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Study of the indirect exchange interaction in a strained graphene nanoribbon

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ABSTRACT

In this paper we study the indirect exchange interaction in a strained graphene ribbon of finite width within a frame of the s–d model. Our calculations show that magnetic ordering of the spins of impurities varies periodically with increasing distance between atoms and asymptotically tends to zero. We analyze the dependence of the exchange interaction on the directions of deformation in the film, as well as on the external magnetic field.

1. Introduction

Numerous studies show the growing interest of researchers for the effects arising in graphene subjected to a magnetic field. Perspectives of different applications of spintronics are confirmed, both theoretically and experimentally [1–3]. Many studies show that graphene is also a material with unique adsorptive properties [4–9].

Recently, scientists have investigated graphene sheets deformation, spurring a significantly renewed interest in this field. One of the reasons for this is the remarkable ability to create giant pseudo-magnetic fields by applying mechanical stress to a sheet of graphene [10].

All of the above brought us to the idea of considering the complex collective effects associated with the interaction of electrons with impurities of a deformed crystal lattice of graphene [11–16]. The objective of the present study is to gain insight into the properties of these interactions responsible for the intriguing emergent behavior of giant pseudo-magnetic fields generation. Note that the magnetic fields considered in the framework of our study can easily be mapped to the pseudo-magnetic ones, given the mathematical equivalence between both quantities.

2. Dispersion relation for strained graphene

We examine a layer of graphene, consisting of two sublattices a and b in the Hückel approximation [18], solely considering the dynamics of π−electrons. More specifically, in the framework of our model we take into account the kinetic energy of the electrons combined with the energy of the impurity electrons, while neglecting: (i) the energy of the electrons from the inner shells of atoms; and (ii) electrons involved in the formation of σ-type chemical bonds; (iii) vibrational energy of the crystal lattice. The Hamiltonian of the system takes the common form for the s–d exchange models [12,13,19]

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}}, \tag{1}$$

where

$$\mathcal{H}_0 = \sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}} a^{\dagger}_{\mathbf{k}\sigma} a_{\mathbf{k}\sigma} + \omega_0 \sum_R S^z_R,$$

is the Hamiltonian of noninteracting conduction electrons and impurities, and

$$\mathcal{H}_{\text{int}} = \sum_{\mathbf{p},\mathbf{p}'} J(\mathbf{q}) \sum_{\sigma,\sigma'} S_{\mathbf{q}} \sigma_{\sigma\sigma'} a^{\dagger}_{\mathbf{p}\sigma} a_{\mathbf{p}'\sigma'},$$

is the term responsible for the indirect exchange interaction. Here $S_q = \sum_R \exp(iqR)S_R$ (S_R being the spin vector of an impurity localized at the point R), $\mathbf{q} = \mathbf{p} - \mathbf{p}'$, $J(\mathbf{q})$ is the Fourier transform of the interaction potential between d-impurity with the conduction electrons, $\sigma_{\sigma\sigma'}$ are the Pauli matrices, $\omega_0 = -\mu \mathbf{B}/\hbar$ is the Zeeman frequency (μ is the total magnetic moment of an impurity), which will be used here to quantify the influence of an external constant magnetic field $\mathbf{B}$ applied along the z-axis; $a^{\dagger}_{\mathbf{k}\sigma}$ and $a_{\mathbf{k}\sigma}$ are respectively the creation and annihilation operators of electrons in the conduction band with spin σ and wave vector $\mathbf{k}$ with the dispersion $\epsilon(\mathbf{k}) = \epsilon_{\mathbf{k}}$.

Here, we use the following dispersion relation for the longitudinal component, $k_{\parallel}$, of the wave vector in a graphene nanoribbon [17]:

$$E_m(k_{\parallel}) = \pm \hbar v_F \sqrt{k_{\parallel}^2 + \frac{(m+\alpha)^2 \pi^2}{W^2}}, \tag{2}$$

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where v_F is the Fermi velocity. For a nanoribbon of width W, the transverse component of the wave vector can take discrete values according to the relation $k_\perp W = \pi m$, where $m = 0, \pm 1, \pm 2 \ldots$ is the integer for a ribbon sub-band. In the dispersion relation (2), the quantity α depends on the crystallographic orientation of the graphene nanoribbon and is related to the forbidden gap $\Delta = 2\delta E |\alpha|$, where $\delta E = \hbar v_F \pi / W$ is the energy difference between the sub-bands. Note that in graphene nanoribbons, the wave function must vanish at the ribbon edge, in contrast to the periodic boundary conditions commonly used in carbon nanotubes (CNTs). This leads to a drastically different quantization condition compared to the CNTs case. For instance, with CNTs, the relation $k_\perp W_{CNT} = 2\pi m$ holds, resulting in a twice larger energy gap between the sub-bands for the same length of circumference W.

