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Anomalous anisotropic magnetoresistance effects in graphene

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We investigate the effect of external stimulus (temperature, magnetic field, and gases adsorptions) on anisotropic magnetoresistance (AMR) in multilayer graphene. The graphene sample shows superlinear magnetoresistance when magnetic field is perpendicular to the plane of graphene. A non-saturated AMR with a value of −33% is found at 10 K under a magnetic field of 7 T. It is surprisingly to observe that a two-fold symmetric AMR at high temperature is changed into a one-fold one at low temperature for a sample with an irregular shape. The anomalous AMR behaviors may be understood by considering the anisotropic scattering of carriers from two asymmetric edges and the boundaries of V_+(V_-) electrodes which serve as active adsorption sites for gas molecules at low temperature. Our results indicate that AMR in graphene can be optimized by tuning the adsorptions, sample shape and electrode distribution in the future application. [http://dx.doi.org/10.1063/1.4894519]

I. INTRODUCTION

Graphene has been considered as a potential functional material due to its linear dispersion for charge carriers,[1] high carrier mobility,[2,3] quantum Hall effect,[4,5] etc. Recently, magnetoresistance (MR) effect in graphene has attracted considerable interests due to its potential applications in magnetic sensors.[6–8] A large linear non-saturated MR up to 100% was found in graphene,[9] which is quite different from the traditional MR with quadratic magnetic field dependence and a relatively small magnitude (smaller than 1%).[10] This linear MR was attributed to the inhomogeneities or disorder of graphene based on both quantum and classical models.[11–15] Moreover, the linear MR was also found to be anisotropic when the magnetic field is applied parallel and perpendicular to the basal plane of graphene.[16] This anisotropic magnetoresistance (AMR) effect results from the resistivity dependence on the magnetic field orientation with respect to the graphene basal plane. As the charge carriers are confined in the 2D graphene,[17] it is hard to change the trajectories of the carries by a Lorentz force when applying a magnetic field parallel to the graphene basal plane. On the contrary, the trajectories of the carries can be changed by a magnetic field applied perpendicular to the graphene basal plane. Recently, Liao *et al* reported an AMR effect in graphene and found that the MR is dependent on the angle between the magnetic field orientation and the basal plane of graphene.[18] AMR effect found in graphene indicates that graphene is one of the potential AMR materials for developing ultra-thin AMR devices.

However, the influences of external stimulus (i.e., temperature, gases adsorptions) on AMR in graphene are less studied and still unclear.[16,18] The lack of such studies significantly limits the

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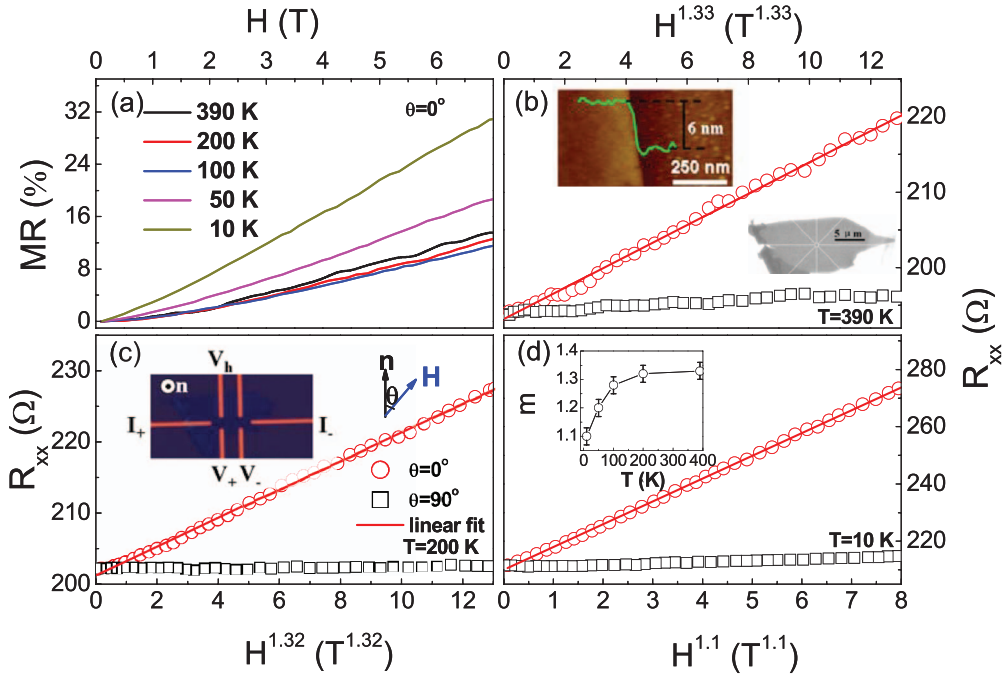

