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Hopping magnetoresistance in ion irradiated monolayer graphene

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HIGHLIGHTS

- Magnetoresistance of ion irradiated graphene samples with VRH mechanism of conductivity was measured.
- In perpendicular magnetic fields negative magnetoresistance was observed.
- In parallel magnetic fields positive magnetoresistance was observed.
- Theoretical interpretation of the observed results was proposed.

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ABSTRACT

Magnetoresistance (MR) of ion irradiated monolayer graphene samples with a variable-range hopping (VRH) mechanism of conductivity was measured at temperatures down to $T=1.8$ K in magnetic fields up to $B=8$ T. It was observed that in perpendicular magnetic fields, hopping resistivity R decreases, which corresponds to negative MR (NMR), while parallel magnetic field results in positive MR (PMR) at low temperatures. NMR is explained on the basis of the "orbital" model in which perpendicular magnetic field suppresses the destructive interference of many paths through the intermediate sites in the total probability of the long-distance tunneling in the VRH regime. At low fields, a quadratic dependence ($|\Delta R/R| \sim B^2$) of NMR is observed, while at $B > B_*$, the quadratic dependence is replaced by the linear one. It was found that all NMR curves for different samples and different temperatures could be merged into common dependence when plotted as a function of B/B_*. It is shown that $B_* \sim T^{1/2}$ in agreement with predictions of the "orbital" model. The obtained values of B^* also allowed us to estimate the localization radius ξ of charge carriers for samples with a different degree of disorder. PMR in parallel magnetic fields is explained by suppression of hopping transitions via double occupied states due to alignment of electron spins.

1. Introduction

Defects are very useful tool to modify and control the physical properties of true two-dimensional (2d) material – monolayer graphene, therefore, disordered graphene attracts a lot of attention [1–7]. The evolution of optical (Raman spectra) and electronic (conductivity) properties of graphene with increasing disorder has been investigated in many papers (see, for example, [8–11]). It has been shown in [11] that an increase of disorder leads to gradual change of the mechanism of conductivity from metallic one in pristine films to the regime of weak localization, and then to the mechanism of variable-range hopping (VRH) of strongly localized charge carriers. In this work, we report the results of magnetoresistance (MR) measurements in samples of monolayer graphene strongly disordered with ion irradiation. MR in the VRH regime of conductivity has been observed earlier in different graphene-based structures: fluorinated graphene [4], graphene exposed to ozone [5], graphene oxide [6]. In Ref. [7], MR was measured in monolayer graphene flakes subjected to Ga^+ ion irradiation. In one highly disordered sample, the negative MR was observed which was attributed to the crystalline-boundary scattering. In Ref. [9], disorder in graphene was introduced by C^+ ion irradiation with energy 35 keV. It was shown that at high dose of irradiation, the conductivity is described by the VRH mechanism, but no MR

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measurements were conducted. We are not aware about study of hopping MR in series of monolayer graphene samples with gradually increased degree of disorder.

2. Experimental results and discussion

The investigated samples belong to a series of samples disordered by a different dose of ion irradiation. All samples were fabricated by means of electron-beam lithography (EBL) on the common large-scale (5 × 5 mm) monolayer graphene film and divided into 6 groups. Initial sample before EBL was marked as sample 0. The samples from the first group, marked as sample 1, were not irradiated, while 5 other groups were subjected to different doses (from 5×10^{13} up to 1×10^{15} cm^{-2}) of irradiation with carbon ions with energy 35 keV. In Ref. [10], concentration of structural defects N_D was determined for each group of samples using measurements of the Raman scattering. For samples 1, 2, 3 and 4, the values of N_D in units of 10^{12} cm^{-2} were 1, 3, 6 and 12 respectively. In Ref. [11], it was shown that the temperature dependence of resistivity $R(T)$ in initial sample 0 has a metallic character, while in slightly irradiated sample 1, conductivity is characterized by the regime of “weak localization”. Measurements of MR in this sample showed a remarkable agreement with a theoretical model [12]. In more irradiated samples 2–4, dependence $R(T)$ is described by the variable-range hopping (VRH) mechanism of conductivity typical for strongly localized carriers [13]. As known, the form of $R(T)$ in VRH depends on the structure of the density-of-localized states $g(\mu)$ in the vicinity of the Fermi level (FL) μ: when $g(\mu)\neq 0$, $R(T)$ is described by the “Mott VRH” which in 2d has the form of “$T^{-1/3}$–law”:

$$R(T) = R_0 \exp(T_M/T)^{1/3}, \quad T_M = C_M[g(\mu)\xi^2]^{-1}. \tag{1}$$

Here R_0 is a prefactor, $C_M = 13.8$ is the numerical coefficient, ξ is the radius of localization. The Coulomb interaction between localized carriers leads to their redistribution in the vicinity of the FL. This results in a “soft” Coulomb gap around FL, which in 2d has a linear form $g(\epsilon) = |\epsilon - \mu|(\kappa/e^2)$, where κ is the dielectric constant of the material. In this case, $g(\mu) = 0$ and VRH is described by the “Efros–Shklovskii (ES) VRH” or “$T^{-1/2}$–law”:

$$R(T) = R_0 \exp(T_{ES}/T)^{1/2}, \quad T_{ES} = C_{ES}(e^2/\kappa\xi) \tag{2}$$

Here $C_{ES} = 2.8$ is the numerical coefficient. Measurements of MR in samples 2–4 were performed at temperatures down to 1.8 K in magnetic fields up to $B = 8$ T in perpendicular and in-plane (parallel) $B_{\parallel}$ geometry. It was observed that $B_{\perp}$ leads to negative magnetoresistance (NMR) while $B_{\parallel}$ results in positive magnetoresistance (PMR) at low temperatures, Fig. 1. This anisotropy shows unambiguously that MR in perpendicular and parallel fields has different origin: NMR is determined by the orbital mechanisms, while PMR is determined by the spin polarization. In that order we will discuss the results of measurements.

