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RKKY Interaction in Disordered Graphene

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We investigate the effects of nonmagnetic disorder on the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction in graphene by studying numerically the Anderson model with on-site and hopping disorder on a honeycomb lattice at half filling. We evaluate the strength of the interaction as a function of the distance R between two magnetic ions, as well as their lattice positions and orientations. In the clean limit, we find that the strength of the interaction decays as $1/R^3$, with its sign and oscillation amplitude showing strong anisotropy. With increasing on-site disorder, the mean amplitude decreases exponentially at distances exceeding the elastic mean free path. At smaller distances, however, the oscillation amplitude increases strongly and its sign changes on the same sublattice for all directions but the armchair direction. For random hopping disorder, no sign change is observed. No significant changes to the geometrical average values of the RKKY interaction are found at small distances, while exponential suppression is observed at distances exceeding the localization length.

I. INTRODUCTION

An unconventional behavior of the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction between magnetic impurities in undoped graphene was recently reported.[1–3] Rather than the conventional $1/R^2$ decay expected for two-dimensional systems, where R is the distance between the two magnetic moments, the RKKY interaction is found to fall off as $1/R^3$ at the Dirac (neutrality) point. Furthermore, it was found that, due to particle-hole symmetry, only ferromagnetic (antiferromagnetic) interactions are allowed when two impurities are located on the same (different) sublattice.[1]

In a recent experiment, the authors of Ref. 4 measured the Kondo effect on graphene samples with a large number of vacancies, confirming that these defects induce local magnetic moments.[5,6] Thus, upon increasing the control over the location of such defects, one might be able to also measure the RKKY interaction as a function of distance and location of local moments. Indeed, a direct detection of the RKKY interaction is feasible with the recent development of a technique to measure the magnetization curves of individual atoms using spin-polarized scanning tunneling spectroscopy.[7,8] With this technique, the orientation and distance dependence of the exchange interactions can be observed precisely.

The influence of disorder on the RKKY interaction in conventional metals has been thoroughly studied.[9–11] These studies found that the main effect of weak disorder is to randomize the electron phase, resulting in an exponential decrease of the ensemble-averaged interaction amplitude with distance. However, the average does not properly characterize the typical interaction strength, as any particular disorder configuration has long-range correlations. Indeed, the typical value, identified as the geometrical average ($J^{\rm geo}_{\rm RKKY} \equiv e^{\langle (1/2)\ln[J_{\rm RKKY}]^2\rangle_{\rm avg}}$) is found to have the same power-law behavior with distance as the amplitude of the interaction in the clean limit. Consequently, at least for conventional metals, weak disorder is not likely to cause any critical change in physical properties which derive from the RKKY interaction. It is only when the system approaches the localized regime that the geometrical average is exponentially suppressed.[12]

In light of these facts, our study focuses on two main questions. The first is how a pair of magnetic impurities in disordered graphene will interact in general. We consider impurities located along any lattice orientation, and not only along the zigzag and armchair lines. The second is how this interaction changes with increasing disorder strength.

II. KPM FORMULATION OF RKKY INTERACTION

Let us begin by considering a general expression for the RKKY exchange coupling constant in terms of the unperturbed (disorder-free) electronic Green's function $G^{(0)}(\mathbf{r}_i, \mathbf{r}_j, \omega)$,[13]

$$J_{\rm RKKY} = J^2 \frac{S(S+1)}{4\pi S^2} \int d\omega\, f(\omega)\, {\rm Im}\Big[G^{(0)}(\boldsymbol{r}_j, \boldsymbol{r}_i, \omega) \times G^{(0)}(\boldsymbol{r}_i, \boldsymbol{r}_j, \omega)\Big] \quad (1)$$

$$= J^2 \frac{S(S+1)}{4\pi S^2} {\rm Im} \int d\omega\, f(\omega) \times \sum_{n,m} \frac{F^{ij}_{nm}}{(E_n - \omega + i\delta)(E_m - \omega + i\delta)}. \quad (2)$$

Here, J is the local coupling constant between the localized magnetic impurities and the itinerant electrons, S is the magnitude of the impurity spin, i (j) is the site index of a magnetic impurity located at position $\boldsymbol{r}_i$ ($\boldsymbol{r}_j$), $f(\omega) = [e^{(\omega-\mu)/T}+1]^{-1}$ is the Fermi-Dirac distribution function, and $F^{ij}_{nm} = \psi^*_n(\boldsymbol{r}_i)\psi_n(\boldsymbol{r}_j)\psi^*_m(\boldsymbol{r}_j)\psi_m(\boldsymbol{r}_i)$, with $\psi_n(\boldsymbol{r}_i)$ denoting the eigenfunction corresponding to the eigenenergy E_n of the unperturbed electronic Hamiltonian (i.e., in the absence of magnetic disorder). The lattice constant a and $\hbar$ are set to unity in all calculations.

