Pseudodipole forces in metals and alloys of rare earths and actinides

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The skew scattering of electrons which is responsible for the anomalous Hall effect also leads to a pseudodipole interaction of localized spins. For low-temperature phase transitions, the example of a simple cubic lattice is used to show that the pseudodipole interaction may lead to an anisotropy in spin space and coordinate space. It may also lead to nonconservation of the spin in the magnetic unit cell. © *1995 American Institute of Physics.*

In this letter we discuss the nature of the pseudodipole interaction of ions of rare earths and actinides in metals and alloys. This interaction was originally introduced phenomenologically by Van Vleck[1] in an effort to explain the magnetic anisotropy of ions with a spin $S=1/2$. The structure of this interaction is the same as that of the ordinary dipole interaction, but it falls off with distance more rapidly than R^{-3}. If the state of the ion is characterized by a total angular momentum J, the pseudodipole interactions of two ions can be described by

$$V_{pd}(\mathbf{R})=\tfrac{1}{3}G(\mathbf{R})[\mathbf{J}_1\cdot\mathbf{J}_2-3(\mathbf{J}_1\cdot\hat{R})(\mathbf{J}_2\cdot\hat{R})], \tag{1}$$

where $\hat{R}=\mathbf{R}$.

We will show that the pseudodipole interaction is generated by the so-called skew scattering of conduction electrons by localized spins in the same way as for the RKKY interaction, i.e., ordinary exchange scattering. To the best of our knowledge, this is the first derivation of the pseudodipole interaction from a microscopic model.

A pseudodipole interaction has been invoked to explain the magnetic anisotropy of transition metals.[2] The question of a possible effect of the pseudodipole interaction on the critical dynamics of ferromagnets was discussed in Refs. 3–5. According to Ref. 6, however, it is apparently unimportant in that case.

As is shown below, in the case of low-temperature phase transitions, a pseudodipole interaction of nearest neighbors, $G(R_{nn})=G_{nn}$, may turn out to be comparable to the ordinary exchange interaction. As a result, since the pseudodipole interaction couples spin space with coordinate space, several qualitatively new phenomena arise. We will take a brief look at them in the particular case of antiferromagnets having a simple cubic lattice. We will show that the pseudodipole interaction may lead to an anisotropy in spin space and coordinate space and to nonconservation of the spin at a site within a magnetic unit cell.

Skew scattering was apparently first introduced in a paper by Kondo,[7] where it was shown that there is an exchange interaction between localized spins and the orbital angular momentum $\mathbf{l}$ of conduction electrons; in our case this interaction is proportional to $\mathbf{J}\cdot\mathbf{l}$. The corresponding contribution to the scattering amplitude is proportional to $i[\mathbf{k}'\times\mathbf{k}]\cdot\mathbf{J}$, where $\mathbf{k}$ and $\mathbf{k}'$ are the momenta of the electron before and after the scattering. This contribution describes a skew scattering since it changes sign under an interchange of $\mathbf{k}$ and $\mathbf{k}'$. This skew scattering leads to an anomalous contribution to the Hall conductivity—one proportional to χH, where χ is the susceptibility of the subsystem of localized spins.

We should point out that further study of the anomalous Hall effect required incorporating details of the skew scattering which are important near the Fermi surface, such as the Kondo effect and resonant scattering in mixed-valence systems.[8,9] Below we are interested in the interaction of localized spins at temperatures high in comparison with the Kondo temperature. Accordingly, the particular features of the behavior of the skew scattering near the Fermi surface are unimportant for our purposes. We will be working from the results of Refs. 7 and 10.

Following Refs. 7 and 10, we write the interaction of the conduction electrons with localized spins in the form

$$V=-\frac{1}{N}\sum_{\mathbf{k}',\mathbf{k}} a^{+}_{\mathbf{k}\mu}a_{\mathbf{k}\nu}\left\{\tfrac{1}{2}(g_J-1)\Gamma_{\mathbf{k}',\mathbf{k}}\boldsymbol{\sigma}_{\mu\nu}+\left(\frac{i}{2}\right)(2-g_J)\delta_{\mu\nu}F_{\mathbf{k}',\mathbf{k}}[\mathbf{k}'\times\mathbf{k}]\right\}\cdot\mathbf{J}, \tag{2}$$

where $\boldsymbol{\sigma}$ is the Pauli vector, and $F_{\mathbf{k}',\mathbf{k}}/k^2=F_2$ is the energy of the skew interaction. For the small radius of the f orbit, r_f, the second term in (2) arises as the result of an expansion of the exchange interaction up to second order in the parameter[7] kr_f. It is smaller than the first term by a factor on the order of $(kr_f)^2$. In this approximation, $F_{\mathbf{k}',\mathbf{k}}$ is independent of $\mathbf{k}$ and $\mathbf{k}'$.