In order to introduce the effects of mechanical stress into our graphene model, we base our analysis on the gauge theory. Under the constraint of external mechanical stress, an internal stress field will appear within the graphene, thus counterbalancing the initial deformation effects. The induced external stress field can conveniently be represented and characterized by an effective vector potential, $\mathbf{A}'$, which acts as a gauge field and imparts changes to the momentum of the electrons in the graphene [20,21]. In a deformed and strained graphene, all the interatomic bonds are not equivalent, so that the three hopping integrals, γ_i ($i = 1, 2, 3$), can have different values. Correspondingly, we introduce the gauge vector potential $\mathbf{A}' = (A'_x, A'_y)$, solely generated by the applied mechanical stress. Taking into account the non-equivalence of hopping integrals, one can write an effective vector potential in the form [22]

$$A'_x = \frac{\sqrt{3}}{2}(\gamma_3 - \gamma_2),$$
$$A'_y = \frac{1}{2}(\gamma_2 + \gamma_3 - 2\gamma_1). \tag{3}$$

In the case of a weakly deformed crystal lattice, and further assuming that the atomic displacement is small compared with the lattice constant a, the hopping integrals can be expanded in a series, which at the leading order in the relative atom displacement reads [23]

$$\gamma_i = \gamma + \frac{\beta \gamma}{a^2} \rho_i (\mathbf{u}_i - \mathbf{u}_0), \tag{4}$$

where γ is the unperturbed hopping integral, ρ_i the radius vector of the nearest-neighbors, $\mathbf{u}_i$ the displacement vector of the ith atom, $\mathbf{u}_0$ the displacement vector of the central atom, and β the electronic Grüneisen parameter given by

$$\beta = -\frac{\partial \ln \gamma}{\partial \ln a} \approx 2. \tag{5}$$

In the continuum limit and at the leading order, the displacement of each carbon atom can be expanded as

$$\mathbf{u}_i - \mathbf{u}_0 \propto (\rho_i \nabla) \mathbf{u}(\mathbf{r}), \tag{6}$$

where $\mathbf{u}(\mathbf{r})$ is the deformation field, assumed here to be small $\rho_i \| \nabla \mathbf{u} \| \ll u_0$. Substituting Eq. (4) in Eq. (3), while accounting for Eq. (6), we can finally obtain the components of the effective gauge field

$$A'_x = \frac{c\beta\gamma}{a}(u_{xx} - u_{yy}),$$
$$A'_y = -\frac{2c\beta\gamma}{a} u_{xy}, \tag{7}$$

where c is a numerical factor depending on the detailed model of chemical bonding, which generally can be fairly well estimated by taking the value $c \approx 1$ [22]. Taking into account the corrections related to the introduction of the gauge field, the momentum in the effective dispersion should be replaced by the generalized momentum

$$k \rightarrow k + A'.$$

Depending on the direction in which the ribbon is deformed, the dispersion relation will take two different forms

$$E_m(k_\parallel) = \pm \hbar v_F \sqrt{\left(k_\parallel + \frac{\pi}{3a} C_\parallel\right)^2 + \frac{(m+\alpha)^2 \pi^2}{W^2}} \quad \text{(longitudinal deformation)},$$

$$E_m(k_\parallel) = \pm \hbar v_F \sqrt{k_\parallel^2 + \frac{(m + C_\perp + \alpha)^2 \pi^2}{W^2}} \quad \text{(transverse deformation)},$$

(8)

where $C_\perp$ and $C_\parallel$ are the parameters associated with the introduction of the generalized momentum in the effective dispersion, which characterize the deformation of graphene nanoribbon via the corresponding gauge field. The quantities $C_{\parallel,\perp}$ correspond to a momentum, normalized in a way that 2π corresponds to the Brillouin zone edge.