Using a zero-temperature approximation ($T = 0$) and changing to an integral form, Eq. (2) can be recast as

$$J_{\rm RKKY} = -J^2\frac{S(S+1)}{2S^2}\int_{E<0} dE \int_{E'>0} dE'\ \frac{F(E,E')}{E-E'}, \tag{3}$$

where $F(E,E') = {\rm Re}[\rho_{ji}(E)\rho_{ij}(E')]$, μ is the Fermi energy, and $\rho_{ij}(E) = \langle i|\delta(E-H)|j\rangle$, which can be calculated numerically using the kernel polynomial method (KPM).[14] In the KPM, the matrix elements $\rho_{ij}(E)$ are expressed as sums over order-M Chebyshev polynomials on the energy E with coefficients obtained through an efficient recursion relation involving matrix elements of the system Hamiltonian.

$$\rho_{ij} = \frac{1}{\pi\sqrt{1-E^2}}\left[g_0\,\mu_0^{ij} + 2\sum_{l=1}^{M} g_l\,\mu_l^{ij}\,T_l(E)\right], \tag{4}$$

where $T_l(E)$ is the $l^{\rm th}$ Chebyshev polynomial, $\mu_l^{ij} = \langle i|T_l(H)|j\rangle$, and g_l are the Jackson kernels coefficients. The sum is taken up to a cutoff number M. One can obtain the expansion coefficients μ_l^{ij} using the recurrence relation of Chebyshev polynomials, namely, $T_{l+1} = 2HT_l(H) - T_{l-1}(H)$ with $T_0(H) = 1$ and $T_1(H) = H$. Implicit in Eq. (4) is the normalization of the energy spectrum to a band of unity width. As our unperturbed electronic Hamiltonian with on-site disorder, we employ the single-band Anderson tight-binding model on a honeycomb lattice,

$$H = -t\sum_{\langle i,j\rangle} c_i^+ c_j + \sum_i w_i\ c_i^+ c_i, \tag{5}$$

where t (≈ 2.67 eV for graphene) is the hopping energy, c_i (c_i^+) annihilates (creates) an electron at site i, w_i is the on-site random disorder energy distributed uniformly between $[-W/2, W/2]$, and $\langle i,j\rangle$ denote nearest-neighbor sites. Periodic boundary conditions are used for all calculations. For clean systems ($W = 0$), the Chebyshev polynomials are calculated up to $M = 3\times10^3$ on a lattice with 5×10^5 sites.

III. RKKY INTERACTIONS IN CLEAN SYSTEM

The RKKY interaction coupling constant between two magnetic impurities is calculated using Eq. (3), of which the results for the clean limit are shown in Fig. 1. In order to better visualize the behavior of the amplitude in the contour plots, we have multiplied $J_{\rm RKKY}$ by R^2, resulting in a smoother ($1/R$) decay. The interactions along the zigzag and armchair directions are shown separately by line plots in Fig. 1. These results are in excellent agreement with previous studies.[1–3] The authors of Ref. 3 used a lattice Green's function method to obtain an RKKY interaction of the form

