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Abstract

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On the Theory of the Anisotropy of Indirect-Exchange Interactions

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It is known that certain antiferromagnets, for example α-Fe_2O_3, carbides of Mn and Co, exhibit spontaneous magnetization. However, the magnitudes of their magnetic moment are extremely small ([1]). Such antiferromagnets are customarily called weak ferromagnets. It is obvious that they are not exchange ferromagnets, since the magnitude of the magnetic moment in ordinary exchange ferromagnets is several orders of magnitude greater (except in the vicinity of the Curie temperature).

A phenomenological theory of the phenomenon of weak ferromagnetism was constructed in ([2]). It turned out that taking account of the magnetic symmetry of the crystals in the expression for the free energy explains this phenomenon. However, in ([2]) the microscopic cause of weak ferromagnetism was not revealed. This was done in ([3]) on the basis of the "new model" of indirect exchange ([4]).

The purpose of the present work is to consider the microscopic source of the indicated and related phenomena from the point of view of a more general model, i.e., to construct a theory of the anisotropy of indirect exchange, magnetoelectric and piezomagnetic effects, starting from a many-electron model ([5]). Thus, the present work is a direct continuation of ([6]).

Let us consider an ionic lattice consisting of two types of sites—magnetic (g) and nonmagnetic (f). To describe the ground state we shall start from the Hamiltonian in the second-quantization representation

$$H = \sum_{(aa')} L(aa')\, a_a^+ a_{a'} + \frac{1}{2} \sum_{a_1 a_2 a_1' a_2'} F(a_1 a_2;\, a_1', a_2')\, a_{a_1}^+ a_{a_2}^+ a_{a_2'} a_{a_1'}. \tag{1}$$

Here and below we shall adhere to the notation of ([6]).

Only $L(aa')$ will contain an additional term due to spin-orbit interaction of the form

$$\left\langle \theta(q) \left| \left\{ \sum_{a''} \nabla U_{a''}(q)\, [S_{a''} \times P_{a''}] \right\} \right| \theta(q) \right\rangle \equiv \Lambda^{LS}. \tag{2}$$

Next, after writing the Hamiltonian in terms of magnetic (g) and nonmagnetic (f) sites, we apply perturbation theory for a degenerate level ([5]). We shall take the overlap of the wave functions of magnetic and nonmagnetic sites as a small parameter. In this case, as usual ([6]), the number of overlaps will correspond to the order of the perturbation-theory series. It is shown in ([6]) that, under such a treatment, the first terms different from zero appear only in the fourth approximation of perturbation theory ([5]). We list them:

$$\varepsilon^4 (H_1 - PH_1P)(H_0 - E_0)^{-1}(H_1 - PH_1 - \Delta_0), \tag{3}$$

$$(H_1 - PH_1 - \Delta_0)(H_0 - E_0)^{-1}(H_1 - PH_1P),$$

$$\varepsilon^4 PH_2(H_0 - E_0)^{-1}H_1(H_0 - E_0)^{-1}H_1P. \tag{4}$$

Here, owing to the spin-orbital interaction, the following will be added to the expression for the work (6):

$$\sum_{(fg\sigma\sigma')} a_{f\sigma}\Lambda^{LS}(fg)\, a_{g\sigma'} + \sum_{gf\sigma\sigma'} a_{g\sigma}\Lambda^{LS}(gf)\, a_{f\sigma'}. \tag{5}$$

It is interesting to note that the first of these expressions depends only on H_1, which contains only transfer integrals. In the second expression there also occur exchange terms due to H_2. In what follows we shall see that (3) leads to a band picture (of the Goodenough type ([7])), while (4) leads to the usual exchange picture. Substituting into (3) and (4) the expression for H_1, taking (2) and H_2 into account, after very cumbersome calculations of all matrix elements we obtain the final expression for the anisotropic indirect-exchange Hamiltonian

$$\sum_{(g_1 g_2)} D^{(3)}_{g_1 g_2}\, [\overline{S}_{g_1} S_{g_2}], \tag{6}$$

where

$$D^{(3)}_{g_1 g_2} = \sum_{(f_1 f_2)} \frac{\Lambda(f_1 g_1)\Lambda(g_1 f_2)\Lambda(f_2 g_2)\Lambda^{LS}(g_2 f_1)}{\Delta_1(g_1 f_2)\Delta_2(f_2 g_2)\Delta_3(g_2 f_1)}, \tag{7}$$

$$\sum_{g_1 g_2} D^{(0)}_{g_1 g_2}\, [S_{g_1} S_{g_2}], \tag{8}$$

where

$$D^{(0)}_{g_1 g_2} = \sum \frac{\Lambda(f_1 g_1) F(g_1 f_2;\, f_2 g_2) \Lambda^{LS}(g_2 f_1)}{\Delta(g_1 f_2;\, f_2 g_2) \Delta(g_2 f_1)}. \tag{9}$$

D^3 and D^0 contain the first order in the spin-orbital interaction. The spin part of the first sum of expression (6) coincides with the expression obtained according to the "new model" of indirect exchange ([4]).