The first term in (2) leads to the familiar RKKY interaction

$$\Delta E^{\mathrm{RKKY}}(\mathbf{R})=\frac{3\pi\Gamma^2(g_J-1)^2\rho(E_F)}{2(2k_FR)^3}\left(\cos 2k_FR-\frac{\sin 2k_FR}{2k_FR}\right)(\mathbf{J}_1\cdot\mathbf{J}_2), \tag{3}$$

where $\rho(E_F)=3/2E_F$ is the density of electron states at the Fermi surface.

The interaction of two ions which results from the second term in (2) can be written

$$\Delta E^{(\mathrm{AS})}(\mathbf{R})=-\frac{(2-g_J)^2}{N^2}\sum_{\mathbf{k}_1,\mathbf{k}_2}F_{\mathbf{k}_1,\mathbf{k}_2}[\mathbf{k}_1\times\mathbf{k}_2]_\alpha[\mathbf{k}_2\times\mathbf{k}_1]_\beta f_{k_2}$$

$$\times(1-f_{k_1})\frac{\cos[(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}]}{\epsilon_{\mathbf{k}_2}-\epsilon_{\mathbf{k}_2}}J_1^\alpha J_2^\beta, \tag{4}$$

where $\epsilon_{\mathbf{k}}$ is the energy of the electron, $f_{\mathbf{k}}$ is the Fermi function, and we need to take the principal value of the sum. It is convenient to rewrite this expression as

$$\Delta E^{(\mathrm{AS})}(\mathbf{R}) = J_{1\alpha} J_{2\beta} \epsilon_{\alpha\mu\nu} \epsilon_{\beta\rho\varphi} \Lambda_{\mu\nu\rho\varphi}(\mathbf{R}),$$

$$\Lambda_{\mu\nu\rho\varphi}(\mathbf{R}) = \frac{(2-g_J)^2}{N^2} \sum_{\mathbf{k}_1,\mathbf{k}_2} |F_{k_1 k_2}|^2 f_{\mathbf{k}_2}(1-f_{\mathbf{k}_1}) \frac{\cos[(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{R}]}{\epsilon_{\mathbf{k}_2}-\epsilon_{\mathbf{k}_1}} k_{1\mu} k_{1\rho} k_{2\nu} k_{2\varphi}. \tag{5}$$

The term proportional to $f_{\mathbf{k}_1} f_{\mathbf{k}_2}$ here is zero, since it changes sign when $\mathbf{k}_1$ is replaced by $\mathbf{k}_2$.

Let us determine the structure of the tensor Λ. The inner sum over $\mathbf{k}_1$ is a second-rank tensor. For cubic crystals it is

$$g_1(\mathbf{R},\epsilon_{\mathbf{k}_2})\delta_{\mu\rho} + g_2(\mathbf{R},\epsilon_{\mathbf{k}_2})\hat{R}_\mu \hat{R}_\rho. \tag{6}$$

Substituting this expression into the sum over $\mathbf{k}_2$, we find the following expression for the tensor Λ:

$$\Lambda_{\mu\nu\rho\varphi} = A(\mathbf{R})\delta_{\mu\rho}\delta_{\nu\varphi} + B(\mathbf{R})\hat{R}_\mu \hat{R}_\rho \delta_{\nu\varphi} + C(\mathbf{R})\delta_{\mu\rho}\hat{R}_\nu \hat{R}_\varphi + D(\mathbf{R})\hat{R}_\mu \hat{R}_\rho \hat{R}_\nu \hat{R}_\varphi. \tag{7}$$

Now substituting (7) into (4), we find

$$\Delta E^{(\mathrm{AS})} = 2[A + \tfrac{1}{3}(B+C)]\mathbf{J}_1\cdot\mathbf{J}_2 + \tfrac{1}{3}(B+C)[(J_1\cdot J_2) - 3(\mathbf{J}_1\hat{R})\cdot(\mathbf{J}_2\hat{R})]. \tag{8}$$

The first term here has the ordinary exchange structure. The second term is the pseudodipole term in (1). The structure of expression (8) is a direct consequence of the cubic symmetry of the crystal and the circumstance that the anomalous scattering is proportional to the cross product $\mathbf{k}_1 \times \mathbf{k}_2$. For crystals of lower symmetry, the expression for $\Delta E^{(\mathrm{AS})}$ is obviously more complicated. In the case of a spherical Fermi surface and a quadratic dispersion, we find the following expressions after some straightforward but lengthy calculations:

$$A = 6\pi\rho(E_F)(2-g_J)^2 F_2^2 (2k_F R)^{-5} \times \left[-\cos 2k_F R + \frac{7\sin 2k_F R}{2k_F R} + \frac{18\cos 2k_F R}{(2k_F R)^2} - \frac{18\sin 2k_F R}{(2k_F R)^3} \right], \tag{9}$$

$$B = C = \frac{3\pi}{2}\rho(E_F)(2-g_J)^2 F_2^2 (2k_F R)^{-4} \times \left[\sin 2k_F R + \frac{10\cos 2k_F R}{2k_F R} - \frac{46\sin 2k_F R}{(2k_F R)^2} - \frac{108\cos 2k_F R}{(2k_F R)^3} + \frac{108\sin 2k_F R}{(2k_F R)^4} \right]. \tag{10}$$

