Ruderman-Kittel-Kasuya-Yosida interaction of magnetic moments in nanosized systems

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Nanosized spherical system of magnetic moments interacting indirectly via the Ruderman-Kittel-Kasuya-Yosida mechanism is studied. The interaction energy that determines the temperature of the ferromagnetic ordering depends strongly on the system size. Obtained in the mean-field approximation, dimensional and concentration dependencies of the Curie temperature testify to the necessity of taking into account the finite size of such systems to calculate their features. Results may concern both artificially constructed nanosystems and naturally arising formations (such as clusters of magnetic ions in diluted magnetic semiconductors, etc.).

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In systems with free carriers of high concentration (metals or degenerate semiconductors), indirect magnetic impurities' interaction of Ruderman-Kittel-Kasuya-Yosida (RKKY) type is considered as one of the basic mechanisms of the magnetic ordering.[1] There are a lot of papers dealing with RKKY interaction in three-, two-, and one-dimensional systems of *infinite* size.[1–4] However, to our knowledge, nobody considered how that interaction should be modified for systems of the *finite* size. It is actual because every potentially interesting electronic device is either characterized by nanosizes or includes some nanosized formations.

For example, though two-dimensional structures represent the most natural systems for the embedding in the traditional semiconductor technology, almost all theoretical works (including those "attached" to concrete experiments) are dealt with the three-dimensional systems. In addition, it is known[5] that magnetic impurities in diluted magnetic semiconductors (DMS) incline to the correlation and can form nanosized clusters. In this connection, the question arises concerning the influence of the impurity correlation on their interaction (for instance, by means of the RKKY mechanism). In the present paper, we consider that problem for the case of the spherical system of the finite radius.

Estimating the energy $w(r)=-J(r)\mathbf{S}_1\mathbf{S}_2$ of the indirect RKKY interaction of magnetic impurities with spins $\mathbf{S}_1$, $\mathbf{S}_2$ spaced at the distance $\mathbf{r}$ is based on making use of the expression

$$J(r)=\frac{J_{pd}^2}{N^2}\exp(-r/l)\sum_{\mathbf{q}}{}'\sum_{\mathbf{k}}e^{i\mathbf{q}\cdot\mathbf{r}}\frac{f(E_{\mathbf{k}})}{E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}}, \tag{1}$$

obtained in the second order of the perturbation theory.[1] Here, N is the number of lattice sites, J_{pd} is the exchange energy for the interaction of the impurity spin with a free charge carrier, $E_{\mathbf{k}}$ is the carrier energy, and $f(E_{\mathbf{k}})$ is the Fermi-Dirac function which in the degenerate case equals $f(E_{\mathbf{k}})=1$ at $k<k_F$ and $f(E_{\mathbf{k}})=0$ at $k>k_F$, where k_F is the Fermi momentum. The prime by the sum over q means that $q\neq 0$. The exponent $e^{-r/l}$ in Eq. (1) reflects the finite carrier mean free path l.

In the continual approximation, the summation in Eq. (1) is replaced by the integration which performed usually over *all* $k<k_F$ and $|\mathbf{k}+\mathbf{q}|>\mathbf{k_F}$. Then the standard result corresponding to the case of the infinite system reads[1]

$$J(r)=-I_0\Phi(r)\exp(-r/l),$$

$$\Phi(r)=\left(\frac{a}{r}\right)^4[\varphi(r)\cos\varphi(r)-\sin\varphi(r)], \tag{2}$$

where

$$I_0=\frac{1}{32\pi^3}\left(\frac{ma^2}{\hbar^2}J_{pd}^2\right),\quad \varphi(r)=2k_{F0}r, \tag{3}$$

a is the lattice constant, and $k_{F0}=(3\pi^2p)^{1/3}$ is the Fermi momentum of carriers of the concentration p.