The expressions for the D orbital ([3]) differ from ours, since our treatment was based on a many-electron theory. Such a treatment has one further advantage. As is evident, formula (6) simultaneously includes both the band and the exchange picture. The predominance of one or the other model (band or exchange) will depend on the type of compound under consideration.

From formula (6) one can readily obtain the Hamiltonian for weak ferromagnetism. For example, applying the symmetry of α-Fe_2O_3 to (6) leads to the Hamiltonian that was obtained from an expansion of the thermodynamic potential in a series in the relative magnetization, with subsequent use of crystallographic and magnetic symmetry.

Thus the phenomenon of weak ferromagnetism is an intrinsic property of the crystal itself, and is not an effect associated with any defect of the crystal.

Formula (6) may also be used to describe the purely antiferromagnetic case. In this case, using the temperature Green' s function technique ([8,9]), the frequency of the uniform precession was calculated, and it was shown that in the spin-wave spectrum there is a gap proportional to $(Z_1 J_1 / Z_2 J_2)^{1/2} D_{ZZ}$*.

If we now retain also the second order in the spin-orbital interaction, then a term from (2) and (3) will be added to the Hamiltonian,

$$\sum_{g_1 g_2} S_{g_1} \Gamma_{g_1 g_2} S_{g_2}, \tag{10}$$

where $\Gamma_{g_1 g_2}$ is a tensor. In view of the fact that we shall not need this tensor in what follows, its explicit expression is not given here because of its unwieldiness.

We now proceed to consider the microscopic mechanism of the magnetoelectric effect ([1]). For this purpose, in expression (2) it is necessary

* These results were reported at the conference on ferro- and antiferromagnetism in Leningrad in 1960.

include matrix elements of electric or magnetic fields. For example, in an electric field the term

$$\Lambda^{\mathrm{III}} \div e(\theta^*_m(q)\, \overline{E}\, q\, \theta_n(q)). \tag{11}$$

is added to H_1. Using the scheme set forth, with allowance for (11), one can obtain the Hamiltonian describing magnetoelectric phenomena

$$\sum_{g_1 g_2} D^{(0)}_{g_1 g_2}(E)[\mathbf{S}_{g_1}\mathbf{S}_{g_2}] + \sum_{g_1 g_2} D^{(3)}_{g_1 g_2}(E)[\mathbf{S}_{g_1}\mathbf{S}_{g_2}], \tag{12}$$

$$\sum_{g_1 g_2} \mathbf{S}_{g_1}\Gamma_{g_1 g_2}(E)\mathbf{S}^*_{g_2}. \tag{12'}$$

The components D and Γ are the following:

$$D^{(3)}_{gg_2}(x) = \sum_{f_1 f_2} \frac{\Lambda(f_1 g_1)\Lambda(g_1 f_2)\Lambda^{LS}(f_2 g_2)\Lambda^{\mathrm{III}}_x(g_2 f_1)}{\Delta_1(f_1 g_1)\Delta_2(g_1 f_2)\Delta_3(f_2 g_2)},$$

$$\Gamma^{(3)}_{xx} = \sum_{f_1 f_2} \frac{\Lambda(f_1 g_2)\Lambda^{LS}_x(g_1 g_2)\Lambda^{\mathrm{III}}(f_2 g_2)\Lambda^{LS}_x(g_2 f_1)}{\Delta_1(f_1 g_1)\Delta_2(g_1 f_2)\Delta_3(f_2 g_2)}. \tag{13}$$

From (13) we conclude that, owing to the nonorthogonality of the wave functions arising from deformation of an orbit in an external field, a spin dependence appears in an electric field. In work ([11]), on the basis of purely inductive considerations, an expression was written down that qualitatively resembles (12′). But in work ([11]) the existence of an internal inhomogeneous crystallographic electric field was also assumed.