FIG. 1. (a) Magnetic field dependence of MR measured for $\theta = 0^\circ$ at 390, 200, 100, 50 and 10 K, respectively. (b) R_{xx} as a function of $H^{1.33}$ at 390 K. (c) R_{xx} as a function of $H^{1.32}$ at 200 K. (d) R_{xx} as a function of $H^{1.1}$ at 10 K. The up and bottom inset of (b) show the topography signals (green line) across the step between graphene and the substrate, and SEM image of the graphene sample, respectively. The inset of (c) shows schematics for the sample configuration, and inset of (d) shows the temperature dependence of the fitting parameter m.

application of graphene in magnetic sensors. In this work, we have systematically studied the AMR effects in graphene at various temperatures and magnetic fields. A MR of 30% and an AMR of −33% are observed in irregular-shaped graphene. Interestingly, a two-fold symmetric AMR at high temperature is observed to change into a one-fold one at low temperature for a graphene sample with an irregular shape. The anomalous AMR behaviors can be understood based on an anisotropic scattering of carriers from two asymmetric edges and the boundaries of $V_+(V_-)$ electrodes, which serve as active adsorption sites for gas molecules at low temperature.

II. EXPERIMENTAL

The graphene sample was transferred onto thermally grown SiO_2 (300 nm-thick) on heavily doped Si substrates by a mechanical exfoliation from Kish graphite by scotch tape. The sample is of a hexagon shape with asymmetric edges as shown in the bottom inset of Fig. 1(b). Electrode contacts of Ti (10 nm)/Au (50 nm) were fabricated by electron beam lithography, followed by annealing in Ar atmosphere at 400°C for an hour to improve the contact. The Atomic Force Microscope (AFM) was used to observe the morphology of graphene. As seen from the up inset of Fig. 1(b), the thickness of graphene is about 6 nm. This means the sample is a multilayer graphene. The AMR and Hall resistance were measured by a Physical Property Measurement System (PPMS, Quantum Design) with a Hall bar geometry as shown in the inset of Fig. 1(c). Out-of-plane rotation of the magnetic field was used to measure AMR. During the measurements, the angle between magnetic field and the direction (n), i.e., the normal direction of the sample's plane is defined as θ, as shown in the inset of Fig. 1(c), and the magnetic field is always kept perpendicular to the current direction. The MR of graphene is defined as $MR(H) = [R(H)/R(0)\text{-}1]\times 100\%$, where $R(H)$ and $R(0)$ represents the resistance with and without magnetic field, respectively. The AMR of graphene is defined as $AMR(\theta) = [R(\theta)/R(0)\text{-}1]\times 100\%$, where $R(\theta)$ and $R(0)$ represents the resistance at θ and $\theta = 0^\circ$, respectively.

