

Specific Features of Indirect Interaction in an Impurity Graphene Bilayer in the Framework of the s – d Model

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Abstract—The specific features of the Ruderman–Kittel–Kasuya–Yosida interaction in an impurity graphene bilayer have been calculated in the framework of the s – d model. Hydrogen atoms are considered as impurities. The calculations have demonstrated that the antiferromagnetic impurity-spin ordering is preferable at short distances, while as the distance increases, the ordering becomes ferromagnetic. The dependences of the exchange interaction constant on the problem parameters are shown.

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1. INTRODUCTION

The second half of the 20th century, without overstatement, can be called the age of microelectronics. Over these 50 years, the world had seen a technological revolution, which had become possible due to digital logics and information technologies based on it. However, in any devices, from first transistors to modern microprocessors striking by their computing powers, microelectronics has mainly used a single property of the electron, namely, its charge. However, the electron has one more specific quantum-mechanical characteristic, namely, the intrinsic angular momentum, or spin (and its related magnetic moment), which had not attracted special attention of developers and researchers until recently. Today, the situation has changed, and a new technology, called the “spintronics” (spintronics originates from spin transport electronics or spin-based electronics), has become topical [1–3]. We remind that, in an external magnetic field, the spin-induced intrinsic magnetic moment of the electron is oriented either parallel to the vector of the magnetic induction (upward) or antiparallel (downward). In particular, ferromagnets have been used in the devices based on the spin effect.

Numerous studies devoted to the investigation of a promising material, i.e., graphene [4, 5], especially, impurity graphene [6–9], have been performed recently. In particular, the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction in the long-wavelength approximation for the dispersion law was investigated in [10–13]. The results of these works have shown that the indirect exchange interaction decreases to zero as the distance between the impurity atoms increases, which agrees with the general notions on the character of exchange interactions.

Large variations in the electronic and magnetic properties are expected upon doping or adsorption of different atoms (alkali, alkali-earth, and transition metals) on the graphene surface [14]. Currently, great attention of researchers has been focused on the calculations of variations in the electronic, magnetic, conducting, and other properties of graphene due to the attachment of a single impurity atom to its surface [15, 16]. It is known from the data obtained during the analysis of the properties of the substances with itinerant electrons that a strong modification of the electronic or magnetic properties is possible upon the adsorption of the impurity with d - or f -electrons [17]. The most known fact is apparently the Kondo effect, or the variation in the conducting properties upon the incorporation of the impurity with varying the temperature.

In view of all the listed factors make the consideration of the collective effects associated with the interaction of d - or f -impurities with electrons of the crystal lattice of graphene, i.e., the RKKY interaction [13], without the use of the long-wave approximation for the dispersion law, is an important problem.

We note that the graphene sheets treated with the hydrogen plasma can be used to fabricate devices of spintronics with controlled magnetic properties of the material by varying the external parameters.

2. BASIC EQUATIONS

Let us consider a double graphene layer consisting of two sublattices a and b in the Hückel approximation [18] taking into account the dynamics of π -electrons only. In this model, we take into account the kinetic energy of electrons and the energy of electrons of the impurity and neglect the energy of electrons of the

inner shells of the atoms and electrons participating in the formation of the σ -type chemical bonds, as well as the energy of atomic vibrations of the crystal lattice.

Let us choose the Hamiltonian of the system in the form standard for the s - d exchange models [14],

$$\begin{aligned} H &= H_0 + H_{\text{int}}, \\ H_0 &= \sum_{k, \sigma} \varepsilon_k a_{k\sigma}^+ a_{k\sigma} + \sum_{k, \sigma} \tilde{\varepsilon}_k b_{k\sigma}^+ b_{k\sigma} + \omega_0 \sum_R S_R^Z, \\ H_{\text{int}} &= \sum_{pp'} J(\mathbf{q}) \sum_{\sigma\sigma'} S_q \sigma_{\sigma\sigma'} a_{p\sigma}^+ a_{p'\sigma'} + \\ &+ \sum_{pp'} J(\mathbf{q}) \sum_{\sigma\sigma'} S_q \sigma_{\sigma\sigma'} b_{p\sigma}^+ b_{p'\sigma'}, \end{aligned} \quad (1)$$

