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ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

Weak Ferromagnetism along the Triad Axis and the Basal Anisotropy Caused by the Dzyaloshinskii–Moriya Interaction and the Cubic Electric Field of the $FeBO_3$ Crystal

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Abstract—Based on the spin Hamiltonian and taking into account the cubic invariant of the crystal field and the Dzyaloshinskii–Moriya interaction, the weak ferromagnetic moment along the triad axis and the basal anisotropy of $FeBO_3$ crystals are calculated in the approximation of second-order perturbation theory.

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1. INTRODUCTION

Although iron borate crystals have been synthesized and investigated quite thoroughly for a long time, they continue to attract the interest of researchers, since they are convenient objects for constructing various models related to magnetism [1]. These crystals are characterized by a relatively simple lattice, a high Néel temperature, narrow lines of antiferromagnetic resonance [2], and a number of isostructural diamagnetic analogs. Thus, Dmitrienko et al. [1] were the first to determine in 2014 the magnitude and sign of vector components in the Dzyaloshinskii–Moriya interaction (for iron borate crystals) [1]. Note that we confirm and use the work by Dmitrienko et al. [1] in our study, which follows from the calculated data and experiment on hexagonal anisotropy (see Section 4). In this work, we take into account the sign of the basal hexagonal anisotropy of $FeBO_3$ and calculate the weak ferromagnetic moment along the triad axis of $FeBO_3$ crystals due to the influence of the cubic electric field and the Dzyaloshinskii–Moriya interaction. The calculations were performed in the approximation of second-order perturbation theory. The calculated matrix elements used in Section 3, are presented in the Appendix. Note that the free energy was considered earlier in [3], but in a less correct form, although it gave the same result. For the exchange interaction term, the molecular field approximation was used. The quantitative evaluation of the obtained expressions for the basal hexagonal anisotropy and the weak ferromagnetic moment along the triad axis was made using electron paramagnetic resonance (EPR) data on $MBO_3 + Fe^{3+}$ crystals isostructural to iron borate (M = Ga, In, Sc, Lu) (see Section 4). The main results of this work are presented in the Section 5.

2. PHENOMENOLOGICAL DESCRIPTION OF ENERGY OF HEXAGONAL ANISOTROPY AND WEAK FERROMAGNETIC MOMENT ALONG TRIAD AXIS OF $FeBO_3$ CRYSTALS

Magnetic properties of iron borate crystals are described by the free energy [4]

$$\Phi = M\left[\frac{1}{2}B\mathbf{M}^2 + \frac{1}{2}a\cos^2\theta + d_{DM}(L_xM_y - L_yM_x) + q\sin^3\theta\cos\theta\cos3\varphi + tM_z\sin^3\theta\sin3\varphi\right]; \quad (1)$$

$$\mathbf{M} = \frac{\mathbf{M}_1 + \mathbf{M}_2}{M}, \quad \mathbf{L} = \frac{\mathbf{M}_1 - \mathbf{M}_2}{M},$$
$$M = 2|\mathbf{M}_1| = 2|\mathbf{M}_2| = Ng\beta sB_{5/2}(x),$$

$\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations, N is the Avogadro number, g is the spectroscopic splitting factor, β is the Bohr magneton, S is the spin of the iron ion equal to 5/2, and $B_{5/2}(x)$ is the Brillouin function. All constants in expression (1) have a dimension of magnetic field. Despite the relatively simple crystal structure of $FeBO_3$ (calcite), the behavior of the magnetic system during the rotation of the antiferromagnetism vector $\mathbf{L}$ in the (111) plane relative to the last two terms in (1) is rather complex [3, 4]. Such a complex behavior (which can be seen in Fig. 1, which shows the distribution of cubic crystal axes), obtained from EPR spectra in $ScBO_3 + Fe^{3+}$ [5] and $CaCO_3 + Mn^{2+}$ [6] crystals isostructured to iron borates is char-