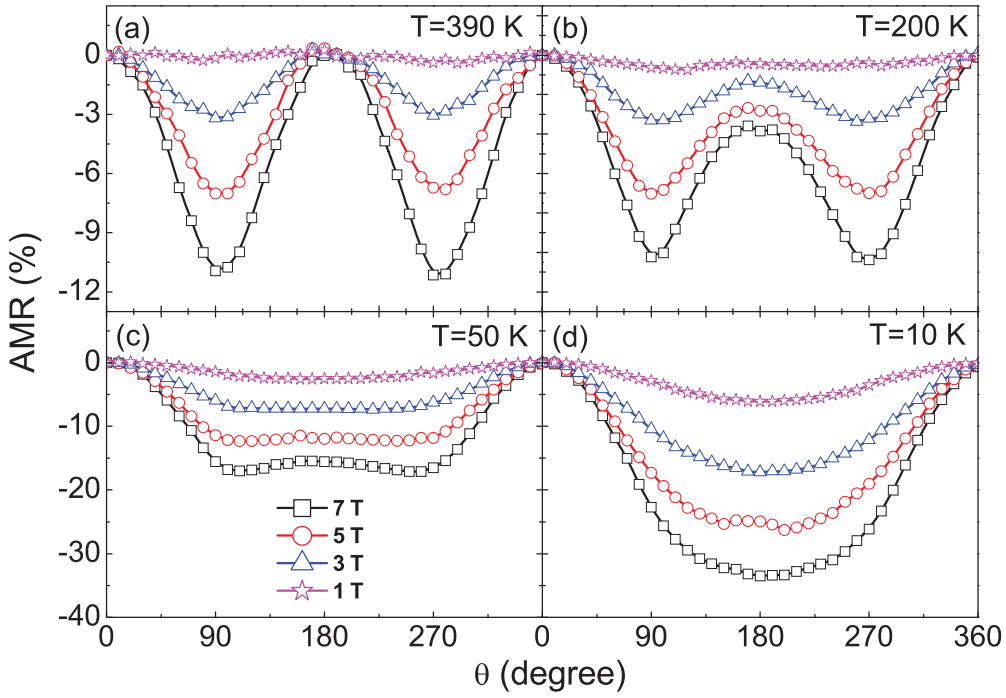

FIG. 2. AMR as a function of θ at (a) 390 K, (b) 200 K, (c) 50 K, and (d) 10 K under various magnetic fields.

III. RESULTS AND DISCUSSIONS

The magnetic field ($\theta = 0^\circ$, perpendicular to the plane) dependence of MR in graphene at various temperatures is shown in Fig. 1(a). The MR shows a non-linear dependence of temperature at a fixed magnetic field of 7 T. First, the MR value decreases from 13.6% at 390 K to 11.4% at 100 K. Then, MR increases with further decrease of temperature from 100 to 10 K, and reaches to about 30% at 10 K. As shown in Figs. 1(b)–1(d), the longitudinal resistance R_{xx} at $\theta = 0^\circ$ shows linear dependence on H^m (superlinear MR), where m is a fitting parameter larger than 1. It is noted that the superlinear MR is dependent on the temperature. As shown in the inset of Fig. 1(d), m increases with increasing temperature as shown in the inset of Fig. 1(d).

It is known that the increase of the disorder, where current paths no longer align with the applied voltage, can change the quadratic MR gradually towards linear MR. When the disorder is sufficiently strong, the linear MR can appear.[12] The decrease of m at low temperature suggests an increased disorder or inhomogeneity at low temperature. Compared with the superlinear MR at $\theta = 0^\circ$, R_{xx} at $\theta = 90^\circ$ remains almost constant with the magnetic field for different temperatures as shown in Figs. 1(b)–1(d), respectively. It is well known that a magnetic field at $\theta = 0^\circ$ could change the trajectories of carriers into a set of circles parallel to the basal plane of graphene due to Lorentz force, and a corresponding quadratic positive MR effect could be induced according to the classical theory,[10] which is proportional to $(eB\tau/m)^2$, where τ is the relaxation time, m is the effective mass of the electrons, e is the charge of electron, and B is the magnetic field.[13] There is a long relaxation time and near zero effective mass in graphene[2], therefore, large magnetoresistance can be expected. However, due to the inhomogeneous spatial distribution of the carrier mobility, linear and superlinear magnetic field dependence of MR are expected in graphene,[11–13] just as shown in Fig. 1. On the other hand, as the charge carriers are confined in the 2D graphene, it is hard to change the trajectories of the carries by a Lorentz force perpendicular to the graphene basal plane ($\theta = 90^\circ$). Therefore, the scattering of the carriers in graphene is difficult to be tuned and R_{xx} at $\theta = 90^\circ$ remains almost constant as shown in Fig. 1.