3. Indirect exchange interaction

The Fröhlich method of calculation for the indirect exchange interactions [24] is based on the assumption that the matrix elements satisfy the inequality $|\mathcal{H}_{int}| \ll |\mathcal{H}_0|$. Essentially, it consists in two stages. First, a transition from the representation (1) to a new representation is carried out using a unitary transformation, $U = \exp(-\mathcal{L})$, where $\mathcal{L}$ is the anti-Hermitian operator satisfying the relation

$$\mathcal{H}_{int} + [\mathcal{H}_0, \mathcal{L}] = 0. \tag{9}$$

With this new representation, the Hamiltonian takes the modified form

$$\mathcal{H} \rightarrow \tilde{\mathcal{H}} = \mathcal{H}_0 + \frac{1}{2}[\mathcal{H}_{int}, \mathcal{L}] + O(\mathcal{H}_{int}^3). \tag{10}$$

A formal solution of the operator Eq. (9) reads

$$\mathcal{L} = \frac{1}{i\hbar} \lim_{\varepsilon \to 0} \int_{-\infty}^{0} e^{\varepsilon t} \mathcal{H}_{int}(t)\, dt,$$
$$\mathcal{H}_{int}(t) = \exp\left(\frac{i\mathcal{H}_0 t}{\hbar}\right) \mathcal{H}_{int} \exp\left(-\frac{i\mathcal{H}_0 t}{\hbar}\right).$$

The second step is to average expression (10) for the transformed Hamiltonian, $\tilde{\mathcal{H}}$, over the states of the interaction field, i.e. average using the equilibrium density matrix for the electron subsystem. This, particularly, means that the second term in the perturbation theory applied to Eq. (10), namely $\mathcal{H}_s = \frac{1}{2} \langle [\mathcal{H}_{int}, \mathcal{L}] \rangle$, ceases to depend on the electron's creation and annihilation operators, but still depends on the spin operators of different impurity atoms. Therefore, this term actually represents the *operator of the indirect interaction*. After some straightforward algebra, the operator $\mathcal{L}$ takes the form

$$\mathcal{L} = \sum_{p,p'} J(\mathbf{p}) \left\{ \frac{(S_q^x - iS_q^y) a_{p,\sigma}^\dagger a_{p',-\sigma}}{\epsilon_{p'} - \epsilon_p + \hbar\omega_0} + \frac{(S_q^x + iS_q^y) a_{p,-\sigma}^\dagger a_{p',\sigma}}{\epsilon_{p'} - \epsilon_p - \hbar\omega_0} \right.$$
$$\left. + \frac{S_q^z a_{p,\sigma}^\dagger a_{p',\sigma}}{\epsilon_{p'} - \epsilon_p} + \frac{S_q^z a_{p,-\sigma}^\dagger a_{p',-\sigma}}{\epsilon_{p'} - \epsilon_p} \right\}. \tag{11}$$

Substituting Eq. (11) into Eq. (10), and using the thermodynamic averaging yields

$$\mathcal{H}_s = \sum_{p_{1,2} R_{1,2}} M_1 \{ S_{R_1}^- S_{R_2}^- \exp\{i(p_1 - p_2)(R_1 - R_2)\}$$
$$+ S_{R_1}^- S_{R_2}^+ \exp\{i(p_2 - p_1)(R_1 - R_2)\}\}$$
$$+ \sum_{p_{1,2} R_{1,2}} M_3 \{ S_{R_1}^+ S_{R_2}^+ \exp\{i(p_1 - p_2)(R_1 - R_2)\}$$
$$+ S_{R_1}^+ S_{R_2}^- \exp\{i(p_2 - p_1)(R_1 - R_2)\}\}$$
$$+ \sum_{p_{1,2} R} M_2 S_R^z, \tag{12}$$

where the following notations for the exchange interaction constants are introduced:

$$M_1 = \frac{1}{2} J_{p_1-p_2} J_{p_2-p_1} \frac{\exp(-\beta\epsilon_{p_1})-\exp(-\beta\epsilon_{p_2})}{\epsilon_{p_1}-\epsilon_{p_2}+\hbar\omega_0},$$

$$M_2 = 2 J_{p_1-p_2} J_{p_2-p_1} \frac{\hbar\omega_0 \exp(-\beta\epsilon_{p_1})(1-\exp(-\beta\epsilon_{p_2}))}{(\hbar\omega_0)^2-(\epsilon_{p_2}-\epsilon_{p_1})^2},$$

$$M_3 = \frac{1}{2} J_{p_1-p_2} J_{p_2-p_1} \frac{\exp(-\beta\epsilon_{p_1})-\exp(-\beta\epsilon_{p_2})}{\epsilon_{p_1}-\epsilon_{p_2}-\hbar\omega_0}.$$

The last term in Eq. (12), namely $\sum_{p_{1,2}R} M_2 S_R^z$, describes the Knight shift, well-known in the theory of magnetic resonance [20], while the other terms are responsible for the indirect exchange interaction.

