# Dzyaloshinskiĭ-Moriĭ electron-nuclear interaction in orthoferrites

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It is shown that if the experimental NMR data for the orthoferrite $YFeO_3$ are interpreted correctly, they do not only indicate unambiguously the existence of antisymmetric indirect hyperfine interaction (an electron-nuclear analog of the Dzyaloshinsky–Moriy interaction) but also yield a quantitative estimate of the interaction. The contribution of this interaction to the hyperfine interaction anisotropy tensor may exceed the corresponding contribution of the symmetric anisotropic terms.

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The Dzyaloshinskiĭ-Moriĭ interaction, or the antisymmetric exchange

$$V_{as} = \mathbf{D}_{12} \cdot [\mathbf{S}_1 \mathbf{S}_2] \quad (1)$$

($\mathbf{D}_{12}$ is the Dzyaloshinskiĭ vector, $\mathbf{S}_1$ and $\mathbf{S}_2$ are the spins of the magnetic ions) is one of the principal interactions in a wide class of magnetic materials, especially in the weak ferromagnets, determining their magnetic, magnetooptic, and resonance characteristics. Moreover, in these materials there is, besides the antisymmetric exchange interaction of the electron spins (1), an electron-nucleus antisymmetric exchange

$$V_{as}^{el-nucl} = \mathbf{d}_{12} \cdot [\mathbf{I}_1 \mathbf{S}_2], \quad (2)$$

where $\mathbf{I}_1$ is the nuclear spin, and $\mathbf{d}_{12}$ is the electron-nuclear analog of the Dzyaloshinskiĭ vector. Such an interaction, and the possibility of its experimental observation in the weak ferromagnets, were first studied by Ozhogin.[1] In Ref. 2 a microscopic theory of the Dzyaloshinskiĭ-Moriĭ electron-nuclear interaction was constructed, along with quantitative estimates. Nevertheless, the problem of experimental observation and determination of the magnitude of the electron-nuclear antisymmetric exchange has not been solved at present.

In the present work, it is shown that a more detailed analysis of the known experimental data on the field dependence of the $^{57}Fe$ NMR frequency in the orthoferrite $YFeO_3$ not only unambiguously demonstrates the existence of the electron-nuclear antisymmetric exchange, but allows us to find its magnitude.

First we consider the coupling of the local field of the magnetic structure of the orthoferrites to the $^{57}Fe$ nucleus. Taking account of the four-component magnetic structure of the orthoferrites $RFeO_3$ with a nonmagnetic R ion (La, Y, Lu), the local field at the nucleus $^{57}Fe_i$ in one of the 4*b* positions can be written in the form[3,4]

$$H_{loc}(i) = a_G(i)\mathbf{G} + a_F(i)\mathbf{F} + a_C(i)\mathbf{C} + a_A(i)\mathbf{A} + \hat{a}(i)\mathbf{G} \quad (3)$$

(**G**, **F**, **C**, **A** are the basis vectors of the magnetic structure, so that $\mathbf{G}^2 + \mathbf{F}^2 + \mathbf{C}^2 + \mathbf{A}^2 = 1$), where the first four terms represent the contribution of the isotropic hyperfine interaction (HFI), and the last, the anisotropic HFI. The notation of Eq. (3) takes into account that in the orthoferrites *F*, *C*, and *A* are $\sim 10^{-2}$ *G*, and the components of the anisotropic local field tensor $\hat{a}$ are less than $\sim 1\%$ of the primary isotropic contribution $H_0 = a_G G$.

Information on the anisotropy of the HFI is taken from study of the field dependence of the $^{57}Fe$ NMR frequency in the domains. We shall consider a specific example, the orthoferrite $YFeO_3$, with a known magnetic configuration $\Gamma_4(G_x, A_y, F_z)$.

In an applied magnetic field **h** parallel to the *c*-axis, the field dependence of the NMR frequency has the form

$$\nu_{\Gamma_4}(\mathbf{h}\|\mathbf{c}) = 1 - a_{xx} + (a_{zx}G_x + a_F F_z)h + h^2/2, \quad (4)$$

where for convenience all "field" quantities ($a_{zx}$, $a_F$, $a_{xx}$, $h$) are specified in units of $H_0$ (in $YFeO_3$ at $T = 4.2$ K we have $H_0 = 551$ kOe),[5] and $\nu$ is in units of $\nu_0 = \gamma H_0/2\pi$ ($\gamma/2\pi = 0.138$ Mhz/kOe). Here and below we assume for definiteness that in the configuration $\Gamma_4$ for $F_z > 0$ the component $G_x > 0$.