In the case of finite system sizes or magnetic ions' clustering, the classic expression (2) for the energy of the RKKY interaction should be rectified. For simplicity, we consider the case when magnetic ions form the spherical cluster. Due to the quasineutrality, its radius R determines not only the area where ions are arranged but also the region where carriers, produced by those ions, are localized. In other words, the carriers are contained in the potential well of the radius R. Therefore, the carrier momentum k and its variation q are limited by the intervals

$$k_1\leqslant k\leqslant k_F,\quad k_1\leqslant q\leqslant k_2, \tag{4}$$

where

$$k_1\approx\pi/R,\quad k_2\approx\pi/a. \tag{5}$$

In addition, due to the spatial quantization, the distance between the energy levels of carriers grows which leads to increasing the Fermi energy and Fermi momentum while decreasing the well size: $k_F=k_F(R)$. Together, they complicate calculations and the final expression for $J(r)$ turns out to be more bulky than the canonical expression (2).

The finite mean free path l results in smearing energy levels of carriers due to their collisions. Therefore, the lowest value k is defined by the system size and equals $k_1\approx\pi/R$ only if the collision broadening $\hbar/\tau$ of levels is less than the energy $\pi^2\hbar^2/2mR^2$ of the first level. That condition could be written in the form

$$\frac{\pi^2}{R^2}-\frac{2k_F}{l}>0, \tag{6}$$

meaning that our approach relates to the small enough systems only. If, for instance, $l/a=10$ and $ak_F=1$, then R

$\lesssim 10a$. In the general case, one could use the value

$$k_1 = \sqrt{\max\left\{\left(\frac{\pi^2}{R^2} - \frac{2k_F}{l}\right), 0\right\}} \tag{7}$$

as the left boundary of inequalities (4).

To proceed one should estimate how the Fermi momentum depends on the cluster size. The total number of free carriers in the cube of the size R with the spherical Fermi surface is defined by the number of cells of the volume $(2\pi)^3$ in the phase space and in the limit of $k_F R \to \infty$ equals $N_F \sim k_F^3 R^3/(2\pi)^3$. For a finite $k_F R$ value, the number of carriers is defined by the number of points in the wave number space with coordinates divisible by $(2\pi/R)$. As none of those coordinates could equal zero, in that case

$$N_F \sim (k_F R/2\pi)^3 - \alpha(k_F R/2\pi)^2,$$

where the correction (proportional to $\alpha \sim 1$) is associated with the "forbidden" points, positioned in the coordinate planes of the wave number space and with the "correct" points neighboring the Fermi surface but not fallen inwards, as well. For the carrier concentration $n = N_F/R^3$, it follows that $n \sim k_F^3(1 - \alpha/k_F R)$. For $n = \text{const}$ and $k_F R \gg 1$ that leads to

$$k_F(R) - k_{F0} \approx \frac{\alpha}{3R},$$

where $k_{F0} \equiv k_F(\infty)$ is the Fermi momentum in the infinitely large system. Everywhere below, we use k_F to mean the value

$$k_F(R) = k_{F0} + \frac{1}{R}. \tag{8}$$

Let us turn now to calculating the energy of the RKKY interaction. Assuming $E_{\mathbf{k}} = \hbar^2 k^2/2m$, $E_{\mathbf{k+q}} = \hbar^2(k+q)^2/2m$ and designating the angle between $\mathbf{r}$ and $\mathbf{q}$ as α, and the angle between $\mathbf{k}$ and $\mathbf{q}$ as θ, we obtain

$$J(r) = \frac{2mJ_{pd}^2}{\hbar^2 N^2} \exp(-r/l) a^4 {\sum_q}' \frac{e^{iqr\cos\alpha}}{q} \sum_k \frac{f(E_k)}{2k\cos\theta + q}, \tag{9}$$

or, in the continual approximation,

$$J(r) = \frac{4}{\pi} I_0 \exp(-r/l) a^4 \int_{k_1}^{k_2} q\, dq \int_0^{\pi} e^{iqr\cos\alpha} \times \sin\alpha d\alpha \int_{k_1}^{k_F} k^2 dk \int_0^{\pi} \frac{\sin\theta d\theta}{2k\cos\theta + q}. \tag{10}$$

Noncomplicated but laborious calculations lead to rather cumbersome results which could be represented in the relatively simple form by the help of the operator $\hat{L}$ determining the value of the double definite integral $\int_{k_1}^{k_F} dk \int_{k_1}^{k_2} (\ldots) dq$ with the primitive function $\Psi(k,q)$:

$$\hat{L}\Psi(k,q) = \Psi(k_1,k_1) - \Psi(k_F,k_1) + \Psi(k_F,k_2) - \Psi(k_1,k_2).$$