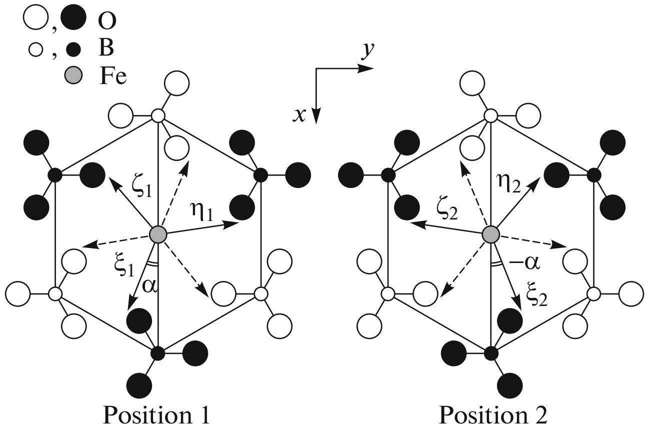

Fig. 1. Effective positions of BO_3^{3-} ions and cubic electric field axes for two inequivalent positions of the ion M in the MBO_3 lattice (M = Fe, Ga, In, Lu, Sc) [5].

acteristic of the effective basal anisotropy and vector of ferromagnetism **M** along the triad axis of the $FeBO_3$ crystal. The first term in Eq. (1) characterizes the isotropic exchange energy in the crystal; the second term describes the uniaxial anisotropy; the third term is for the Dzyaloshinskii interaction, which leads to the appearance of weak ferromagnetism in the basal plane (111); the last two terms are the anisotropy energy in the (111) plane; θ and φ are the polar and azimuthal angles of vector **L**, measured from the triad axis (z) and from the crystal symmetry plane (x axis), respectively (see Fig. 1). Phenomenological expressions for the effective basal anisotropy and the weak ferromagnetic moment along the triad axis are obtained by minimizing the free energy (1) along θ and M_z and have the form [3, 7]

$$E_q\sin^6\theta\cos6\varphi = -\frac{(qM)^2}{4M(a+d_{DM}^2/B)}\sin^6\theta\cos6\varphi,$$
$$M_z = -\frac{tM}{B}\sin^3\theta\sin3\varphi. \quad (2)$$

3. CALCULATION OF HEXAGONAL ANISOTROPY AND WEAK FERROMAGNETIC MOMENT ALONG THE TRIAD AXIS OF $FeBO_3$ CRYSTAL BASED ON SPIN HAMILTONIAN TAKING INTO ACCOUNT THE INFLUENCE OF CUBIC ELECTRIC FIELD AND DZYALOSHINSKII–MORIYA INTERACTION

The spin Hamiltonian taking into account two nonequivalent positions of Fe^{3+} ions and Dzyaloshinskii–Moriya interaction ions takes the form [3, 8]

$$\hat{H} = g\beta\mathbf{H}_j^{\text{eff}}\mathbf{s}_j + \frac{1}{3}D_{cf}O_{2j}^0 + \frac{F_{cf}}{180}O_{4j}^0 - \frac{a_{cf}}{180}[O_{4j}^0 - 20\sqrt{2}(O_{4j}^3\cos3\alpha_{cfj} - \tilde{O}_{4j}^3\sin3\alpha_{cfj})] + d_{DM}(s_{x1}s_{y2} - s_{y1}s_{x2}). \quad (3)$$

Here, the first term (exchange interaction energy) in Eq. (3) is written in the molecular field approximation, **s** is the ion spin operator, O_n^m are the equivalent spin operators that are given together with their matrix elements, for example, in [9, 10], and α_{cfj} is the angle between the projections of the axis of the cubic crystal field on the (111) plane and the plane of symmetry of the crystal at position j (Fig. 1; for a more detailed description, see [7, 8]). The second, third, and fourth terms (for the Hamiltonian constant a_{cf}) describe the interactions of axial and cubic symmetry. The last term describes the Dzyaloshinskii–Moriya interaction.

The exchange term in (3) can be written in the zeroth approximation in perturbation theory assuming that the quantization axis is determined by angles θ_j and φ_j measured from the triad axis and from the symmetry plane, respectively. The Hamiltonian in the rotating coordinate system can be written as in [10] with the expressions responsible for the Dzyaloshinskii–Moriya interaction [11]:

