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RKKY interactions between nanomagnets of arbitrary shape

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Abstract. – The RKKY interaction between well-separated magnetic particles in a nonmagnetic metallic matrix is calculated. It turns out that the net interaction can be mapped onto an RKKY interaction between two point-like effective moments. The effective moments exhibit a strongly oscillating dependence on the particle's size, shape, and orientation, but their magnitudes are governed by scaling laws. As a rule, magnetostatic interactions tend to suppress the RKKY effect in particles larger than about 1 nm. Surface roughness leaves the effective-moment picture unaltered but tends to yield a moderate reduction of the effective moments. The results are discussed in the context of magnetic recording, spin-glass magnetism, and cluster physics.

Introduction. – Interactions between small magnetic particles embedded in a nonmagnetic metallic matrix have attracted much attention during the past few years. This refers in particular to the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which is important for the understanding of the giant magnetoresistance of granular magnetic materials. This refers, in particular, to the magnetoresistance of granular nanostructures such as Co/Ag and Co/Cu [1-3], where there is a competition between interparticle and Zeeman interactions. A related problem is the nature of spin-glass interactions between nanoclusters [4-7].

The RKKY mechanism describes the interaction of two local magnetic moments (spins) in a sea of free electrons. Due to exchange, itinerant electrons are subject to a spin-dependent local potential, and in second-order perturbation theory the energy of the electron gas depends on whether the two localised spins are parallel or antiparallel. RKKY oscillations are akin to electron-density or Friedel oscillations caused by nonmagnetic impurities in metals and indicate that the spatial resolution of free-electron waves is of order $1/k_{\mathrm{F}}$. (For typical noble-metal hosts, such as Cu and Ag, $1/k_{\mathrm{F}}$ is about 0.8 Å.) Alternatively, the oscillations may be interpreted as rudimentary electron shells formed around impurities.

In spite of its shortcomings [8], the RKKY theory is frequently used as a starting point to describe interactions between $3d$ moments in spin glasses, granular media, and multilayers. Advanced band structure calculations [9] are now able to extend the free-electron RKKY picture to complicated intermetallics and supercells containing hundreds of transition metal atoms per unit cell. However, complex nanostructures and random macroscopic magnets remain beyond the scope of realistic first-principle calculations.

The RKKY interaction between two localized spin moments m and m' is given by the well-known expression

$$J(\xi) = J_0 \frac{2k_{\mathrm{F}}\xi \cos(2k_{\mathrm{F}}\xi) - \sin(2k_{\mathrm{F}}\xi)}{(2k_{\mathrm{F}}\xi)^4} m\, m' \,. \tag{1}$$

Here ξ is the distance between the interacting atomic moments, J_0 is an interaction parameter which depends on the effective mass of the conduction electrons, and k_{F} is the Fermi wave vector of the electron gas. In this work, the local moments are assumed to be known, although in reality the local moments depend self-consistently on the nearest-neighbour and RKKY exchange interactions, on the magnetocrystalline anisotropy, on the magnetostatic self-interaction, and on the magnetostatic field. The solution of the corresponding micromagnetic problem goes far beyond the scope of this work but requires the integral RKKY interaction as an input.

To evaluate the net RKKY interaction between two interacting nanomagnets, eq. (1) must be replaced by a sum over many atomic pairs or by an integral. It is convenient to rewrite eq. (1) as $J(\xi) = J_0 F(2k_{\mathrm{F}}\xi) m\, m'$, where $F(\eta) = (\eta \cos\eta - \sin\eta)/\eta^4$. The integral interaction is then

$$J(|\mathbf{R} - \mathbf{R}'|) = J_0 \int F(2k_{\mathrm{F}}|\mathbf{R} - \mathbf{R}' + \mathbf{r} - \mathbf{r}'|) M(\mathbf{r}) M'(\mathbf{r}') \mathrm{d}\mathbf{r}\, \mathrm{d}\mathbf{r}' \,. \tag{2}$$

Here $M(\mathbf{r})$ and $M'(\mathbf{r}')$ are the local magnetizations of the two nanoclusters (fig. 1).