where $S_q = \sum_R \exp(i\mathbf{q}\mathbf{R}) S_R$; $\mathbf{q} = \mathbf{p} - \mathbf{p}'$; $J(q)$ is the Fourier image of the interaction potential of the d -impurity with electrons in the conduction band; $\sigma_{\sigma\sigma'}$ are the Pauli matrices; frequency ω_0 is proportional to the external constant magnetic field applied along axis z ; S_R is the spin vector of the impurity arranged in point R ; and $a_{k\sigma}^+$, $a_{k\sigma}$, $b_{k\sigma}^+$, and $b_{k\sigma}$ are the birth and delete operators of the electrons in the conduction band with spins σ and wave vectors \mathbf{k} , which have the dispersion laws $\varepsilon(k) = \varepsilon_k$ and $\tilde{\varepsilon}(k) = \tilde{\varepsilon}_k$, respectively. For the graphene bilayer, $\varepsilon_k = -\tilde{\varepsilon}_k$.

Let us represent the dispersion law of the electrons of the graphene bilayer in the form [19]

$$E_k^{++}(V) = \pm \sqrt{\varepsilon_{k, \text{adsorb}}^2 + \frac{t_{\perp}^2}{2} + \frac{V^2}{4}} \pm \sqrt{\frac{t_{\perp}^4}{4} + (t_{\perp}^2 + V^2) \varepsilon_{k, \text{adsorb}}^2}, \quad (2)$$

where $\varepsilon_{k, \text{adsorb}}$ is the energy of electrons of the double layer of graphene changed as a result of attachment of impurity atoms, t_{\perp} is the integral of electron hopping between the layers (~ 0.3 eV), and V is the electrostatic interaction potential between the graphene layers.

The dispersion law of electrons for a single-layer impurity graphene has the following form [20]:

$$\varepsilon_{k, \text{adsorb}} = \frac{1}{2} \left[\varepsilon_a + \varepsilon_k \pm \sqrt{(\varepsilon_a - \varepsilon_k)^2 + 4 \frac{N_{\text{imp}}}{N} |V_{ka}|^2} \right].$$

Here, ε_a is the energy of the impurity atom, ε_k is the energy of electrons of the single-layered graphene [5], V_{ka} is the hybridization potential, N is the number of unit cells, and N_{imp} is the number of the impurity atoms.

The Fröhlich method for the calculation of indirect interactions [21] is based on the assumption that the inequality $|H_{\text{int}}| \ll |H_0|$ is fulfilled for the matrix elements; this method consists of two stages. The first stage is the passage from representation (1) to a new representation using the unitary transformation $U = \exp(-L)$, where L is the anti-Hermit operator satisfying the condition

$$H_{\text{int}} + [H_0, L] = 0. \quad (3)$$

In a new representation, the Hamiltonian H takes the form

$$H \longrightarrow \tilde{H} = H_0 + \frac{1}{2} [H_{\text{int}}, L] + O(H_{\text{int}}^3). \quad (4)$$

The solution of operator Eq. (3) is the following expression [1]:

$$L = \frac{1}{i\hbar} \lim_{\varepsilon \rightarrow 0} \int_{-\infty}^0 e^{\varepsilon t} H_{\text{int}}(t) dt,$$

$$H_{\text{int}}(t) = \exp\left(\frac{iH_0 t}{\hbar}\right) H_{\text{int}} \exp\left(-\frac{iH_0 t}{\hbar}\right).$$

The second stage involves averaging expression (4) for the transformed Hamiltonian \tilde{H} by the states of the carrier field of the interaction, namely, in averaging with an equilibrium statistical matrix for the electronic subsystem. Thus, the second-order term of the perturbation theory $H_{\text{ss}} = \frac{1}{2} \langle [H_{\text{int}}, L] \rangle$, where the angular brackets denote the average with the equilibrium statistical matrix for the electronic subsystem, ceases to depend on the birth and delete operators in expansion (4), but still depends on spin operators of different impurity atoms; therefore, it has the meaning of the operator of the indirect interaction.