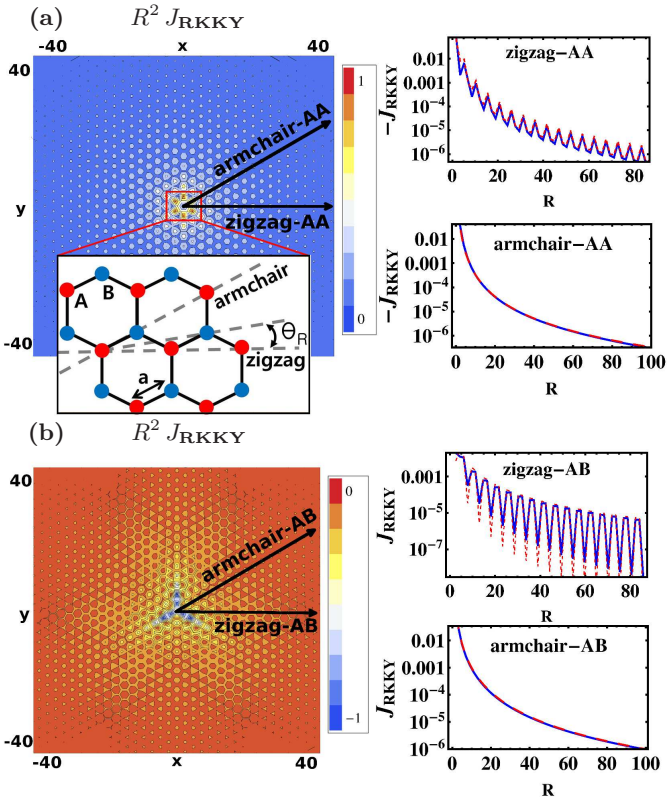

FIG. 1. (Color online) Plots of the RKKY interaction strengths between a magnetic impurity at the origin and another at: (a) a site from the same sublattice (AA) and (b) a site from a different sublattice (AB). In the contour plots, the amplitudes are multiplied by the square of the distance to facilitate visualization. The lattice constant is set to unity. The numerical data is for clean graphene ($W = 0$). Calculations using the kernel polynomial method and lattice Green's function method are represented as solid blue and dashed red lines, respectively.

$$J^0_{\rm AA} = -J^2\frac{1+\cos[(\boldsymbol{K}^+-\boldsymbol{K}^-)\cdot\boldsymbol{R}]}{R^3}, \tag{6}$$

$$J^0_{\rm AB} = J^2\frac{3+3\cos[(\boldsymbol{K}^+-\boldsymbol{K}^-)\cdot\boldsymbol{R}+\pi-2\theta_R]}{R^3}, \tag{7}$$

where all the coefficients are set to unity, $\boldsymbol{K}^\pm = (\pm2\pi/3\sqrt{3}, 2\pi/3)$ are the Dirac points in the Bloch momentum space, $\mathbf{R} = \mathbf{r}_i - \mathbf{r}_j$, and θ_R is defined in the inset of Fig. 1a. For a direct comparison, plots of Eqs. (6) and (7) are shown in Fig. 1 along with the results calculated from Eq. (3). As expected from the particle-hole symmetry of the spectrum, the magnetic impurity on the

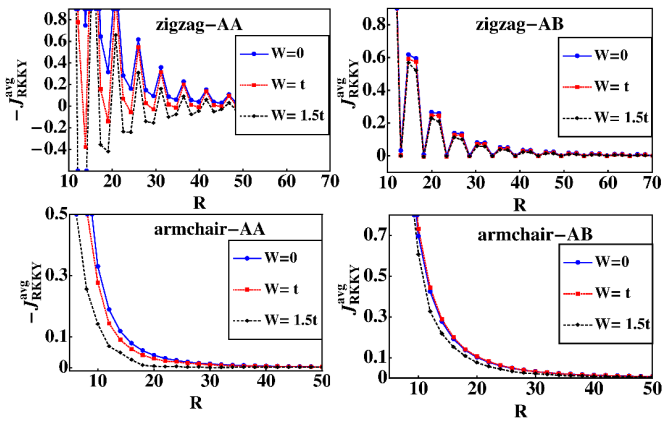

FIG. 2. (Color online) Plots of the RKKY interaction strength along the (a) zigzag and (b) armchair directions in the diffusive regime, as averaged over 1.6×10^3 different disorder configurations. A lattice with 1.8×10^5 sites and a polynomial degree cutoff of $M = 5 \times 10^3$ are used in these numerical calculations.

origin has ferromagnetic correlations with the impurities on the same sublattice (Fig. 1a), while antiferromagnetic correlations develop for impurities on different sublattices (Fig. 1b).

IV. RKKY INTERACTIONS IN DISORDERED SYSTEM

A. Diagonal defects

In order to investigate the effect of on-site nonmagnetic disorder in graphene, we consider 1.6×10^3 different disorder configurations for each value of W and then evaluate the matrix elements ρ_{ij} through the KPM with $M = 5 \times 10^3$ on a lattice with 1.8×10^5 sites.

For weak (strong) disorder strength, the system is in the diffusive (localized) regime, where the actual value of W for which this crossover occurs depends on the lattice size and has been determined by evaluating the localization length (see Fig. 4). The average amplitude of the RKKY interaction in the diffusive regime is shown in Fig. 2. Similar to conventional metals, the interaction decays exponentially with increasing disorder strength as

$$J^{\rm avg}_{\rm RKKY} \sim J^{\rm clean}_{\rm RKKY}\ e^{-R/l_e}, \qquad (8)$$

where l_e is the mean free path and $J^{\rm clean}_{\rm RKKY}$ is the interaction amplitude in the clean limit. It is worth noticing that the sign of the interaction oscillates when the impurities are located along the zigzag-AA direction.