Then

$$J(r) = \frac{1}{\pi} I_0 \exp(-r/l)(ak_F)^4 \hat{L}\Psi_r(k,q), \tag{11}$$

where

$$\Psi_r(k,q) = \frac{1}{(2k_F r)^4}[\Phi_1(k)c_1(k,q) - \Phi_2(k)c_2(k,q) + \Phi_3(k,q)], \tag{12}$$

$$\Phi_1(k) = 2kr\cos 2kr - \sin 2kr,$$

$$\Phi_2(k) = 2kr\sin 2kr + \cos 2kr, \tag{13}$$

$$c_1(k,q) = \mathrm{Si}[(2k+q)r] - \mathrm{Si}[(2k-q)r],$$

$$c_2(k,q) = \mathrm{Ci}[(2k+q)r] - \mathrm{Ci}(|2k-q|r), \tag{14}$$

$$\Phi_3(k,q) = \cos qr(1 + 2k^2r^2)\ln\left|\frac{2k+q}{2k-q}\right| + qr\left(\sin qr - \frac{1}{2}qr\cos qr\right)\left(\ln\left|\frac{2k+q}{2k-q}\right| - \frac{4k}{q}\right), \tag{15}$$

$$\mathrm{Si}(x) = \int_0^x \frac{\sin t}{t} dt, \quad \mathrm{Ci}(x) = -\int_x^{\infty} \frac{\cos t}{t} dt. \tag{16}$$

To the traditional situation $(R \to \infty)$ there correspond $k_1 \to 0$ and $k_2 \to \infty$. In that case, $c_1 \to \pi$, $c_2 \to 0$, $\Phi_3 \to 0$, and

$$\hat{L}\Psi \to \frac{\pi}{(2k_F r)^4}\Phi_1(k_F) = \pi\frac{2k_F r\cos 2k_F r - \sin 2k_F r}{(2k_F r)^4}.$$

Hence Eq. (11) reduces to the standard expression (2). For finite values k_1 and k_2, the interaction energy $J(r)$ should be calculated with Eqs. (11)–(15).

The local effective RKKY-field H_{RKKY}, generated in a given point, is defined by the relation $\mu H_{\mathrm{RKKY}} = \Sigma_i J(r_i)$, where r_i is the distance from that point to the ith magnetic impurity and μ is the impurity magnetic moment. In the continual approximation, the sum could be replaced by the integral $\mu H_{\mathrm{RKKY}} = \int J(r')d^3r'$, where the integration is spread over the volume occupied by impurities. Contrary to the infinite system, the value of that integral depends on the position of the considered point. For the spherical system, the effective field could be characterized by the value

$$\mu H_{\mathrm{RKKY}}^0 = 4\pi \int_{r_{\min}}^{R} J(r) r^2 dr \tag{17}$$

of that integral in the center of the sphere. Here $r_{\min}$ is the minimum distance between impurities determined by the discreteness of the crystal lattice (for instance, the minimum distance between extrinsic Mn atoms, replacing Ga atoms in GaAs lattice, amounts to $r_{\min} = a/\sqrt{2} \approx 4$ Å).

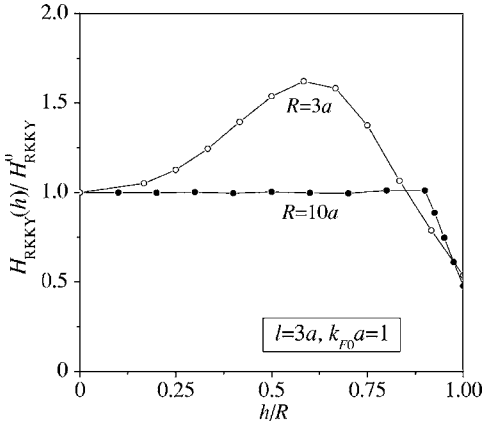

FIG. 1. Spatial distribution of the local effective field H_{RKKY} within the sphere of the radius $R=10a$ and $R=3a$ at $l=3a$ and $k_{F0}a=1$.