After elementary transformations, operator L has the form

$$\begin{aligned} L &= \sum_{pp'} J(\mathbf{q}) \sum_{\sigma} \left\{ \frac{(S_q^x - iS_q^y) a_{p, \sigma}^+ a_{p', -\sigma}}{\varepsilon_{p'} - \varepsilon_p + \hbar\omega_0} \right. \\ &+ \frac{(S_q^x - iS_q^y) b_{p, \sigma}^+ b_{p', -\sigma}}{\varepsilon_{p'} - \varepsilon_p + \hbar\omega_0} + \frac{(S_q^x + iS_q^y) a_{p, -\sigma}^+ a_{p', \sigma}}{\varepsilon_{p'} - \varepsilon_p - \hbar\omega_0} \\ &+ \frac{(S_q^x + iS_q^y) b_{p, -\sigma}^+ b_{p', \sigma}}{\varepsilon_{p'} - \varepsilon_p - \hbar\omega_0} + \frac{S_q^z a_{p, \sigma}^+ a_{p', \sigma}}{\varepsilon_{p'} - \varepsilon_p} \left. \right\} \end{aligned} \quad (5)$$

$$+ \frac{S_q^z b_{p,\sigma}^+ b_{p',\sigma}}{\varepsilon_{p'} - \varepsilon_p} + \frac{S_q a_{p,-\sigma}^+ a_{p',-\sigma}}{\varepsilon_{p'} - \varepsilon_p} + \frac{S_q b_{p,-\sigma}^+ b_{p',-\sigma}}{\varepsilon_{p'} - \varepsilon_p} \left\}.$$

Substitution of (5) into (4) and thermodynamic averaging give

$$\begin{aligned} H_{ss} = & \sum_{p_1 p_2 R_1 R_2} M_1 \exp(i(p_1 - p_2)(R_1 - R_2)) S_{R_1}^- S_{R_2}^- \\ & + \exp(i(p_2 - p_1)(R_1 - R_2)) S_{R_1}^- S_{R_2}^+ \\ & + \sum_{p_1 p_2 R_1 R_2} M_3 \exp(i(p_1 - p_2)(R_1 - R_2)) S_{R_1}^+ S_{R_2}^+ \\ & + \exp(i(p_2 - p_1)(R_1 - R_2)) S_{R_1}^+ S_{R_2}^- + \sum_{p_1 p_2 R} M_2 S_z^R, \end{aligned} \quad (6)$$

where we introduced the following notation of constants of the exchange interaction:

$$M_1 = \frac{1}{2} J_{p_1 - p_2} J_{p_2 - p_1} \left\{ \frac{\exp(-\beta \varepsilon_{p_1}) - \exp(-\beta \varepsilon_{p_2})}{\varepsilon_{p_1} - \varepsilon_{p_2} + \hbar \omega_0} + \frac{\exp(-\beta \tilde{\varepsilon}_{p_1}) - \exp(-\beta \tilde{\varepsilon}_{p_2})}{\tilde{\varepsilon}_{p_1} - \tilde{\varepsilon}_{p_2} + \hbar \omega_0} \right\},$$

$$M_2 = \hbar J_{p_1 - p_2} J_{p_2 - p_1} \left\{ \frac{2 \hbar \omega_0 \exp(-\beta \tilde{\varepsilon}_{p_1}) (1 - \exp(-\beta \tilde{\varepsilon}_{p_2}))}{(\hbar \omega_0)^2 - (\tilde{\varepsilon}_{p_2} - \tilde{\varepsilon}_{p_1})^2} + \frac{2 \hbar \omega_0 \exp(-\beta \tilde{\varepsilon}_{p_1}) (1 - \exp(-\beta \tilde{\varepsilon}_{p_2}))}{(\hbar \omega_0)^2 - (\tilde{\varepsilon}_{p_2} - \tilde{\varepsilon}_{p_1})^2} \right\},$$

$$M_3 = \frac{1}{2} J_{p_1 - p_2} J_{p_2 - p_1} \left\{ \frac{\exp(-\beta \varepsilon_{p_1}) - \exp(-\beta \varepsilon_{p_2})}{\varepsilon_{p_1} - \varepsilon_{p_2} - \hbar \omega_0} + \frac{\exp(-\beta \tilde{\varepsilon}_{p_1}) - \exp(-\beta \tilde{\varepsilon}_{p_2})}{\tilde{\varepsilon}_{p_1} - \tilde{\varepsilon}_{p_2} - \hbar \omega_0} \right\}.$$

The last term in (6) describes the Night shift [1] well known in the theory of the magnetic resonance, while other terms are responsible for the indirect interaction.