To better characterize the amplitude of the interaction, we have also calculated the geometrical average ($J^{\rm geo}_{\rm RKKY}$) for both diffusive and localized regimes (Fig. 3). In Fig. 3a, one can see that the geometrical average for a weakly disordered system remains long ranged and has a decaying behavior similar to the clean system.

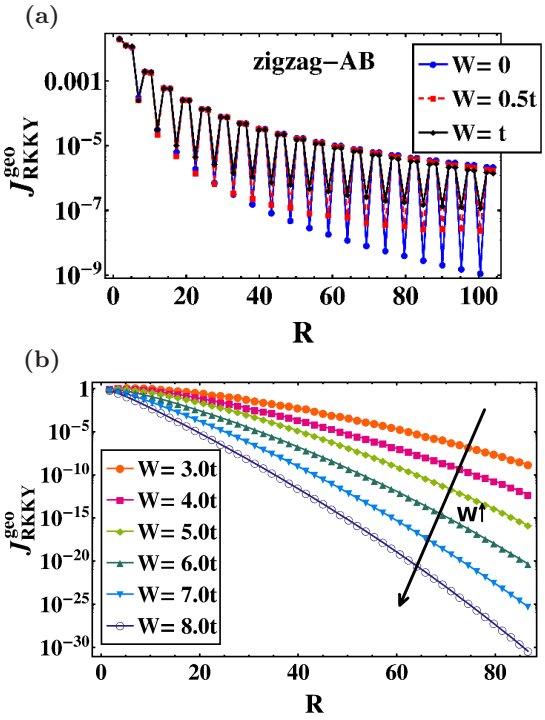

FIG. 3. (Color online) Plots of the geometrical average over 1.6×10^3 different disorder configurations of the RKKY interactions for (a) weak and (b) strong disorder. The same lattice size and polynomial cutoff of Fig. 2 are used.

As mentioned earlier, studies of conventional metals[9–11] have shown that the geometrical average (i.e., the typical value) of the RKKY interaction in weakly disordered systems has a power law dependence with the same exponent of the clean limit, namely, $J^{\rm geo}_{RKKY} \sim 1/R^{\alpha}$ (e.g., $\alpha = 2$ in a two-dimensional electron gas). Due to the unconventional distance dependence (Eqs. (6) and (7)) caused by the pseudogap at the Dirac point of clean graphene, one may expect two possibilities. If the pseudogap is not filled by disorder, the geometrical average value of the amplitude is expected to have the same exponent of the clean system, namely, $\alpha = 3$. However, if it is filled, then the geometrical average value should approach the conventional power law of a two-dimensional electron gas, namely, $\alpha = 2$. Our calculations show that the former is the correct answer. This is in accordance with the fact that short-range disorder preserves the pseudogap in graphene.[15] Therefore, the presence of weak short-range disorder in undoped graphene is not anticipated to induce any major change in physical properties related to the RKKY interaction. The situation is drastically different in the localized regime, where the geometrical average values is exponentially suppressed with distance, as shown in Fig. 3b. This behavior is captured by the following relation,[12]

$$J^{\rm geo}_{\rm RKKY} \sim e^{-R/\xi}, \qquad (9)$$

where ξ is the localization length. Fig. 4 presents the mean free path and the localization length obtained by

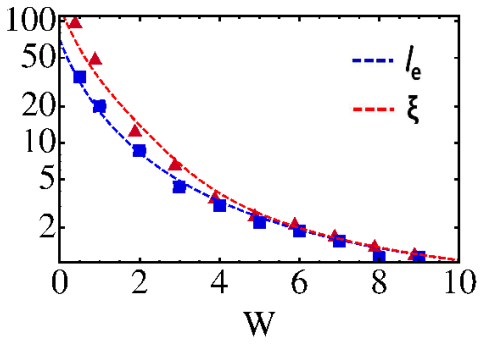

FIG. 4. (Color online) Plots of the mean free path l_e and the localization length ξ as functions of the disorder strength W (in units of t). The blue dashed line represents a fitting to the relation $l_e = c_1/W^2$ with $c_1 = 60$, whereas the red dashed line represents the resulting localization length[Eq. (10)], with $v_F\Lambda = \sqrt{10}$ treated as another fitting constant.