Figure 2 shows the angular dependence of AMR at different temperatures with various H. The maximum value of AMR increases along with H, and it shows a two-fold symmetry with varying

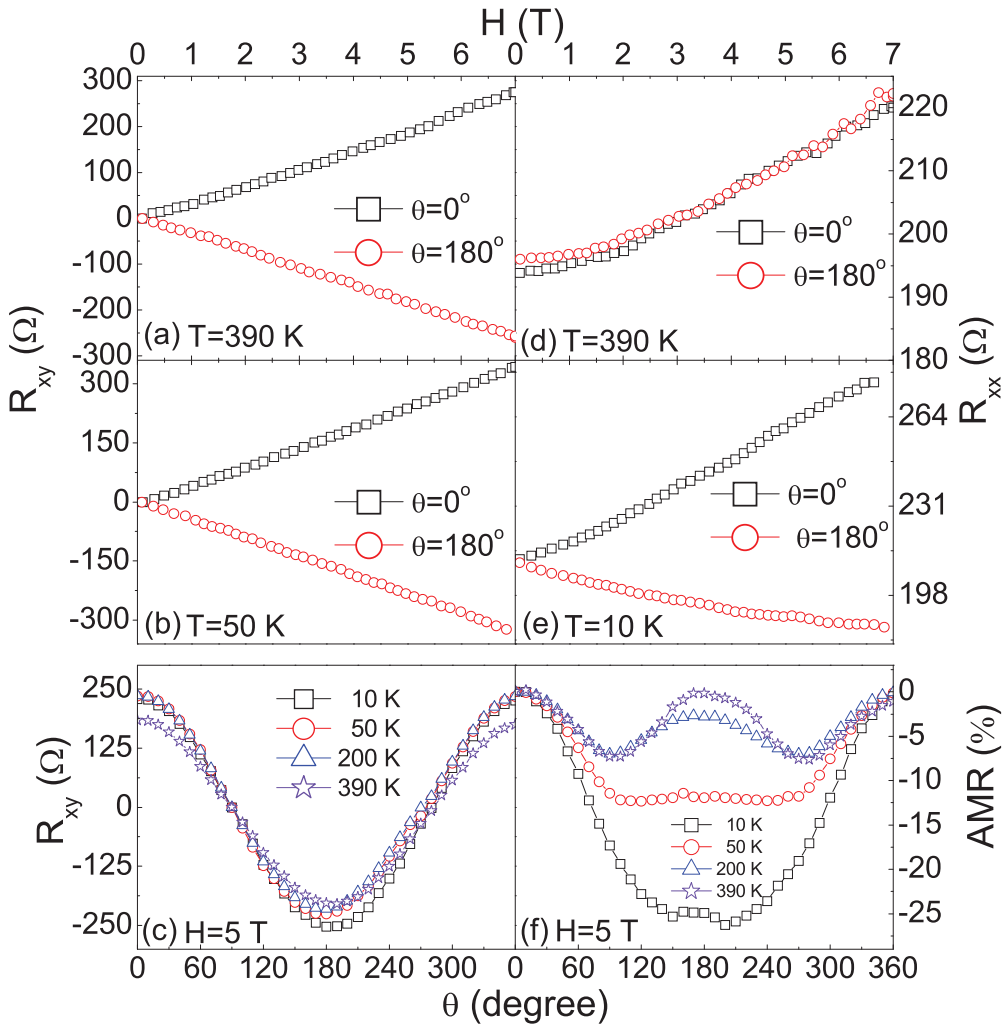

FIG. 3. R_{xy} as a function of the magnetic field at (a) 390 K and (b) 10 K. R_{xx} as a function of the magnetic field at (d) 390 K and (e) 10 K. The angular dependence of (c) R_{xy} and (f) AMR.