Let us denote the exchange interaction of localized impurity spins in a direct space. For this purpose, let us apply the inverse Fourier transformation (in the recip-

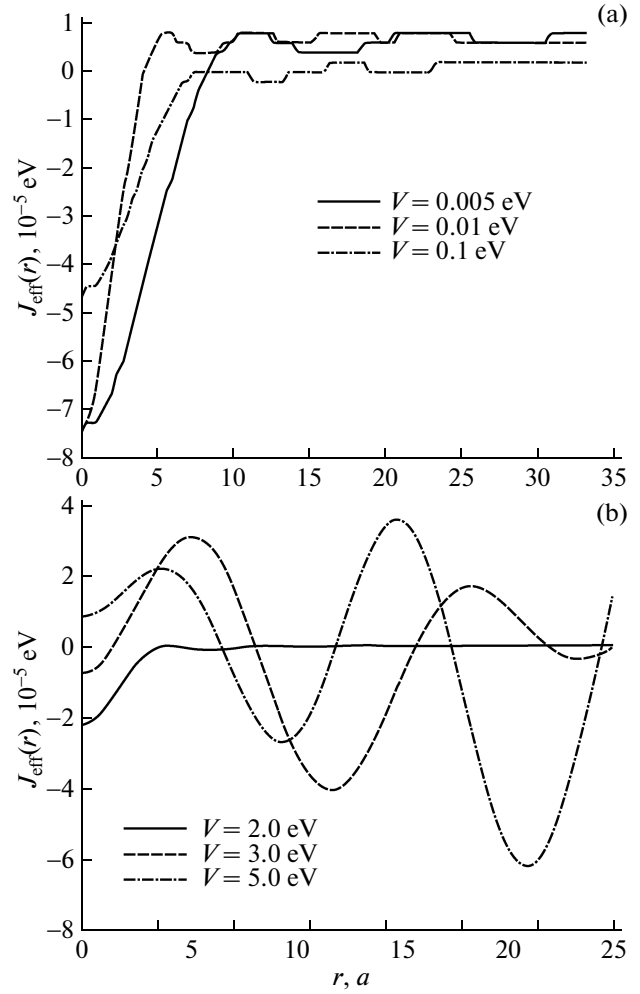


Fig. 1. Dependences of the exchange interaction constant J_{eff} on distance r for different values of the electrostatic potential V . The distance between the localized spins in all plots is expressed in units of the lattice constant $a = 2.49 \text{ \AA}$.

rocal space, we performed the integration over the Brillouin zone)

$$J_{\text{eff}} = \int_0^{\frac{\pi}{3a}} dk_{x1} \int_0^{\frac{\sqrt{3}}{3} k_{x1}} dk_{y1} \int_0^{\frac{\pi}{3a}} dk_{x2} \int_0^{\frac{\sqrt{3}}{3} k_{x2}} dk_{y2} M_{1,3}(k_{x1}, k_{y1}, k_{x2}, k_{y2}) \times \exp\{i(k_{x1} - k_{x2})x\} \exp\{i(k_{y1} - k_{y2})y\} dk_{y2}.$$

We note that this dependence already contains the oscillating part and, when evaluating the integrals by the saddle point method, we can show the presence of the exponential decay, i.e., strong decrease in the value of the indirect interaction upon increasing the distances between the impurities. We note that this behavior is always characteristic of the mechanisms of the indirect exchange through the conduction electrons; it is caused by the local character of the interaction between the electrons and impurity atoms.

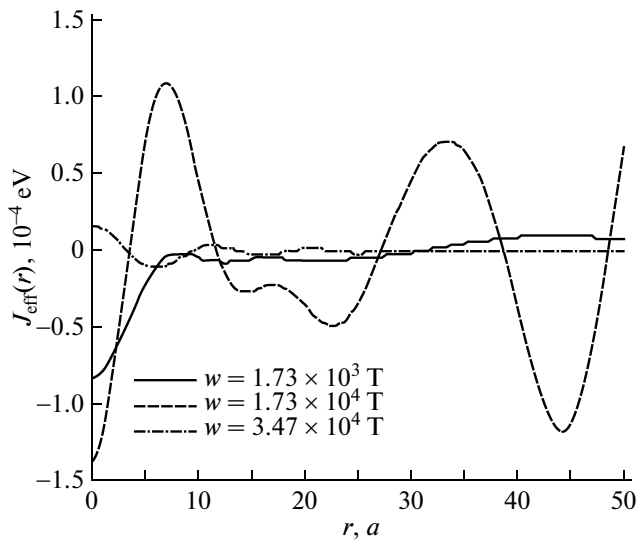


Fig. 2. Dependences of the exchange interaction constant J_{eff} on distance r for different values of the external magnetic field w .

