparing figures 4(a) and (b), differences were obvious. The charge neutrality point for $B = 0\,\mathrm{T}$ is shifted to higher back-gate voltage with respect to the other sweep direction of the back-gate voltage (as already noted in figure 1). As in figure 4(a) the maxima in figure 4(b) are bent, although in a different direction. To investigate this behavior in more detail, the position of the charge neutrality point versus the magnetic field is depicted in figure 4(c) for each sweep direction. It is clearly visible that by increasing the magnetic field the splitting between the charge neutrality points shrinks. Whereas the splitting stays more or less constant up to 0.3 T, it is drastically reduced between 0.3 and 5 T and remains more or less constant with a splitting of about 2 V for higher fields.

The two curves in figure 4(c) are used to shift and recalibrate each back-gate voltage sweep in figures 4(a) and (b) in such a way that each field effect (at different magnetic fields) shows its maximum at $V_{\mathrm{BG}} = 0\,\mathrm{V}$. The result is shown in figure 4(d) for the backwards sweep direction. The expected Landau fan with the linear behavior of the peaks can be seen. In figure 4(e), the typical Shubnikov–de Haas oscillations at fixed back-gate voltage ($V_{\mathrm{BG}} = 10\,\mathrm{V}$) are shown. The resistance is plotted against nh/eB, with a carrier concentration $n = 7.5 \times 10^{15}\,\mathrm{m^{-2}}$. We find minima in the oscillations at $\nu_{\mathrm{min}} = 4i$, with $\nu = nh/eB$ being the filling factor and i an integer. This and the optical contrast indicate that the sample used here is a single-crystal bilayer [12]. For graphene the charge carrier concentration is manipulated by the back-gate voltage. Thus, the magnetic field dependence of the hysteresis does not depend on the charge carrier concentration as is observed for the whole back-gate voltage range. Therefore, the magnetic field dependence observed here shows the interaction of the charge traps and the external magnetic field. With rising magnetic field the influence of the charges traps on the field effect of graphene decreases. Clearly, at about $B = 5\,\mathrm{T}$ a transition to a much weaker influence occurs. Hence, the ability to trap and detrap charges is dramatically changed by the magnetic field. An adequate model is still needed for describing this magnetic field dependence of the charge traps.

We show here that graphene is used to detect and to distinguish charging effects in its vicinity. A common difficulty in measuring the dielectric properties of molecular solids at low temperature is that frequently they contract faster than the electrodes, and if the electrodes cannot follow the contraction, the sample cracks [13]. Graphene helps us to avoid this difficulty, and by just measuring the field effect, the characteristics of charging effects close to graphene can be measured and the dielectric properties of the material can be accessed.

In conclusion, we have used graphene to probe, at low temperature, charge traps in the underlying substrate. It was demonstrated that the splitting in the field effect strongly depends on the sweep rate. A bilayer graphene was used here to determine the lifetime of the charge traps to be 16.9 min. The measurements carried out here have shown clearly that the hysteresis effect in the graphene devices depends strongly on the magnetic field. By rising the magnetic field the hysteresis was strongly suppressed. Above 5 T the hysteresis remained constant.

Acknowledgments

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