One could judge how much the local field H_{RKKY} would be nonuniform inside the sphere by making note that in the case with the interaction length l being comparable or shorter than the radius R, the local field at the surface of the sphere should be approximately half as large as its value in the center of the sphere. It is not hard to show that for any point inside the sphere being offset by the distance $h \leqslant R$ from its center, one could employ the relation

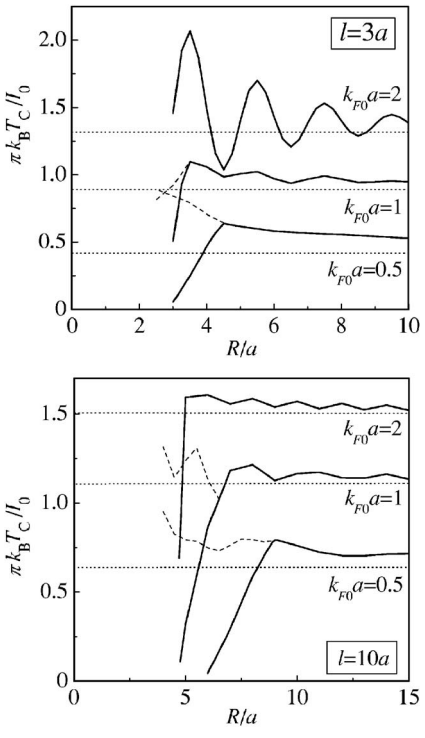

FIG. 2. Dependencies $T_{\mathrm{C}}(R)$ of the Curie temperature on the radius of spherical systems with various carrier concentrations. Upper panel: $l=3a$; lower panel: $l=10a$. Dotted lines indicate the values $T_{\mathrm{C}}(R=\infty)$; dashed curves show the "standard" (corresponding to $k_1=0$) behavior of the dependencies $T_{\mathrm{C}}(R)$ at small R.

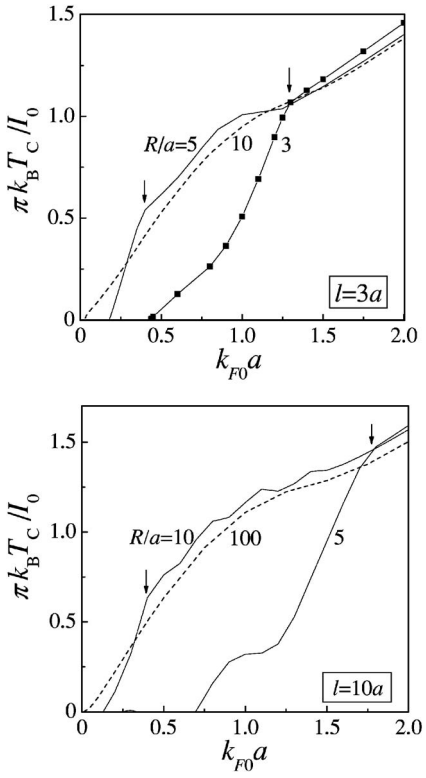

FIG. 3. Dependencies $T_{\mathrm{C}}(k_{F0})$ of the Curie temperature on the carrier Fermi momentum for spherical systems of various radii. Upper panel: $l=3a$; lower panel: $l=10a$. Arrows indicate where the condition (6) is reached.

$$\mu H_{\mathrm{RKKY}} = 4\pi \int_{r_{\min}}^{R+h} J(r) r^2 F(r) dr, \quad F(r) = \begin{cases} 1, & r < R-h, \\ \dfrac{R^2-(h-r)^2}{4rh}, & R-h < r < R+h, \end{cases} \tag{18}$$

instead of Eq. (17). In particular, for the field at the sphere surface ($h=R$), one could find

$$\mu H^S_{\mathrm{RKKY}} = 2\pi \int_{r_{\min}}^{2R} J(r)(1-r/2R) r^2 dr. \tag{19}$$