$$\hat{H} = g\beta H_j^{\text{eff}}s_{zj} + \sum_{m=0}^{2}a_{2j}^mO_{2j}^m + \sum_{m=0}^{4}a_{4j}^mO_{4j}^m + \sum_{m=1}^{4}\tilde{a}_{4j}^m\tilde{O}_{4j}^m + d_{DM}[(s_{x1}s_{x2}\cos\theta_1\cos\theta_2 + s_{y1}s_{y2} + s_{z1}s_{z2}\sin\theta_1\sin\theta_2)\sin(\varphi_2-\varphi_1) + (s_{x1}s_{z2}\cos\theta_1\sin\theta_2 + s_{z1}s_{x2}\sin\theta_1\cos\theta_2)\sin(\varphi_2-\varphi_1) + (s_{x1}s_{y2}\cos\theta_1 - s_{y1}s_{x2}\cos\theta_2)\cos(\varphi_2-\varphi_1) + (s_{z1}s_{y2}\sin\theta_1 - s_{y1}s_{z2}\sin\theta_2)\cos(\varphi_2-\varphi_1)]. \quad (4)$$

Here, the signs of the rotating coordinate system are omitted for simplicity, and $\varphi_2 - \varphi_1 \approx 180°$ is the difference between the orientations of the sublattices $j = 1$ and $j = 2$. We can take into account the weak ferromagnetic moment along the triad axis and the basal anisotropy in the second approximation of perturbation theory using expressions for $\tilde{a}_{4j}^1\tilde{O}_{4j}^1$, where

$$\tilde{a}_{4j}^1 = -a_{cf}(\sqrt{2}/12)\sin^2\theta_j\cos\theta_j\sin3(\varphi_j+\alpha_j)$$

plus the last term in (4):

$$|d_{DM}|(s_{z1}s_{y2}\sin\theta_1 - s_{y1}s_{z2}\sin\theta_2);$$

in formulas (1)–(4), d_{DM} value for $FeBO_3$ is negative [1].

The expression for the energy in the approximation of second-order perturbation theory was given, for example, in [12]:

$$W'' = \sum_{m_1',m_2'} \frac{\langle m_1, m_2|\hat{H}''|m_1', m_2'\rangle\langle m_1', m_2'|\hat{H}''|m_1, m_2\rangle}{W^0_{m_1,m_2} - W^0_{m_1',m_2'}},$$

where $\hat{H}''$ is the perturbation operator, $|m_1, m_2\rangle$ and $|m_1', m_2'\rangle$ are the wavefunctions of ions 1 and 2, respectively, in the ground and excited states; m_1, m_2 and m_1', m_2' have the meaning of the corresponding magnetic quantum numbers; and the denominator is the difference between the energy of the ground and excited states in the zero approximation. The calculated matrix elements, in which the constants $\tilde{a}^1_{4j}$ and the trigonometric functions in the last term of Eq. (4) were omitted for simplicity, are presented in the Appendix. The energy levels in the zero and second approximations of the perturbation theory in relation to the above expressions are

$$\begin{aligned} W_{j,\pm 1/2} &= \pm\frac{1}{2}g\beta H_j^{\text{eff}} - s\sqrt{2}\frac{|d_{DM}|a_{cf}}{g\beta H_j^{\text{eff}}} \\ &\times \sin^3\theta_j\cos\theta_j\sin 3(\varphi_j + \alpha_j), \\ W_{j,\pm 3/2} &= \pm\frac{3}{2}g\beta H_j^{\text{eff}} + 9s\sqrt{2}\frac{|d_{DM}|a_{cf}}{2g\beta H_j^{\text{eff}}} \\ &\times \sin^3\theta_j\cos\theta_j\sin 3(\varphi_j + \alpha_j), \\ W_{j,\pm 5/2} &= \pm\frac{5}{2}g\beta H_j^{\text{eff}} - s^2\sqrt{2}\frac{|d_{DM}|a_{cf}}{g\beta H_j^{\text{eff}}} \\ &\times \sin^3\theta_j\cos\theta_j\sin 3(\varphi_j + \alpha_j). \end{aligned} \tag{5}$$