Genkin and Sapozhnikov [10] have evaluated eq. (2) for two interacting spheres. For a homogeneously magnetized sphere $M(\mathbf{r})$ interacting with a point dipole $M'(\mathbf{r}') = m'\delta(0)$ they obtained a comparatively simple expression containing the sine integral $\mathrm{si}(x)$. The integration over $\mathbf{r}'$, that is the transition from a point dipole interacting with a sphere to two interacting spheres, turned out to be possible but led to quite cumbersome expressions. Furthermore, Vargas and Altbir [11] found short-distance corrections due to the discrete nature of the crystalline moment distribution in spheres.

In the limit of large distances R between the spheres, the net RKKY interaction reduces to a physically transparent effect given by the remarkable equation [10]

$$J(R) = J_0 F(2k_{\mathrm{F}}R) m_{\mathrm{eff}}\, m'_{\mathrm{eff}} \,, \tag{3}$$

where the effective spin moment

$$m_{\mathrm{eff}} = \frac{\pi M}{2k_{\mathrm{F}}^3} (\sin(2k_{\mathrm{F}}R_{\mathrm{c}}) - 2k_{\mathrm{F}}R_{\mathrm{c}} \cos(2k_{\mathrm{F}}R_{\mathrm{c}}) \,. \tag{4}$$

Here M and R_{c} are the magnetization and the radius of the sphere, respectively.

This letter deals with well-separated nanoparticles of *arbitrary* shape. First, the question is answered whether and under which circumstances it is possible to define effective moments of the type eq. (4) for aspherical particle shapes. Then a general equation for the effective spin moments is derived, and, finally, a few explicit results are reported.

Calculation and results. – Even in the exactly solvable case of interacting spheres it is not possible to derive effective moments for short distances between the spheres, and the validity of eq. (4) is limited to the asymptotic case of large distances [10]. This means, for example, that there is no point in considering dense arrays of Co clusters [12], and we have to analyze the limit of well-separated magnetic particles. The distance between the clusters is given by

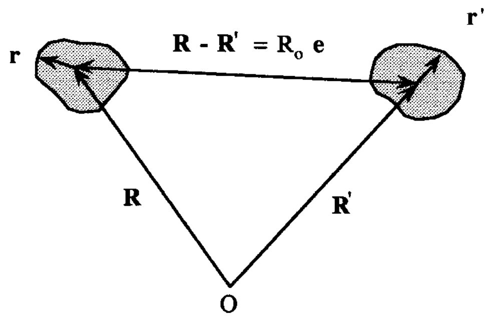

Fig. 1

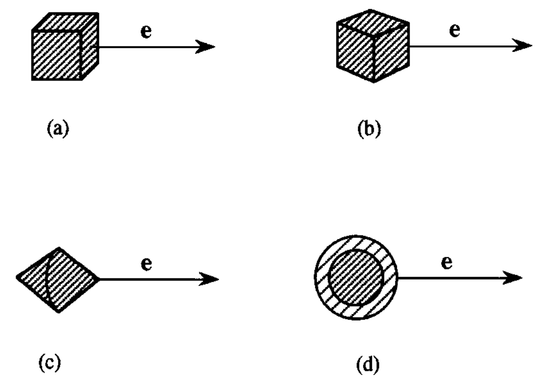

Fig. 2

Fig. 1. – Geometry of magnetic nanoparticles embedded in a nonmagnetic metallic matrix.

Fig. 2. – Some particle geometries for which effective moments are calculated: (a) aligned cube, (b) inclined cube, (c) double cone, and (d) sphere with surface roughness.

$\mathbf{R}_0 = R_0\mathbf{e} = \mathbf{R} - \mathbf{R}'$, so that for $R_0 \gg r$ and $R_0 \gg r'$ it is possible to rewrite the distance in the argument of eq. (2) as

$$|\mathbf{R}_0 + \mathbf{r} - \mathbf{r}'| = R_0 + (\mathbf{r} - \mathbf{r}') \cdot \mathbf{R}_0/R_0 + \dots. \tag{5}$$

Next we have to substitute this expression into the RKKY kernel $F(2k_\mathrm{F}(|\mathbf{R}_0 + \mathbf{r} - \mathbf{r}'|))$. The expansion with respect to $\mathbf{r} - \mathbf{r}'$ is reminiscent of but not equivalent to an ordinary multipole expansion, because RKKY interactions oscillate on an atomic $1/k_\mathrm{F}$ length scale. As a consequence, the conventional "smallness" multipole criterion $R \gg |\mathbf{r} - \mathbf{r}'|$ known from basic electrodynamics must be replaced by the stronger "quasi plane wave" multipole criterion $R \gg k_\mathrm{F}|\mathbf{r} - \mathbf{r}'|^2$. Pictorially, the perturbative quantum-mechanical oscillations of the free-electron gas, which are responsible for the RKKY interaction, are conceived as planar waves by the interacting clusters. Note that the plane-wave criterion puts the effective spin approach by Genkin and Sapozhnikov [10] in a broader context and makes it possible to evaluate eq. (2) for well-separated particles of arbitrary shapes.