The results of numerical calculations (see Fig. 1) with $r_{\min}=a/\sqrt{2}$, $l=3a$, and $k_{F0}a=1$ show that for $R=10a$ the part of the sphere where the effective field differs from H^0_{RKKY} by no more than 20% amounts to about 85% of its volume and the average field $\bar{H}_{\mathrm{RKKY}}=(3/R^3)\int_0^R H_{\mathrm{RKKY}}(h)h^2dh \approx 0.93H^0_{\mathrm{RKKY}}$ (the same for $R=3a$ amounts to $\bar{H}_{\mathrm{RKKY}} \approx 1.14H^0_{\mathrm{RKKY}}$). Hence one could, in the first approximation, ignore the nonuniformity of the effective field and consider it as nearly uniform and equal to $\bar{H}_{\mathrm{RKKY}} \approx H^0_{\mathrm{RKKY}}$.

In the mean-field theory, Curie temperature T_{C} of the ferromagnetic state arising due to the RKKY interaction is defined by the simple relation[1]

$$k_B T_C \sim \mu \bar{H}_{RKKY} \approx \mu H^0_{RKKY}. \quad (20)$$

It is of interest to understand how the so-defined Curie temperature depends on the system size R at k_{F0}=const or varies with the carrier concentration (determined by the Fermi momentum[6]) in systems of fixed (but different) sizes.[7] Corresponding dependencies are shown in Figs. 2 and 3.

The dependence $T_C(R)$ turns out to be nonmonotone, with pronounced oscillations of the period $2a$ at higher k_{F0} values. They resulted from that part of the interaction energy $J(r)$ which is defined by the function $\Phi_3(k,q)$ given by Eq. (15), namely, by the terms $\sin qr$ and $\cos qr$ with $q=k_2=\pi/a$. That provides for the observed period.

At $R\sim l$, the Curie temperature noticeably exceeds its value for $R\to\infty$ (shown by dotted lines in Fig. 2). But the most significant distinction of the dependence $T_C(R)$ (associated with accounting the finite k_1 value) is observed at small R. As soon as the condition (6) is reached, the drop of T_C is observed, as compared to the "standard" (corresponding to $k_1=0$) dependence (shown by dashed curves in Fig. 2). However, that does not indicate the impossibility of existing ferromagnetism due to RKKY interaction in small clusters. Let us consider, for example, two DMS samples of GaAS(Mn)-type with magnetic impurities that supply charge carriers responsible for the interaction. Let the first one be the sample where magnetic impurities of the relative concentration $x\sim 10^{-3}$ are uniformly distributed over the volume and the Fermi momentum equals $k_{F0}a\approx 0.5$, and the second one the sample where the same number of impurities (per unit volume) are gathered in spherical drops with $x'\sim 10^{-1}$ resulting in $k'_{F0}a\approx 2$.[6] It follows from Fig. 2 that if $R=3-7a$ the Curie temperature in the "drop case" could be much higher than in the uniform sample.

As for the dependence $T_C(k_{F0})$, the Curie temperature increases almost monotonously with k_{F0} (i.e., with raising the carrier concentration). In that case also, reaching the condition (6) results in considerably decreasing T_C (down to zero at some finite size $R\sim l$) as compared to the $T_C(k_{F0})$ dependence for $R\to\infty$.

Of course, it would be instructive to study the considered problem for two-dimensional systems as well. Unfortunately, so far we have not succeeded in obtaining the relevant explicit relations, though one would expect similar dimensional effects for a quasi-2D case (the film of the finite nanosized thickness) too.

In conclusion, we studied nanosized spherical systems of magnetic moments interacting indirectly via the RKKY mechanism. The interaction energy which determines the temperature T_C of ferromagnetic ordering depends strongly on the system size. Obtained in the mean-field approximation, dimensional and concentration dependencies of the Curie temperature testify to the necessity of taking into account the finite size of such systems to calculate their features. Results may concern both artificially constructed nanosystems and naturally arising formations (such as clusters of magnetic ions in diluted magnetic semiconductors,[8,9] etc.).

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[7] Another way to express T_C in terms of H_{RKKY} is to relate it to the minimum effective field (at the sphere surface): $k_B T_C \sim \mu H^S_{RKKY}$. The Curie temperature so defined is half as large as that shown in Figs. 2 and 3.
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