θ at 390 K. The AMR at $\theta = 180^\circ$ is nearly zero at 390 K and does not change with increasing H. Interestingly, different AMR values are observed at $\theta = 0^\circ$ and 180° for temperatures lower than 390 K. The absolute value of AMR at $\theta = 180^\circ$ increases with decreasing the temperature, and reaches to a maximum of 33% under $H = 7$ T at 10 K. The key point is that AMR as a function of θ shows a one-fold symmetry at 10 K.

As is known, Hall resistance R_{xy} as a function of θ shows a one-fold symmetry.[19] In order to check whether R_{xy} has contribution to R_{xx}, the angular dependence of R_{xy} was measured from 390 to 10 K at various magnetic fields. According to the Hall measurement, the carriers are n-type electrons, and the carrier density is increased from 2.2×10^{19} to 2.8×10^{19} /cm^3 with increasing the temperature from 10 to 390 K. Fig. 3 shows the comparison of R_{xy} and R_{xx} as a function of H and θ. As seen from Figs. 3(a) and 3(b), R_{xy} increases linearly with increasing H for $\theta = 0^\circ$, and when H is reversed (i.e. $\theta = 180^\circ$), R_{xy} changes sign. R_{xy} shows very little difference from 390 to 10 K as shown in Fig. 3(c). As shown in Figs. 3(d) and 3(e), the dependence of R_{xx} on H for $\theta = 0^\circ$ and 180° are nearly the same, and show opposite trends with increasing H at 390 and 10 K, respectively. The angular dependence of AMR from 390 to 10 K at 5 T is shown in Fig. 3(f), which changes from two-fold symmetry to a one-fold one. With decreasing the temperature, R_{xy} shows nearly no change, therefore, the symmetry change of AMR with decreasing the temperature can not be induced by the Hall effect.