An estimate of terms of the type (12′) gives a value underestimated by two orders of magnitude in comparison with experiment. Therefore the author of work ([12]), as the micromechanism, adopts the mechanism proposed in work ([13]). In work ([13]) the magnetoelectric effect is explained not by spin-orbit interaction but by the dependence of the exchange integral on the electric field. This conclusion of work ([12]) was based on expression (5) of work ([11]). But, as was indicated above, expression (5) of work ([11]) can be compared only with expression (12′), which is associated with the second order in the spin-orbit interaction. In our treatment, in addition to those indicated, there is also a spin-dependent part whose coefficients contain the first order of the spin-orbit interaction, the exchange integral, and overlap integrals.

As is clear from the foregoing, there is no need for the artificial assumption ([13]) of a dependence of the exchange integral on the electric field in order to explain magnetoelectric effects. The many-electron theory of magnetoelectric phenomena automatically leads to three mechanisms: the exchange mechanism ([11]), the band mechanism, and a higher-order band mechanism in the spin-orbit interaction.

The results obtained are a consequence of the consistent application of perturbation theory to the many-electron model.

In conclusion, we note that this same scheme was applied to explaining the micromechanism of piezomagnetic ([1]) and other phenomena related to them. Preliminary calculations show that this effect appears in a higher, 8th, approximation. In this case we expand in a series, with respect to the small displacement of the lattice, the coefficients depending on the displacement F from formula (1) and U from formula (2), and continue the application of the perturbation theory set forth, now to a new Hamiltonian in which acoustic phonon operators also participate. Apparently, optical phonons will also play a role here, since the expansion is carried out in small oscillations relative to magnetic and nonmagnetic sites.

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* It follows immediately from this that, in the antiferromagnetic case, not only the spin frequency but also the nuclear-resonance frequency will depend linearly on the electric field as a result of the presence of additional terms (12) in the Hamiltonian. Physically all this is connected with the overlap of electronic orbits and their deformation by the electric field.

It is interesting to note that, in addition to the terms responsible for piezomagnetism, two biquadratic indirect-exchange expressions of the type $(S_{g_1} S_{g_2})^2$ appear without the participation of spin-orbital interaction. One of them—indirect exchange through phonons—was obtained earlier in work ([14]) from purely thermodynamic considerations. Our treatment shows that the microscopic source of the exchange anisotropy ([14]) is indirect exchange with the participation of phonons. The second biquadratic indirect exchange is purely electronic (through nonmagnetic ions, as in the usual case). But it appears in a higher (8th) approximation. Apparently, this latter biquadratic indirect-exchange interaction is the microscopic source of recently discovered experimental facts ([15,16]).

Let us emphasize once again that in the present work we were interested only in obtaining different types of indirect-exchange Hamiltonians.

To obtain the free energy it is necessary to construct a statistical theory. This makes it possible to obtain various temperature-dependent coefficients of the magnetoelectric and piezomagnetic parts of the free energy ([1]).

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CITED LITERATURE

[1] *Itogi nauki, Antiferromagnetism and Ferrites*, Publ. House of the USSR Academy of Sciences, 1963.
[2] I. E. Dzyaloshinskii, ZhETF, 33, 807 (1957).
[3] T. Moriya, Phys. Rev., 120, 91 (1960).
[4] P. W. Anderson, Phys. Rev., 115, 2 (1959).
[5] N. N. Bogolyubov, *Lectures on Quantum Statistics*, Kiev, 1949.
[6] S. V. Vonsovskii, Yu. M. Seidov, DAN, 107, 37 (1956); O. M. Seidov, Dissertation, Azerbaijan State University, Baku, 1956.
[7] Collected translations, *Theory of Ferromagnetism of Metals and Alloys*, 1963.
[8] V. L. Bonch-Bruevich, S. V. Tyablikov, *The Green Function Method in Statistical Mechanics*, Moscow, 1961.
[9] L. N. Zubarev, UFN, 6, 71, 71 (1960).
[10] Yu. M. Seidov, N. G. Guseinov, Izv. AN AzerbSSR, ser. phys.-math. and techn. sciences, No. 5 (1963).
[11] G. T. Rado, Phys. Rev., 6, 609 (1961).
[12] Dn. Astrov, Abstract of dissertation, 1963.
[13] M. Data, J. Kanamori, M. Tachiki, J. Phys. Soc. Japan, 16, 2589 (1961).
[14] C. Kittel, Phys. Rev., 120 (1960).
[15] E. A. Harris, J. Owen, Phys. Rev. Lett., 11, 9 (1963).
[16] D. S. Rodell, I. S. Jacobs et al., Phys. Rev. Lett., 11, 10 (1963).

Note: Figure translations are in progress. See original paper for figures.