The next step is to use the identities $\sin(a+b) = \sin(a)\cos(b) + \cos(a)\sin(b)$ and $\cos(a+b) = \cos(a)\cos(b) - \sin(a)\sin(b)$ to evaluate the function F. Putting the result of this calculation into eq. (2) reproduces eq. (3), but the effective spin moment is now given by

$$m_\mathrm{eff} = \int M(\mathbf{r}) \cos(2k_\mathrm{F}x)\mathrm{d}\mathbf{r}\,, \tag{6}$$

where $x = \mathbf{r} \cdot \mathbf{e}$. Note that eq. (6) is based on a well-defined particle position $\mathbf{R}$. In the case of highly symmetric particles, such as cubes and spheres, $\mathbf{R}$ refers to the particle center, but in general, $\mathbf{R}$ must be adjusted to yield a net RKKY interaction of the form $F(x)$. This is achieved by ensuring $\int M(\mathbf{r}) \sin(2k_\mathrm{F}x)\mathrm{d}\mathbf{r} = 0$. Note that eq. (6) remains valid for small clusters of magnetic atoms and also for aspherical atomic moments, such as rare-earth ions (to be published elsewhere).

It is straightforward to apply eq. (6) to a number of geometries. Of course, in the case of homogeneously magnetized spheres, the present approach reproduces the effective spin moment eq. (4), which was first derived [10] by explicit integration of eq. (2). For homogeneously

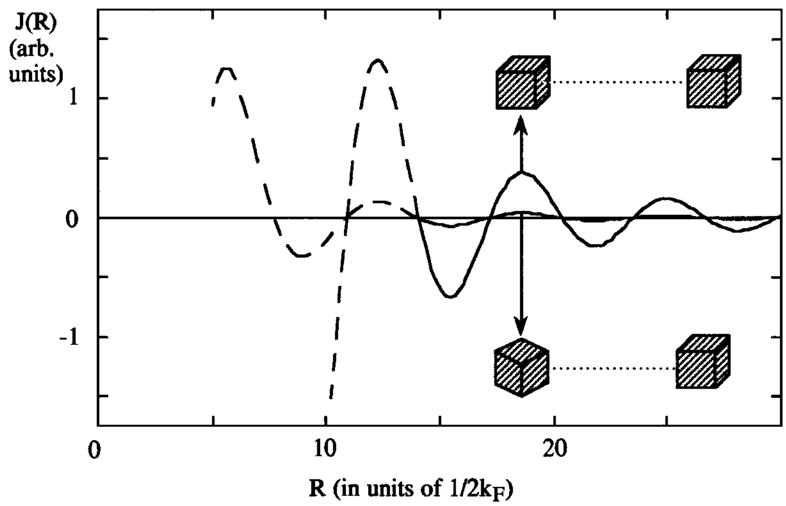

Fig. 3. – Dependence of the net RKKY interaction on the particle orientation (interaction between a cube and a sphere). The dashed parts of the lines indicate the breakdown of the present asymptotic approach. The amplitude ratio is obtained from eqs. (8) and (9) and scales as $k_\mathrm{F}L$.

magnetized and aligned double cones of diameter $2R_\mathrm{c}$ and length $2R_\mathrm{c}$ (fig. 2c) the effective spin moment is

$$m_\mathrm{eff} = \frac{\pi M}{2k_\mathrm{F}^3}(2k_\mathrm{F}R_\mathrm{c} - \sin(2k_\mathrm{F}R_\mathrm{c})). \tag{7}$$

It is interesting to note that the sign of the effective moment oscillates for spheres but not for double cones. However, in both cases the magnitude of the effective moment scales as R_c.