Exchange interaction between localized spins of the impurities is considered in the direct space, so we apply the inverse Fourier transform

$$J_{eff} = \int_0^{\pi/3a} dk_{x_1} \int_0^{\sqrt{3}k_{x_1}/3} dk_{y_1} \int_0^{\pi/3a} dk_{x_2} \int_0^{\sqrt{3}k_{x_2}/3} dk_{y_2} M_{1,3}(k_{x_1}k_{y_1}k_{x_2}k_{y_2})$$
$$\times \exp\{i(k_{x_1}-k_{x_2})x\} \exp\{i(k_{y_1}-k_{y_2})y\}. \tag{13}$$

Note that this quantity implicitly contains an oscillating part. Thus, when evaluating the integrals by the saddle point method, one can show an exponential decay in the magnitude of the indirect interaction with increasing distance between the impurities. Note that such a behavior is always observed in the presence of mechanisms of indirect exchange via conduction electrons; this being due to the local nature of the interaction between electrons and impurity atoms.

4. Results of the numerical analysis

In view of the rather complex dependence of the indirect exchange interaction on the parameters of the problem, the obtained quantities were analyzed numerically. Indirect exchange interaction between localized spins of impurities has been considered in the direct space. For this purpose it is necessary to use the inverse Fourier transform (the integration was carried out over the Brillouin zone in the reciprocal space). The numerical integration was performed using the classical trapezoidal quadrature rule. Calculations were carried out for as long as the relative error does not exceed 1%.

Fig. 1 shows the typical dependence of the exchange interaction term (see Eq. (13)) between the impurity spin components on distance, for different magnitudes of the magnetic field. Note that the different shapes of the curves in Fig. 1 must be due to the strong dependence of the electron dispersion (2) on the applied magnetic field via the Zeeman frequency ω_0. Furthermore, a reduction in the intensity of the magnetic field—equivalent to having a reduced value for ω_0—results in a slight increase in the amplitude of the indirect exchange interaction. Note, the dependence of the distribution of exchange coupling term on the magnetic field is oscillatory with attenuation (Fig. 1). The mechanism behind such a behavior is directly related to the presence of two competing processes: the pairing and de-pairing of electron spins, which are in the resonant interaction state. This observation is physically consistent with the results of the calculations of the exchange interaction in other media [12].

Figs. 2 and 3 show the dependence of the exchange interaction term, J_{eff}, on distance for a graphene nanoribbon mechanically strained in two orthogonal directions, namely transversely and longitudinally. As can be seen from these two figures, the value of the exchange interaction term is approximately ten times greater when considering a longitudinal deformation of the nanoribbon. We attribute this effect to the unboundedness and non-quantized nature of the longitudinal component of the wave vector of the electrons, $k_\parallel$, in sharp contrast with the quantized nature of $k_\perp$ when the nanoribbon is deformed transversely. The above dependence shows have an oscillating form, indicating possible areas of alternating
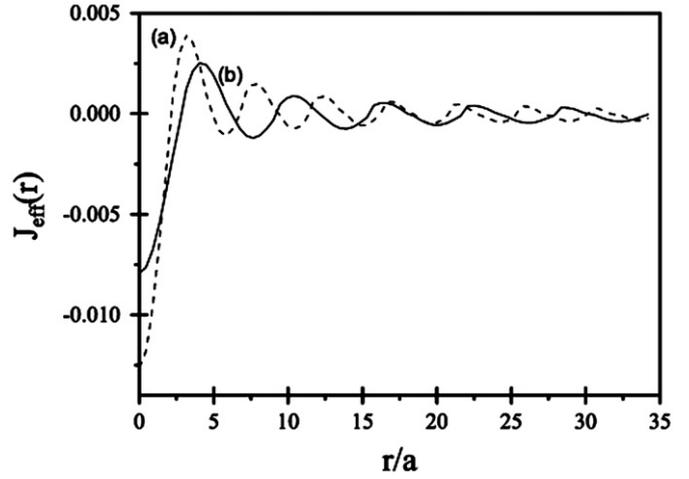