3. RESULTS OF THE NUMERICAL ANALYSIS

By virtue of the rather complex dependence of the obtained operator of the indirect interaction on the problem parameters, the found quantities were analyzed numerically. For example, a typical dependence of the exchange interaction constant (which has the meaning of the energy of the system depending only on the orientation of the impurity spin) between the components of the impurity spin on the distance for different values of the electrostatic potential is presented in Fig. 1.

It is seen from the presented dependence that, at low external electric fields (Fig. 1a), the indirect interaction between the spins decreases to zero with a distance acquiring the ferromagnetic character, while as the field increases (Fig. 1b), the interaction character is oscillatory (which was already mentioned above; this is associated with a local character of the interaction between the electrons and impurity atoms), which indicates the possible alternation of ferromagnetic and antiferromagnetic ordering regions. Let us also note that different trends of the curves presented in Fig. 1 are associated with a strong dependence of the dispersion law of electrons (2) on the applied voltage.

The character of the distance dependence of the exchange interaction constant at different magnetic fields, which is presented in Fig. 2, is similar to the character of the dependence at different electric fields. For low fields, the indirect interaction between the spins decreases with increasing distance, while for high fields, it acquires the periodic character. We note that this result agrees with the results of the calculation of the RKKY interaction in other media. The obtained results indicate an interesting possibility of forming regions with different types of ordering (ferromagnetic

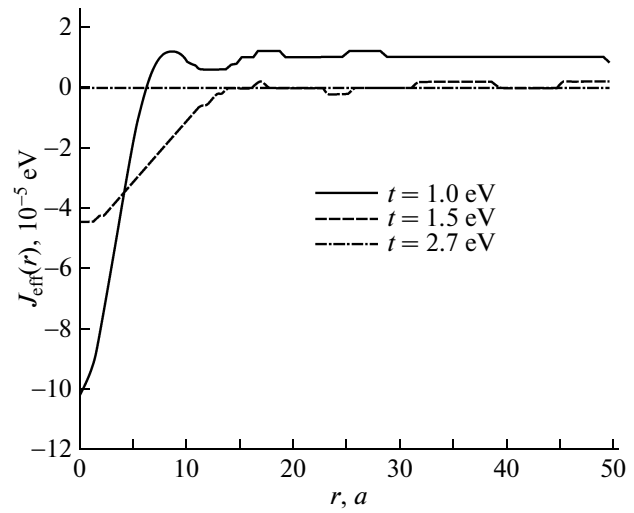


Fig. 3. Dependences of the exchange interaction constant J_{eff} on distance r for different values of the hopping integral t .

or antiferromagnetic) of impurity atoms in response to combinations of external dc electric and magnetic fields, which is undoubtedly important for applications.

The distance dependence of the exchange interaction constant for different values of the hopping integral is presented in Fig. 3. We note that, although this quantity for graphene systems is 2.7 eV, and the presented result is purely theoretical, the obtained dependence shows that the indirect spin interaction is firstly determined by the dispersion law of electrons and strongly depends on the electron energy in the conduction band.

4. CONCLUSIONS

The specific feature of the RKKY interaction of impurity atoms on the surface of a graphene bilayer was studied. The exchange interaction was considered using the total electronic spectrum of the crystal lattice (including the shorter-wavelength region). This dispersion relation involves electron pulses over the entire Brillouin zone.

In summary, we formulate the main conclusions drawn in our work.

(i) The RKKY interaction in the impurity graphene bilayer was investigated; adsorbed hydrogen atoms were used as impurities. A similar approach can be used when considering the adsorption of alkali metal atoms.

(ii) It is seen from the obtained plots that the antiferromagnetic impurity-spin ordering is preferable at short distances, and the character of the interaction becomes ferromagnetic increasing distance.

(iii) An increase in the strength of the electromagnetic field leads to a strongly oscillating dependence of the indirect interaction of impurity spins on the distance, which can be associated with the resonant electronic transitions between the split levels.

(iv) An increase in the external magnetic field also leads to a nonuniform distribution of impurity spins; in fact, we observed the formation of domain structures and the alternation of ferromagnetic and antiferromagnetic ordering regions.

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