fitting the relations Eq. 8 and Eq. 9 to the numerical data. For $W = t$, the localization length is about 10^2, which is close to the longest linear distance possible in our calculations, namely $R_{\max} = 60\sqrt{3}$. Therefore, the system crosses over from the diffusive to the localized regime around $W \sim t$. For uncorrelated, short-range disorder, which allows for intervalley scattering, the localization length is given by $\xi = l_e \exp(\pi\sigma/G_0)$,[16,17] where $\sigma = \frac{4}{\pi}\left[\frac{(v_F\Lambda)^2}{(v_F\Lambda)^2+W^4}\right]$, v_F denotes the Fermi velocity, Λ is the energy cutoff, and $G_0 = e^2/h$ is the conductance quantum. It is well known that the mean free path is inversely proportional to the square of disorder strength ($l_e \sim 1/W^2$). Therefore, one expects the localization length to obey the relation

$$\xi \approx (c_1/W^2)\exp\left[\frac{4(v_F\Lambda)^2}{(v_F\Lambda)^2+W^4}\right], \qquad (10)$$

where c_1 is a fitting constant. Indeed, these relations fit reasonably well the numerical data, as shown in Fig. 4.

B. Off-diagonal defects

To find out the effect of disorder with no sublattice symmetry breaking, we added randomness to the hopping integral and eliminated on-site disorder ($w_i = 0$),

$$H = -\sum_{\langle i,j\rangle} t_{ij} c_i^+ c_j, \qquad (11)$$

where $t_{ij} = t + \Delta t_{ij}$, with Δt_{ij} being distributed uniformly between $[-W/2, W/2]$. We perform the same calculations of the on-site disorder case, but now with a lattice of 2×10^4 sites and a Chebyshev polynomial cutoff $M = 10^3$. For comparison, we plot the results together with those for the on-site disorder calculations in Fig. 5. A total of 4×10^2 configurations of disorder are used, with the thick dashed line indicating the average

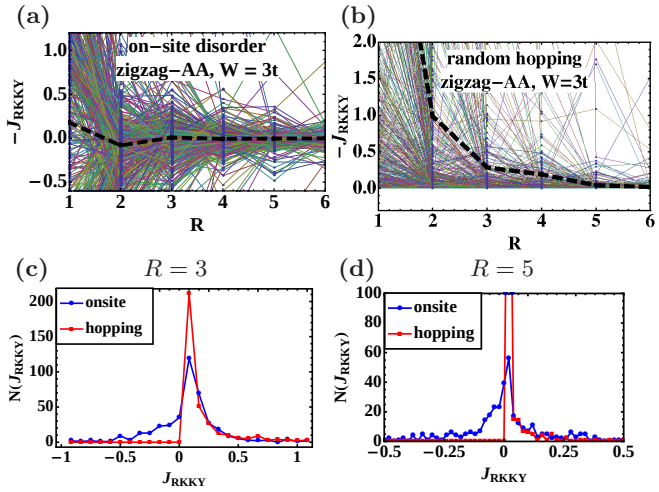

FIG. 5. Plots of the RKKY interactions along the zigzag-AA directions with strength $W = 3t$ of (a) diagonal disorder, (b) off-diagonal (random hopping) disorder, and distributions of the RKKY interactions for (c) $R = 3$, (d) $R = 5$ with 400 realizations. Here the distance unit is $\sqrt{3}a$. A lattice with 2×10^4 sites and a polynomial degree cutoff of $M = 10^3$ are used in these numerical calculations. The black dashed line is the averaged interaction.

value. While the on-site disorder generates random fluctuations in the sign and amplitude of the RKKY interaction (Figs. 5a), the hopping disorders affect only the amplitude, even in the presence of very strong randomness (Fig. 5b). The distributions of the RKKY interactions are shown in Figs. 5c,d and those show that the off-diagonal disorder (red lines) does not change the sign of the RKKY coupling.

V. CONCLUSION

In conclusion, we have confirmed that the RKKY interactions in clean graphene has a strong anisotropy of its sign and oscillation amplitude, and it decays as $1/R^3$ for all directions. Increasing the amount of nonmagnetic, on-site disorder causes the averaged amplitude of the RKKY interaction to decrease exponentially at distances exceeding the elastic mean free path, similarly to what is obtained for conventional metals. At smaller distances, however, the fluctuations of the amplitude are found to increase strongly, with sign oscillations even for a pair of magnetic impurities located on the same sublattice, for all directions except the armchair direction. When the randomness is instead applied to the hopping (off-diagonal disorder), the sign oscillations disappear. This shows that these sign changes at weak disorder potential are caused by the breaking of the sublattice symmetry, since off-diagonal disorder preserves this symmetry. Our calculations also confirm that the geometrical average of the RKKY interaction in disordered graphene has the same power law decay at short distances as in the

clean case. However, it is exponentially suppressed at distances exceeding the localization length. We plan, to extend these studies by considering the effects of long-range disorder and resonant impurities.

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