3. NMR in perpendicular magnetic fields

Fig. 2 shows the MR curves $\Delta R(B)/R(0) \equiv [R(B) - R(0)]/R(0)$ at different T for all three samples on a linear scale. One can see, that NMR at fixed T decreases with an increase of disorder from sample 2 to 4. For samples 2 and 3, NMR increases with decreasing T. For sample 4, $\Delta R/R$ first increases with decreasing T, but at $T < 10$ K, NMR rapidly decreases and the curves seek to change the sign. It could be due to the standard positive MR caused by the shrinkage of the wave functions in perpendicular magnetic fields [13]. Therefore we will discuss the NMR for sample 4 only down to 10 K.

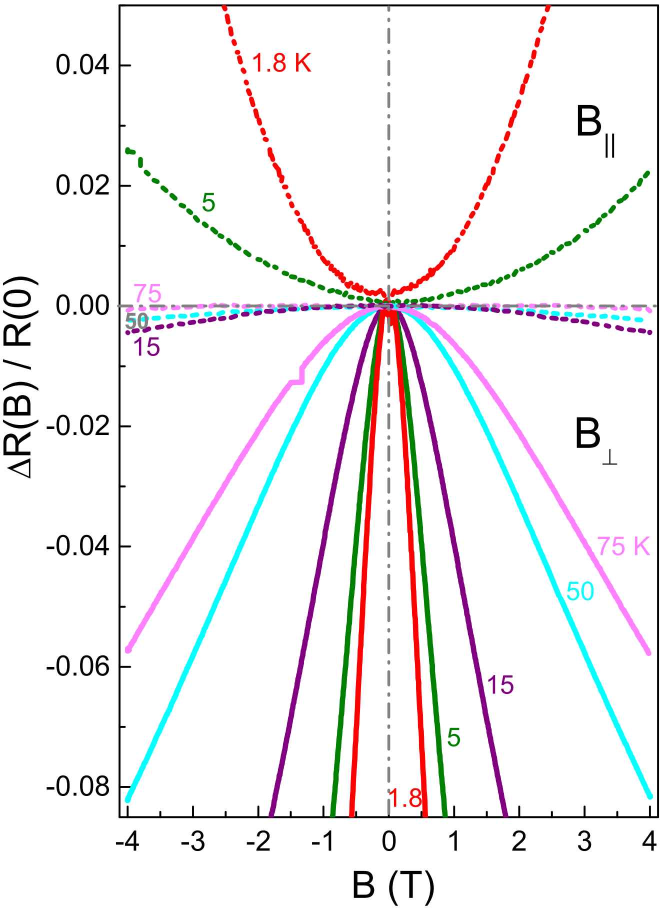

Fig. 1. $\Delta R/R$ as a function of B for sample 3 in parallel and perpendicular magnetic fields at different temperatures in K, shown near each curve.

In Fig. 3, NMR curves are plotted on the log–log scale. On this scale, the slope to the curve is equal to the power m in $\Delta R/R \sim B^m$. Quadratic dependence ($m = 2$) is observed at low fields up to some value B^*. At $B > B^*$, the quadratic dependence is replaced by linear one ($m = 1$) and then by sublinear dependencies. Some values of B^* are shown in Fig. 3 by arrows. Very weak effect of NMR (about 1–2%) in VRH regime was earlier observed in three-dimensional (3d) conductivity in heavily doped and compensated Ge (for a review, see [14]). In 2d, a significant NMR in the VRH regime has been observed in perpendicular magnetic fields in different systems [15–20]. Anisotropy of this effect in perpendicular and parallel fields unambiguously indicates the orbital nature of NMR, because effects due to spin polarization have to be isotropic. Theoretically the effect of orbital NMR in the VRH regime of conductivity has been discussed in [21–27]. The main idea of the model suggested by Nguen, Spivak and Shklovskii [21] is based on the following consideration.

In VRH, only part of localized states with energy levels within the so-called “optimal band” around FL $\epsilon(T)$ is involved in the hopping process. In “Mott VRH”, $\epsilon(T)$ decreases with a decrease of temperature [13]:

$$\epsilon(T) = T^{2/3}[g(\mu)\xi^2]^{-1/3} \tag{3}$$

Correspondingly the hopping distance r_h increases, which gives $r_h \sim T^{-1/3}$:

$$r_h \approx [g(\mu)\epsilon(T)]^{-1/2} \approx \xi(T_M/T)^{1/3}. \tag{4}$$

In “ES VRH”, $r_h \sim T^{-1/2}$. Therefore, at low T, r_h becomes much larger than the mean distance between localized centers, and the