Thus the derivative $(\partial\nu/\partial h)_{h=0}$ under these assumptions is the sum of two terms:

$$[\partial\nu_{\Gamma_4}(\mathbf{h}\|\mathbf{c})/\partial h]_{h=0} = a_{zx}G_x + a_F F_z, \quad (5)$$

which by convention we call ferromagnetic ($a_F F_z$) and antiferromagnetic ($a_{zx} G_x$).

In an applied magnetic field **h** parallel to the *a*-axis, we induce a $\Gamma_4 - \Gamma_2$ ($F_x$, $C_y$, $G_z$) spin-reorientation transition

$$\nu_{\Gamma_{24}}(\mathbf{h}\|\mathbf{a}) = 1 - (a_{xx}G_x^2 \pm 2a_{zx}^{(s)} G_x G_z + a_{zz}G_z^2) + (a_{xz}G_z + a_F F_x \pm a_G G_x)h + h^2/2 \quad (6)$$

($a_{zx}^{(s)}$ is the symmetric part of $a_{zx}$: $a_{zx}^{(s)} = (a_{zx} + a_{xz})/2$), where the $\pm$ signs refer to nuclei in the positions 1,3 and 2,4 (in the designation of Ref. 4). In this case splitting of the NMR frequency occurs, with the size of the splitting given by

$$\Delta\nu = 2(2a_{zx}^{(s)} G_z + h)G_x \quad (7)$$

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and its field dependence is used to find the parameter $a_{zx}^{(s)}$. According to Ref. 5, in $YFeO_3$ $a_{zx}^{(s)} = 3.2\times 10^{-3}$. From the data on splitting of the NMR lines by the spontaneous spin-reorientation transition $\Gamma_4 - \Gamma_2$ similar values are obtained for a series of orthoferrites: $a_{zx}^{(s)} = 3.4\times 10^{-3}$ in $ErFeO_3$ and $|a_{zx}^{(s)}| = 2.9\times 10^{-3}$ in $HoFeO_3$.[4]

The anisotropy tensor $\hat{a}$ of the local field is, in general, nonsymmetric. In fact, if we assume $\hat{a}$ to be symmetric, then in $YFeO_3$ (at $T = 4.2$ K) we have

$$[\partial \nu_{\Gamma_4}(\mathbf{h}\|\mathbf{c})/\partial h]_{h=0} = (3.2-8.6)\cdot 10^{-3} = -5.4\cdot 10^{-3},$$

where we have used the value[5]

$$a_F = 2H_{\text{IHFI}}/H_0 - 1 = -0.79$$

($H_{\text{IHFI}}$ is the contribution of the indirect HFI $^{57}Fe - O^{2-} - Fe^{3+}$ in the local field) and the most accurate value of the angle of inclination of the magnetic sublattice $F_z = 10.9\times 10^{-3}$.[6]

The experimental value of the quantity (5), obtained in Ref. 5, is $-10.2\times 10^{-3}$; that is, almost twice (!) as large. Therefore, the assumption that the HFI anisotropy tensor is symmetric is inconsistent. Comparing (5) with experiment,[5] we find

$$a_{zx} = -1.6\cdot 10^{-3},$$

so that the antisymmetric part

$$a_{zx}^{(a)} = -4.8\cdot 10^{-3},$$

exceeds the symmetric part. We notice that in Ref. 5 the "antiferromagnetic" contribution to the derivative (5) was omitted, although the corresponding to the NMR frequency splitting was taken into account.