As was to be expected, $\Delta E^{(\mathrm{AS})}$ oscillates with the same period ($R_0 = \pi/k_F$) as the RKKY interaction. Both parts of it—the exchange part and the pseudodipole part—are proportional to R^{-4} at large values of R. In other words, they fall off more rapidly than either the RKKY interaction or a dipole interaction. At short range ($2k_F R \ll 1$), according to (9) and (10), the exchange part of $\Delta E^{(\mathrm{AS})}$ is negative, like the RKKY interaction, while the function $G(\mathbf{R}) = B(\mathbf{R}) + C(\mathbf{R})$ is positive.

At distances on the order of interatomic distances, however, Eqs. (9) and (10) are obviously not valid. For the interaction of nearest neighbors we have the estimate

$$B(R_{nn})\sim C(R_{nn})\sim F_2^2/E_F=G_{nn}.$$

In Refs. 7 and 10, F_2 was found from data on the anomalous Hall effect. It was shown[7] that we have $F_2=5$ and 20 meV for erbium and dysprosium, respectively. Values of F_2 were found in Ref. 10 for alloys of rare earth elements with Au, Ag, and Al. Series of F_2 values over the interval 1.6–6.5 meV were found. Adopting $E_F=10$ eV, we thus find that G_{nn} lies between 2.2×10^{-3} K and 0.35 K. In other words, it can range from a value small in comparison with the ordinary dipole interaction of nearest neighbors, $V_d\sim10^{-2}$ K, to a value comparable to the exchange interaction characteristic of low-temperature phase transitions in rare-earth metals and actinides, where $T_N(T_c)$ is on the order of a few degrees.

Let us examine certain physical consequences of the pseudodipole interaction. This interaction is like dipole forces in that it violates the conservation of the total spin of the system and therefore contributes to a relaxation of the uniform magnetization. Accordingly, if the pseudodipole interaction is stronger than the dipole forces and the anisotropy, it determines the frequency dependence of the uniform susceptibility, $\chi(\omega)$. Exceptional cases are ferromagnets with a dipole temperature region with $4\pi\chi>1$ (Ref. 6). In this case, dipole forces lead to demagnetization effects because of their long-range nature, and they cannot be ignored.

For low-temperature phase transitions, the pseudodipole interaction can strongly influence the structure of the ground state. We will illustrate this circumstance in the simple example of an antiferromagnet with a simple cubic lattice and a nearest-neighbor interaction. This approach is justified for pseudodipole forces because these forces fall off rapidly. A consideration of more realistic situations goes beyond the scope of this letter. For nearest neighbors, the Fourier transform of the tensor $R_\alpha R_\beta$ is $2\delta_{\alpha\beta}\cos k_\alpha$, where $\alpha=x,y,z$. As a result, we find the following expression for the spin Green's function in the random-phase approximation (the mean-field approximation):

$$G_\alpha(R)=G_0\left\{1+\frac{2G_0}{T}[J(\cos k_x+\cos k_y+\cos k_z)-G_{nn}\cos k_\alpha]\right\}^{-1}, \tag{11}$$

where $G_0=J(J+1)/3$. Under the condition $J>G_{nn}$, this expression yields the ordinary antiferromagnetic structure $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$. In this case, however, an anisotropy arises in spin space, and the spins become ordered along one of the cubic axes. In the case $G_{nn}>J$, in contrast, a nonstandard ferromagnetic structure of the $(0\frac{1}{2}\frac{1}{2})$ type arises, with spins along the x axis. Their dependence on $\mathbf{R}$ is described by

$$S_x(\mathbf{R})=\tfrac{1}{3}S_0(1+\cos\pi R_y+\cos\pi R_z). \tag{12}$$

As a result, the spin at a site is not a constant: $S_x(0,0,0)=S_0$, $S_x(0,1,0)=S_x(0,0,1)=S_0/3$, and $S_x(0,1,1)=-S_0/3$. This nonconservation of the spin at a site is a consequence of the circumstance that the pseudodipole interaction does not commute with the exchange Hamiltonian.

A pseudodipole interaction should arise when any asymmetry of the scattering in $\mathbf{k}$ space leads to an anomalous Hall effect. Accordingly, a pseudodipole interaction can arise in the cases of skew scattering of types other than the $\mathbf{J}\cdot\mathbf{l}$ coupling discussed above. In particular, this interaction arises in the versions of skew scattering discussed in Refs. 8

and 9. In those cases, however, the pseudodipole interaction will be significant only if the skew scattering occurs over an energy shell on the order of E_F, rather than exclusively near the Fermi surface. Expressions such as (9) and (10) are derived by integrating over the entire Fermi sphere.

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