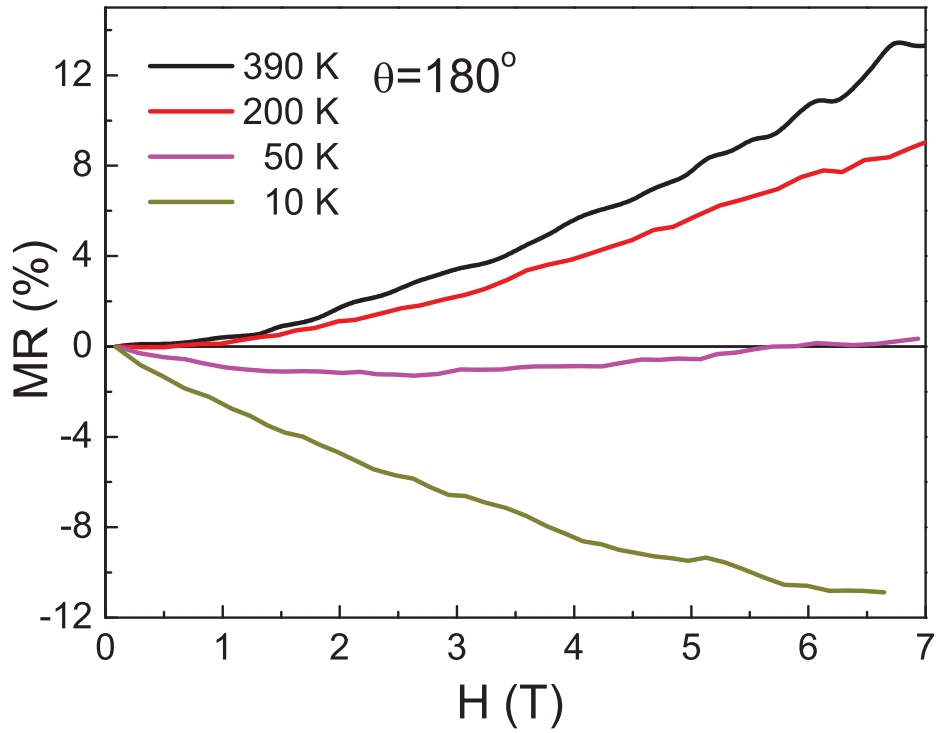

FIG. 4. The magnetic field dependence of MR at $\theta = 180^\circ$ at various temperatures.

The anomalous AMR behavior is possibly related to the irregular shape, the boundaries of the V_+ and V_- electrodes (the inset of Fig. 1) and gas adsorption of graphene. It is noted that the mean-free paths are about micrometer for graphene.[20] Although it is impossible to obtain such large mean-free paths in our multilayer graphene, the sample can also have a large mean free path than conventional metal, such as copper.[21] As seen in the inset of Fig. 1(c), the distribution of the electrodes must result the distribution of the current. Therefore, the carriers near the edges, V_+ and V_- electrodes can influence the current transport. For $\theta = 0^\circ$ and 180°, opposite Lorentz force is applied on the carriers separating the carriers towards the two asymmetric edges resulting in the change of the current distribution. It is noted that V_+ and V_- electrodes are in one side of the edges, which increase the asymmetry of the sample. Therefore, the two asymmetric edges, V_+ and V_- electrodes influence the electronic transport, which may cause anisotropic scattering of carriers. In addition, it is known that there are a lot of dangling bonds at the edges of graphene, which serve as active adsorption sites for gas molecules.[22,23] Moreover, the boundaries of V_+ and V_- electrodes can also be treated as "the edges". During the measurements, the graphene samples were loaded into a Helium atmosphere in the PPMS chamber, thus the sample's edges, graphene near the boundaries of V_+ and V_- electrodes can adsorb gas molecules during cooling. The molecule adsorption may cause more disorders or inhomogeneous spatial distribution of the carriers. As shown in the inset of Fig. 1(d), the superlinear fitting parameter m decreases from 1.33 at 390 K to 1.1 at 10 K, suggesting an increased inhomogeneity for $\theta = 0^\circ$ at low temperature.[24] On the other hand, the increase of disorders or inhomogeneities can result in a negative MR as reported by Zhou *et al.*[9] As seen in Fig. 4, for $\theta = 180^\circ$, a positive MR effect is observed above 50 K, but MR changes to negative at 10 K. This quite different from the MR effect for $\theta = 0^\circ$ as seen in Fig. 1(a). The different temperature dependence of MR between $\theta = 0^\circ$ and 180° thus reflects the anisotropic scattering of the carriers. At low temperatures, the two asymmetric edges, the graphene near the boundaries of V_+ and V_- electrodes may adsorb more molecules thus changes the band structure and electronics transport properties, which may enhance the asymmetry of the two edges. As a result, the anisotropic scattering of the carriers increased correspondingly. Therefore, the absolute value of AMR at $\theta = 180^\circ$ increased with decreasing the temperature. The adsorbing of the molecules at the

