
MAGNETISM
AND FERROELECTRICITY

Exchange Dependence of the Spin-Hamiltonian Constants for Antiferromagnetically Coupled S -Ion Pairs

A. M. Vorotynov, G. A. Petrakovskii, and V. V. Rudenko*

Kirensky Institute of Physics, Siberian Branch of the Russian Academy of Sciences, Akademgorodok 50/38, Krasnoyarsk, 660036 Russia

* e-mail: rvv@iph.krasn.ru

Received November 12, 2009

Abstract—Experimental data that indicate the existence of an exchange-dependent contribution to single-ion spin-Hamiltonian constants for antiferromagnetically coupled S -ion pairs in diamagnetic crystals are reported and generalized.

DOI: 10.1134/S1063783410070164

Earlier, investigation of the effect of a crystal field on magnetic anisotropy of a magnetically concentrated crystal was usually reduced to the choice of an isostructural diamagnetic analog with close lattice parameters (or a series of analogs) [1], which was doped with the corresponding impurity in amounts necessary for the observation of the electron paramagnetic resonance (EPR) spectrum of individual ions. The energy levels, which were calculated using a Hamiltonian written in the single-ion approximation and the molecular-field approximation for a magnetically concentrated crystal with the constants derived from experimental data available in the literature on ion pairs

$MBO_3 + Fe^{3+}$ ($M = Ga, In, Lu, Sc$), $CaO + Mn^{2+}$, $MgO + Mn^{2+}$, and $CsCdCl_3 + Mn^{2+}$ crystals) for the axial spin-Hamiltonian constant D_S [6–9] in order to demonstrate the existence of exchange-dependent contributions.

In Hamiltonians [6–9], the strong isotropic exchange interaction leads to six states characterized by the total spin $S = s_i + s_j$. The energies of these states have the form [6–9]

$$E_S = (J/2)[S(S+1) - s_i(s_i+1) - s_j(s_j+1)].$$

Here, S is the total spin number, which for each multiplet of the pair takes one of the values $S = s_i + s_j, s_i + s_j - 1, \dots, s_i - s_j; s_i = s_j = 5/2; s_i$ and s_j are the spin numbers of ions in the pair; and J is the exchange parameter.

According to [7, 9], the Hamiltonian of the strong exchange interaction for each individual multiplet can be represented in terms of the total spin

$$\mathcal{H} = g\beta HS_z + (D_S/3)O_2^0(S_z).$$

Here, the external field is directed along the axial axis. The fourth-order anisotropic terms are ignored. The spin-Hamiltonian constant D_S has the form [6–9]

$$D_S = 3\alpha_S D_{\text{dip}} + \beta_S D_{cS};$$

$\alpha_S = (1/2)[S(S+1) + 4s_i(s_i+1)]/(2S-1)(2S+3)$ and $\beta_S = [3S(S+1) - 3 - 4s_i(s_i+1)]/(2S-1)(2S+3)$ are the nonlinear functions of the total spin S , which are given in [6–9] and tabulated in [7]; D_{dip} is the dipole interaction constant; and D_{cS} is the constant including the contributions from the crystal field D_{cS} , the local

Constants of the spin Hamiltonian of exchange-coupled S -ion pairs in diamagnetic crystals

Compound	A , Oe	$D_{c0} + D_{cf}$, Oe	D_{dip} , Oe	D'_{dip} , Oe
CaO + Mn ²⁺	35.3	-183.8	-494.4	-498.9
CsCdCl ₃ + Mn ²⁺	-89.7	557.2	-402.1	-407.9
MgO + Mn ²⁺	-15.7	334.3	-567.9	-565.6
GaBO ₃ + Fe ³⁺	-100	1194	-71	-82
InBO ₃ + Fe ³⁺	-55	642	-70	-69
ScBO ₃ + Fe ³⁺	-39	458	-68	-72.6
LuBO ₃ + Fe ³⁺	-8	-9	-78	-77

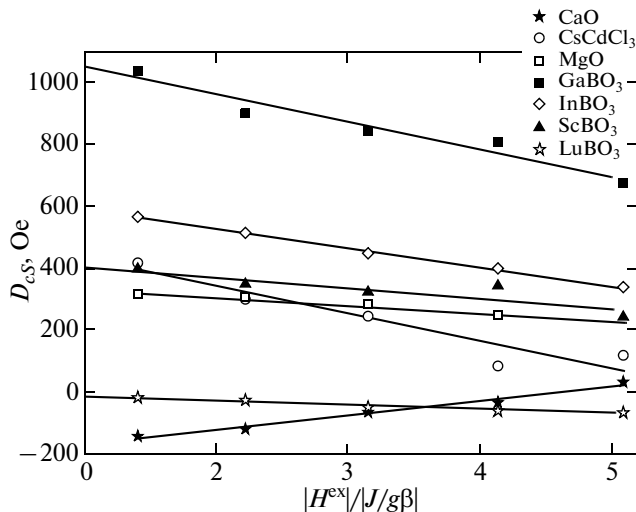
distortion due to the "foreign" pair D_{c0} , and the single-ion exchange mechanism $A\sqrt{S(S+1)}$. The experimental values of D_{cS} can be determined from the expression [6–8, 10]

$$D_{cS} = (D_S/\beta_S) - (3\alpha_S D'_{dip}/\beta_S). \quad (1)$$

The experimental dependence is described using the fitted function

$$D_{cS}(S) = (D_{c0} + D_{cf}) + A\sqrt{S(S+1)}.$$

The expression for the single-ion exchange constant $A\sqrt{S(S+1)}$ is written in this form, because the anisotropic exchange interaction always includes the isotropic exchange parameter. Each multiplet with the total