The nature of the antisymmetric part of the tensor $\hat{a}$ can be connected only to the antisymmetric indirect HFI (AIHFI)

$$\hat{V}_{\text{AIHFI}} = \sum_{i\neq j} \mathbf{d}(ij)\cdot[\mathbf{I}_i\mathbf{S}_j]$$

—the electron-nuclear analog of the Dzyaloshinskiĭ-Moriĭ interaction—so that

$$a_{zx}^{(a)}(i) = -\frac{S}{g_n\beta_n}\sum_j d_y(ij). \tag{8}$$

The interaction appears in the result of the calculation along with the usual indirect hyperfine $^{57}Fe$–$O^{2-}$–$Fe^{3+}$ interaction and the spin-orbit interaction for the $Fe^{3+}$ ion. In accordance with the results of the microscopic theory,[2] the electron-nuclear analog of the Dzyaloshinskiĭ vector, $\mathbf{d}(ij)$, depends on the geometry of the $^{57}Fe$–$O^2$–$Fe^{3+}$ bond:

$$\mathbf{d}(ij) = d(\theta)\,[\mathbf{r}_i\mathbf{r}_j], \tag{9}$$

where $\mathbf{r}_i$, $\mathbf{r}_j$ are the unit radius vectors of the anion-cation bonds, and

$$d(\theta) = d_1 + d_2\cos\theta, \tag{10}$$

where $\theta$ is the cation-anion-cation bond angle.

For a rough estimate of the order of magnitude of the parameter $d$ one can use the relation

$$d/A_{\text{IHFI}} \lesssim \xi/\Delta E,$$

where $\xi$ is the one-electron parameter of the spin-orbit interaction for the $d$-electron; $\Delta E$ is the energy of the $^4T_1$-type excited terms for the $Fe^{3+}$ ion; $A_{\text{IHFI}}$ is the constant in the isotropic indirect HFI:

$$\hat{V}_{\text{IHFI}} = \sum_{i\neq j} A_{\text{IHFI}}(ij)\,\mathbf{I}_i\mathbf{S}_j. \tag{11}$$

Using in our case $\xi \lesssim 5\times 10^2$ cm$^{-1}$, $\Delta E \gtrsim 10^4$ cm$^{-1}$, we find

$$d/A_{\text{IHFI}} \lesssim 5\cdot 10^{-2},$$

which agrees well with the magnitude of the ratio

$$|a_{zx}^{(a)}/H_{\text{IHFI}}| \approx 4.6\cdot 10^{-2}.$$

obtained from analysis of the experimental data.[5]

This ratio can be compared with the ratio of the Dzyaloshinskiĭ field $H_D$ to the exchange field $H_E$: in $YFeO_3$ we have $H_D/H_E \approx 2.2\times 10^{-2}$. That these ratios are comparable in magnitude is reasonable if we consider that in accordance with the exchange-relativistic nature of the Dzyaloshinskiĭ field $H_D/H_E \sim \xi/\Delta E$, which means that $|a_{zx}^{(a)}/H_{\text{IHFI}}| \sim H_D/H_E$. In other words, if $H_{\text{IHFI}}$ is the electron-nuclear analog of the exchange field, then $H_{\text{AIHFI}} = |a_{zx}^{(a)}|$ is the electron-nuclear analog of the Dzyaloshinskiĭ field. In the orthoferrite $YFeO_3$

$$H_{\text{IHFI}} = 58 \text{ kOe}, \quad H_{\text{AIHFI}} = 2.6 \text{ kOe}.$$

For an approximate value of the "electron-nuclear" Dzyaloshinskiĭ field in the weak ferromagnets we can use the relation

$$H_{\text{AIHFI}} \approx (H_D/H_E)H_{\text{IHFI}}.$$

Thus, our analysis of known experimental data[5] on the field dependence of the $^{57}Fe$ NMR frequency in $YFeO_3$ makes it possible to specify quantitatively the electron-nuclear Dzyaloshinskiĭ-Moriĭ interaction $^{57}Fe$–$O^{2-}$–$Fe^{3+}$.

Naturally, this interaction should be observable in other weak ferromagnets as well. We note that in the "easy-planar" phase of such rhombohedral weak ferromagnets as $FeBO_3$, $FeF_3$, $\alpha$-$Fe_2O_3$, the "antiferromagnetic" contribution to the derivative $[\partial\nu(\mathbf{h}\perp\mathbf{c}_3)/\partial h]_{h=0}$ comes only from the electron-nuclear Dzyaloshinskiĭ-Moriĭ interaction:

$$[\partial\nu(\mathbf{h}\perp\mathbf{c}_3)/\partial h]_{h=0} = a_{xy}^{(a)} l_y + a_F m_z, \tag{12}$$

so that its detection and numerical estimate in these compounds is, from the theoretical point of view, easier than in the orthoferrites.

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