Here, without loss of generality with the final result (in the approximation of a strong exchange field), we assumed $\sin\theta_1 = \sin\theta_2 = \sin\theta_j$ (see the last term in (4)). In expression (5) $|d_{DM}| = g\beta|H_{DM}/m|$, where $|d_{DM}|$ and $|m|$ are the absolute values of the Dzyaloshinskii–Moriya field and the magnetic quantum number, respectively. The final expressions for the energy levels take the form

$$\begin{aligned} W_{j,\pm 1/2} &= \pm\frac{1}{2}g\beta H_j^{\text{eff}} - 2s\sqrt{2}a_{cf}\frac{|H_{DM}|}{H_j^{\text{eff}}} \\ &\times \sin^3\theta_j\cos\theta_j\sin 3(\varphi_j + \alpha_j), \\ W_{j,\pm 3/2} &= \pm\frac{3}{2}g\beta H_j^{\text{eff}} + 3s\sqrt{2}a_{cf}\frac{|H_{DM}|}{H_j^{\text{eff}}} \\ &\times \sin^3\theta_j\cos\theta_j\sin 3(\varphi_j + \alpha_j), \\ W_{j,\pm 5/2} &= \pm\frac{5}{2}g\beta H_j^{\text{eff}} - s\sqrt{2}a_{cf}\frac{|H_{DM}|}{H_j^{\text{eff}}} \\ &\times \sin^3\theta_j\cos\theta_j\sin 3(\varphi_j + \alpha_j). \end{aligned} \tag{6}$$

At arbitrary temperatures, the contribution to the weak ferromagnetic moment along the triad axis and the basal anisotropy constant will be determined from the free energy of the crystal

$$F = -\frac{NkT}{2}\sum_j \ln Z_j, \quad Z_j = \sum_{m_j}\exp\left(-\frac{W_{jm_j}}{kT}\right)$$

is the sum of the states of the jth ion.

Let us expand the exponential function in the expression for the free energy of a crystal in a series in $(c(m_j)a_j(\theta_j, \varphi_j, \alpha_j))/kT$. Restricting consideration to the linear expansion term, we get

$$\begin{aligned} F &= -\frac{NkT}{2}\sum_j \ln\sum_{m_j}\exp\left(-\frac{g\beta H_j^{\text{eff}}m_j}{kT}\right) \\ &\times\left(1 - \frac{c_j(m_j)a_j(\theta_j,\varphi_j,\alpha_j)}{kT}\right), \end{aligned} \tag{7}$$

$$a_j(\theta_j,\varphi_j,\alpha_j) = s\sqrt{2}a_{cf}\frac{|H_{DM}|}{H_j^{\text{eff}}}\sin^3\theta_j\cos\theta_j\sin 3(\varphi_j + \alpha_j),$$

$c_j(m_j)$ have values derived from Eqs. (6) and (7). Using the notation $Y_j = \exp(-g\beta H_j^{\text{eff}}/kT)$ and summing over m_j, we rewrite the expression for F as follows

$$\begin{aligned} F &= -\frac{NkT}{2}\sum_j \ln\left\{(Y_j^{1/2} + Y_j^{-1/2})\left(1 - \frac{2a_j}{kT}\right)\right. \\ &\left. + (Y_j^{3/2} + Y_j^{-3/2})\left(\frac{1 + 3a_j}{kT}\right) + (Y_j^{5/2} + Y_j^{-5/2})\left(\frac{1 - a_j}{kT}\right)\right\} \\ &= -\frac{NkT}{2}\sum_j \ln\left(z_{0j} - \frac{a_j}{kT}a_{2j}\right) \\ &= -\frac{NkT}{2}\sum_j\left\{\ln z_{0j} + \ln\left(1 - \frac{a_j}{kT}\frac{z_{2j}}{z_{0j}}\right)\right\}, \end{aligned}$$

where

$$z_{0j} = \frac{Y_j^5 + Y_j^4 + Y_j^3 + Y_j^2 + Y_j + 1}{Y_j^{5/2}},$$

$$z_{2j} = \frac{-Y_j^5 + 3Y_j^4 - 2Y_j^3 - 2Y_j^2 + 3Y_j - 1}{Y_j^{5/2}}.$$

Decomposing the function $\ln[1 - (a_j/kT)(z_{2j}/z_{0j})]$ in a series in a small parameter $(a_j/kT)(z_{2j}/z_{0j})$ and, limiting ourselves to considering only the linear term, we get

$$F = \frac{N}{2}\sum_j\left(a_j\frac{z_{2j}}{z_{0j}}\right). \tag{8}$$

By decomposing the sinusoidal functions a_j in Eqs. (6) and (7), introducing the azimuthal angles $\varphi + \alpha$ and $\varphi + \pi - \alpha$ for the vector of antiferromagnetism similarly to [13] and summing over j, we arrive at the expression

$$F = \frac{N}{2}\sqrt{2}a_{cf}\frac{|H_{DM}|}{H^{\text{eff}}}r(Y)\sin^3\theta[(\cos\theta_1 - \cos\theta_2) \\ \times\sin 3\alpha\cos 3\varphi + (\cos\theta_1 + \cos\theta_2)\cos 3\alpha\sin 3\varphi]. \quad (9)$$