Dependences of the single-ion spin-Hamiltonian constants D_{cS} for antiferromagnetically coupled Mn²⁺–Mn²⁺ and Fe³⁺–Fe³⁺ ions in CaO, CsCdCl₃, MgO, and MBO_3 ($M = \text{Ga, In, Sc, Lu}$) crystals on the quantity $|H^{ex}|/|J/g\beta|$.

spin S is in its own isotropic exchange field that appears in the pair interaction:

$$\begin{aligned} |\mathbf{H}^{ex}| &= (1/g\beta)(\partial|E_S|/\partial|S|) \\ &= |J/g\beta|\sqrt{S(S+1)} \approx |J/g\beta|(S+1/2). \end{aligned}$$

Equation (1) is also in agreement with the data reported in [4], where the Hamiltonian term describing the single-ion exchange mechanism is obtained in the third order of perturbation theory, which, in particular, is linear in the isotropic exchange interaction. The experimental results for the spin-Hamiltonian constants D_S in $MBO_3 + Fe^{3+}$ ($M = \text{Ga, In, Lu, Sc}$), CaO + Mn²⁺, and MgO + Mn²⁺ crystals are presented in the form of dependences on the total spin S in [6]. These dependences were previously fitted to determine the spin-Hamiltonian parameters D_{dip} , $D_{c0} + D_{cf}$, and A , which have been published only partially [6] (numerical experimental values of the spin-Hamiltonian constant D_S for the CaO + Mn²⁺ and MgO + Mn²⁺ crystals are given in [7]). The parameters D_{dip} , $D_{c0} + D_{cf}$, and A calculated from the dependences $D_S(S)$ for the CaO + Mn²⁺, MgO + Mn²⁺, and CsCdCl₃ + Mn²⁺ crystals are presented for the first time (see table). The experimental data on the spin-Hamiltonian constant D_S for the CsCdCl₃ + Mn²⁺ crystal are reported in [10].

The experimental value of the dipole interaction constant $D'_{dip} = \{D_3 - \beta_3[D_{cf} + D_{c0} + A\sqrt{12}]\}/3\alpha_3$ can be determined with a higher accuracy than D_{dip} from the data obtained for the multiplet with $S = 3$. The constants D_3 , $(D_{c0} + D_{cf})$, and A , which are necessary for this determination, can be taken from data on the dependence $D_S(S)$ [6, 7, 10]. This approach to the determination of the dipole interaction constant D'_{dip} made it possible to construct the monotonic dependences $D_{cS}(|H^{ex}|)$ dependences (see figure). Note that, when constructing the experimental dependence described by Eq. (1), the point for $S = 3$ is especially sensitive to the error in the determination of the constant D_3 and the dipole interaction constant because the quantity β_3 is small.

Therefore, the single-ion exchange constant includes the correlation correction $\sqrt{S(S+1)}$, which must be taken into account in writing the Hamiltonian for pairs in terms of the total spin S . As can be seen from the figure, this notation should have a general form.

REFERENCES

1. L. Rimai and T. Kushida, Phys. Rev. **143** (1), 160 (1966).

2. S. Geschwind, Phys. Rev. **121** (2), 363 (1961).
3. G. P. Rodrigue, H. Meyer, and R. V. Jones, J. Appl. Phys. **31** (5), S376 (1960).
4. A. E. Nikiforov, V. Ya. Mitrofanov, and A. N. Men, Phys. Status Solidi B **45** (1), 65 (1971).
5. A. S. Moskvina, I. G. Bostrem, and M. A. Sidorov, Zh. Éksp. Teor. Fiz. **104** (1), 2499 (1993) [JETP **77** (1), 127 (1993)].
6. V. V. Men'shikov, S. G. Ovchinnikov, V. V. Rudenko, A. N. Sudakov, V. I. Tugarinov, and A. M. Vorotynov, J. Magn. Magn. Mater. **267** (3), 289 (2003).
7. E. A. Harris, J. Phys. C: Solid State Phys. **5** (3), 338 (1972).
8. A. M. Vorotynov, S. G. Ovchinnikov, V. V. Rudenko, and A. N. Sudakov, Fiz. Tverd. Tela (St. Petersburg) **42** (7), 1275 (2000) [Phys. Solid State **42** (7), 1313 (2000)].
9. J. Owen, J. Appl. Phys. **32** (3), S213 (1961).
10. S. N. Martynov and S. B. Petrov, Phys. Status Solidi B **149** (1), K41 (1988).

Translated by G. Tsydynzhapov