For cubes of volume L^3, and aligned so that $\mathbf{e}$ is perpendicular to a cube face (fig. 2a), the result is

$$m_\mathrm{eff} = \frac{ML^2}{k_\mathrm{F}}\sin(k_\mathrm{F}L)\,, \tag{8}$$

whereas the RKKY interaction of inclined cubes (fig. 2b) is characterized by

$$m_\mathrm{eff} = \frac{ML}{k_\mathrm{F}^2}(1 - \cos(\sqrt{2}k_\mathrm{F}L))\,. \tag{9}$$

This shows that the effective spin moments depend not only on the particle shape but also on the particle orientation. Note, in particular, that not only the involved trigonometric functions but also the scaling behaviour are different for the two orientations. In the case of inclined cubes, the interaction scales as L, whereas for aligned cubes it exhibits a much stronger L^2 behavior (fig. 3). The latter behavior is related to the well-known fact that the RKKY interaction is particularly strong for parallel magnetic planes, as for example in multilayers. Figure 3 compares the amplitudes of the respective RKKY interactions.

Equation (6) can also be used to study magnetic particles with rough surfaces (interfaces). For simplicity, let us consider interdiffusion so that the average moment decreases linearly over a distance of Δ. For plates of thickness t and area L^2 the effective moment is then

$$m_\mathrm{eff} = \frac{ML^2}{k_\mathrm{F}^2\Delta}\sin(k_\mathrm{F}t)\sin(k_\mathrm{F}\Delta)\,. \tag{10}$$

For spherical symmetry (fig. 2d), the influence of the roughness is calculated most conveniently from the RKKY contribution $\mathrm{d}m_{\mathrm{eff}}$ of a thin magnetic shell of thickness $\mathrm{d}r$ and magnetization $M(r)$:

$$\mathrm{d}m_{\mathrm{eff}} = -\frac{2\pi M(r)}{k_{\mathrm{F}}}\sin(2k_{\mathrm{F}}r)r\,\mathrm{d}r\,. \tag{11}$$

Integration of this equation yields

$$m_{\mathrm{eff}} = \frac{\pi M}{2k_{\mathrm{F}}^4\Delta}(2\sin(2k_{\mathrm{F}}R_{\mathrm{c}})\sin(k_{\mathrm{F}}\Delta) - k_{\mathrm{F}}\Delta\sin(2k_{\mathrm{F}}R_{\mathrm{c}})\cos(k_{\mathrm{F}}\Delta) - $$
$$-2k_{\mathrm{F}}R_{\mathrm{c}}\cos(2k_{\mathrm{F}}R_{\mathrm{c}})\sin(k_{\mathrm{F}}\Delta))\,. \tag{12}$$

For $\Delta = 0$, this equation reduces to eq. (4). In the limit of strong roughness, $\Delta \gg 1/k_{\mathrm{F}}$, both eq. (10) and eq. (12) yield a reduction of the effective moment by a factor of order $1/k_{\mathrm{F}}\Delta$. However, when the roughness involves only a few atomic layers then the net RKKY interaction is only moderately reduced. This is in agreement with numerical calculations by Altbir *et al.* [4].

Discussion and conclusions. – The question arises to what extent other interactions, such as magnetostatic interactions, are able to suppress RKKY interactions on a macroscopic scale [4-6]. This refers, in particular, to the question of spin-glass ordering in nanocomposites exhibiting giant magnetoresistance [7]. As a rule, the electrostatic nature of exchange interactions ensures that the RKKY interactions dominate on an atomic scale, for example in spin glasses. Both RKKY and magnetostatic interactions scale as $1/R_0^3$, so that the net interaction strengths are given by the effective moments. In the magnetostatic case, the moments scale as R_{c}^3, where R_{c} is the particle radius. Since the effective moments considered here scale as R_{c} (or at most as R_{c}^2), there is a transition radius R_{t} above which magnetostatic interactions dominate. According to the present calculations, R_{t} is of order 1 nm, which is comparable with numerical results according to which RKKY interactions and magnetostatic dipole interactions are of equal strengths for clusters of about 100 Co atoms [4]. This transition is also important in the context of future high-density recording media, where long-range interactions may have a deteriorating effect on the storage density and must be analyzed properly.

In conclusion, the present calculations show that the RKKY interaction between well-separated nanoclusters of *arbitrary* shapes can be mapped onto an RKKY interaction of point-like moments characterized by an effective spin moment m_{eff}. An explicit expression for the effective moments is derived, and it is shown that the effective moments exhibit a generally oscillating dependence on the particle's size, shape, and orientation. Surface roughness tends to yield a moderate reduction of effective RKKY moments.

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