In (9), by definition, $\cos\theta_1 - \cos\theta_2 = 2\cos\theta$, $\cos\theta_1 + \cos\theta_2 = 2m_z$. Let us write the expression for the constants of hexagonal anisotropy, which follows from Eqs. (1) and (9) in accordance with the single-ion contribution [4]:

$$q_{cfDM} = N\sqrt{2}a_{cf}r(Y)\left\{\frac{|H_{DM}|}{H^{\text{eff}}}\sin 3\alpha - \frac{1}{3}\cos 3\alpha\right\}, \\ r(Y) = \frac{5}{2}\frac{z_2}{z_0} = \frac{5}{2}\frac{-Y^5 + 3Y^4 - 2Y^3 - 2Y^2 + 3Y - 1}{Y^5 + Y^4 + Y^3 + Y^2 + Y + 1}. \quad (10)$$

This function was introduced by Wolf when calculating the single-ion magnetic anisotropy of cubic crystals [14]. In Eq. (10), a_{cf} is expressed in the energy units.

The effective field of the measured hexagonal anisotropy, which follows from (2), (9), and (10), has the form

$$H_{qcfDM}\sin^6\theta\cos 6\varphi = -\frac{a_{cf}^2[r(Y)/s]^2\{[|H_{DM}|/H^{\text{eff}}]\sin 3\alpha - (1/3)\cos 3\alpha\}^2}{2\{H_A(0) + [H_{DM}^2(0)]/H_E(0)\}B_{5/2}^3(x)}\sin^6\theta\cos 6\varphi. \quad (11)$$

Note that, in Eq. (11), the ratio between the effective fields H_{DM}/H^{eff} multiplied by $M = Ng\beta B_{5/2}(x)$ is equal to the weak ferromagnetic moment [4]. Equating the energy m_z in (1) to (9), we obtain

$$t = N\frac{\sqrt{2}a_{cf}|H_{DM}|}{H^{\text{eff}}}r(Y)\cos 3\alpha,$$

and then define according to (2)

$$m_z\sin^3\theta\sin 3\varphi \\ = -N\frac{\sqrt{2}a_{cf}|H_{DM}|}{BH^{\text{eff}}}r(Y)\cos 3\alpha\sin^3\theta\sin 3\varphi.$$

Based on this expression, we find the measured magnetic moment along the triad axis per a mole of the crystalline substance $FeBO_3$:

$$\sigma_z(T) = m_z M = -\sqrt{2}Ng\beta sB_{5/2}(x)\frac{a_{cf}|H_{DM}|}{2(H^{\text{eff}})^2} \\ \times\frac{r(Y)}{sB_{5/2}(x)}\cos 3\alpha\sin^3\theta\sin 3\varphi. \quad (12)$$

Here a_{cf} is expressed in units of field (oersteds).

4. QUANTITATIVE ESTIMATION OF THE BASAL ANISOTROPY AND WEAK FERROMAGNETIC MOMENT ALONG THE TRIAD AXIS OF $FeBO_3$ CRYSTALS RESULTING FROM A CUBIC ELECTRIC FIELD AND DZYALOSHINSKII–MORIYA INTERACTION

Theoretical estimates of the values of anisotropic interactions in $FeBO_3$ (based on the single-ion model) were made using experimentally determined (EPR) spin Hamiltonian constants for $MBO_3 + Fe^{3+}$ crystals isostructural to iron borate (M = Ga, In, Sc, Lu).