Fig. 1. The dependence of the indirect interaction constant for the nanoribbon of 20 nm width for $C_\parallel = 0.02$ and for two different magnitudes of the magnetic field: (a) $B = 1.73 \times 10^4$ T; (b) $B = 1.73 \times 10^3$ T.
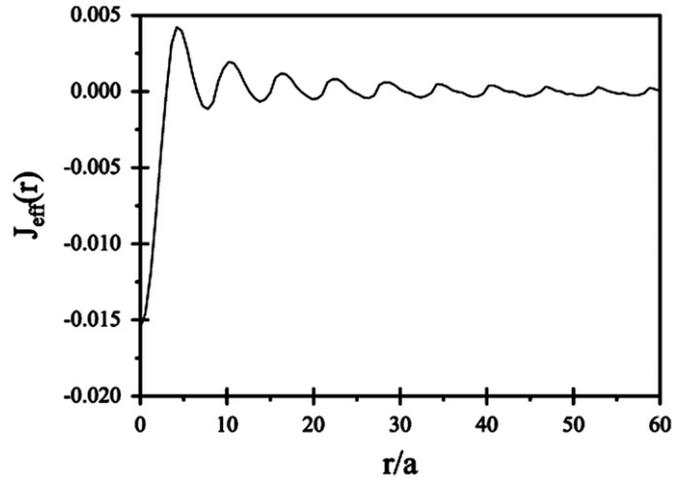

Fig. 2. The dependence of the indirect interaction constant for a nanoribbon of 20 nm width for $B = 1.73 \times 10^4$ T and $C_\parallel = 0.02$ (deformation along k_x).
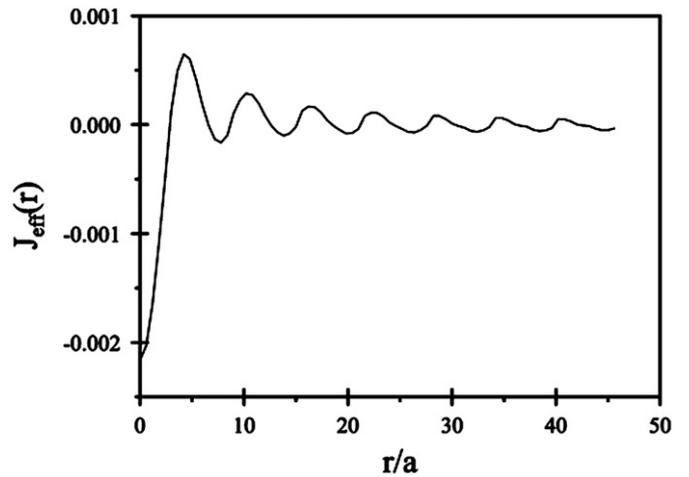

Fig. 3. The dependence of the indirect interaction constant for a nanoribbon of 20 nm width for $B = 1.73 \times 10^4$ T and $C_\perp = 0.02$ (transverse deformation).

ferromagnetic and antiferromagnetic ordering. A localized spin immersed in a conduction electrons cloud, induces a spin polarization of the cloud, which is essentially oscillatory in space. Indeed,

these oscillations are felt by any other spin present in the region of its spatial localization, thereby resulting in a global oscillating potential of interaction between the spins. These results reveal interesting and potentially important applications given the possibility of forming regions with different types of ordering (ferromagnetic or antiferromagnetic) of impurity atoms. Furthermore, the formation of these differently ordered zones can be controlled by varying a combination of two input parameters: namely the external constant magnetic field and the mechanical stress in either the longitudinal or the transverse direction.

5. Conclusions

Specific features of the indirect exchange interaction—the interaction of the electron spins in stressed graphene nanoribbons—studied in the present work, consists in the fact that the exchange interaction was examined on the basis of the Dirac approach so that dispersion relation involves electron momenta near the Dirac points. In conclusion we formulate two principal results of our study:

(i) Increase in the deformation constant reduces the energy of indirect interaction of impurities as a function of the distance.
(ii) When considering indirect interactions in the ribbon, deformed in the transverse direction, the numerical value of the exchange coupling constant is reduced by orders of magnitude.

Acknowledgments

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