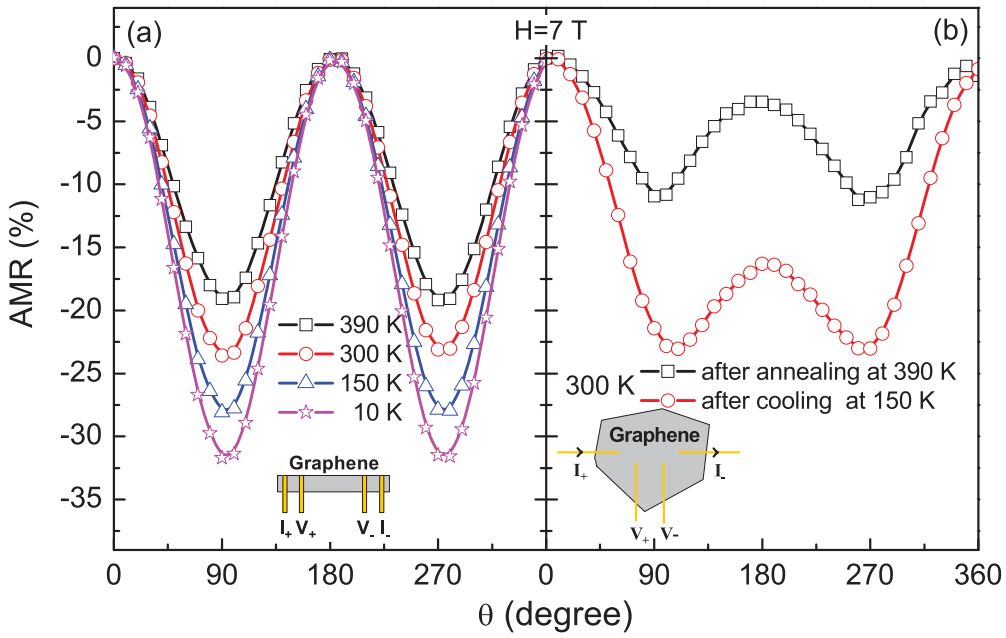

FIG. 5. (a) AMR as a function of θ at various temperatures for a graphene with a rectangular shape. (b) AMR as a function of θ at 300 K under $H = 7$ T after annealing at 390 K and cooling at 150 K for 24 hours, respectively. The insets of (a) and (b) show the schematics of the measured graphene samples.

asymmetric edges and the boundaries of V_+ and V_- electrodes at the low temperature is important for the observed anomalous AMR.

In order to clarify the sample shape and electrode distribution influences on the observed anomalous AMR effects, we prepared a rectangular graphene sample with four symmetric electrodes, as schematically shown in the inset of Fig. 5(a), and measured the AMR under $H = 7$ T at various temperatures. With decreasing the temperature, AMR increases generally, but it does not show any differences between $\theta = 0^\circ$ and 180° even at 10 K. Therefore, the anomalous AMR behaviors are confirmed to be related to the anisotropic scattering of carriers from the two asymmetric edges and the boundaries of V_+ and V_- electrodes.

In order to justify the influence of the possible molecule adsorption, AMR effects of the graphene sample with an irregular shape were measured at 300 K after two different processes, respectively. First, the sample was warmed up to 390 K in order to remove some adsorptions then cooled down to 300 K. Second, the sample was cooled down to 150 K and held for 24 h in order to adsorb more molecules, and then warmed to 300 K. The AMR as a function of θ for the above two processes is shown in Fig. 5(b). It is clearly seen that the absolute value of AMR for the second process is much larger compared to that for the first process, which well supports our hypothesis. As the mean-free path is still much smaller than the sample size (about 10 μm), we believe that only the carriers near the asymmetric edges and the boundaries of the electrodes contribute to the above anomalous AMR effect. If the mean-free path is comparable to the sample size, the anomalous AMR can reach a larger value.

In summary, AMR effects of graphene were systematically studied at various magnetic fields and temperatures. When the magnetic field is perpendicular to the graphene plane, a 30% non-saturated superlinear MR was observed under a magnetic field of 7 T at 10 K. It is found that AMR as a function of θ shows a two-fold symmetry at high temperature, and changes to a one-fold symmetry at low temperature in a graphene sample. Correspondingly, the absolute value of AMR at $\theta = 180^\circ$ increased from 0% at 390 K to 33% at 10 K. The anomalous AMR behaviors could be attributed to the anisotropic scattering of carriers from the asymmetric edges of the irregular shape graphene sample and the boundaries of V_+ and V_- electrodes, which serve as active adsorption sites for gas molecules at low temperature.

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