Estimation using Eq. (12) (taking into account the cubic crystal field and Dzyaloshinskii–Moriya interaction in the second approximation of perturbation theory) gives $\sigma_z \sim 1 \times 10^{-4}$ G cm^3/g (at $T = 0$ K), which is an order of magnitude smaller than the single-ion contribution [13] obtained in the first approximation of the perturbation theory (2.4×10^{-3} G cm^3/g). The experimental value at $T = 77$ K (based on magnetization measurements) is 1.3×10^{-3} G cm^3/g [15]. To illustrate the levels of “strong” and finer interactions, we also give the value of the weak ferromagnetic Dzyaloshinskii–Moriya moment in the basal plane at $T = 77$ K ($\sigma_{xy} = 3$ G cm^3/g) [16].

A quantitative estimate of the hexagonal anisotropy caused by Fe^{3+} ions in $FeBO_3$ with respect to two mechanisms (11) gives $H_{qcfDM}(0) = -1.0 \times 10^{-2}$ Oe (according to EPR) and the experimental value $H_q(0) = -1.1 \times 10^{-2}$ Oe (according to antiferromagnetic resonance) [8, 17]. In Eq. (11), H_D, a_{cf}, and α_{cf} are 100 kOe [2], 130 Oe [5], and 24° [5], respectively, which corresponds to a $FeBO_3$ crystal with the lattice parameters from [7]; $H_E(0) = 2H^{\text{eff}}(0) = 6020$ kOe [2, 18]; $B_{5/2}(x)$ is the Brillouin function for spin $S = 5/2$.

5. CONCLUSIONS

The influence of the cubic crystal field and the Dzyaloshinskii–Moriya interaction on the magnitude of the basal hexagonal anisotropy and weak ferromagnetic moment along the triad axis are considered. The calculated value of H_{qcfDM} appears only in the second order of the perturbation theory and is consistent with EPR data. In future, to determine the presence of other contributions (in particular, “single-ion exchange”) [7] and comparison with experiment, it is desirable to carry out measurements using a highly sensitive magnetometer.

The correct estimation of the basal anisotropy (taking into account the sign of the Dzyaloshinskii–Moriya vector and the single-ion contribution) did not significantly change the final result. The calculated contribution to the weakly ferromagnetic moment along the triad axis taking into account the influence of the cubic electric field and the Dzyaloshinskii–Moriya interaction is an order of magnitude smaller than the experimental value for a single ion; therefore, the main contribution to the weakly ferromagnetic moment along the triad axis comes from the single-ion mechanism [13]. For a better agreement between the calculations and the experiment, it will be necessary to consider additional mechanisms of anisotropic interactions. In conclusion, we note that Section 4 presents comprehensive experimental data on the studied problem.

APPENDIX

Calculation of matrix elements:

$$s = 5/2, \quad m_1 = -1/2, \quad j = 1$$

$$\langle m_1 = -1/2, m_2 = 1/2|\tilde{O}_{4j}^1/(g\beta H^{\text{eff}})|m_1' = -3/2, m_2' = 1/2\rangle\langle m_1' = -3/2, m_2' = 1/2|(-s_{z2}s_{y1})|m_1 = -1/2, m_2 = 1/2\rangle = 15/(g\beta H^{\text{eff}}),$$

$$\langle m_1 = -1/2, m_2 = 1/2|(-s_{z2}s_{y1})|m_1' = -3/2, m_2' = 1/2\rangle \times\langle m_1' = -3/2, m_2' = 1/2|\tilde{O}_{4j}^1/(g\beta H^{\text{eff}})|m_1 = -1/2, m_2 = 1/2\rangle = 15/(g\beta H^{\text{eff}}),$$

$$s = 5/2, \quad m_1 = -1/2, \quad j = 2$$

$$\langle m_1 = -1/2, m_2 = 1/2|\tilde{O}_{4j}^1/(g\beta H^{\text{eff}})|m_1' = -1/2, m_2' = 3/2\rangle\langle m_1' = -1/2, m_2' = 3/2|s_{z1}s_{y2}|m_1 = -1/2, m_2 = 1/2\rangle = 15/(g\beta H^{\text{eff}}),$$

$$\langle m_1 = -1/2, m_2 = 1/2|s_{z1}s_{y2}|m_1' = -1/2, m_2' = 3/2\rangle \times\langle m_1' = -1/2, m_2' = 3/2|\tilde{O}_{4j}^1/(g\beta H^{\text{eff}})|m_1 = -1/2, m_2 = 1/2\rangle = 15/(g\beta H^{\text{eff}}),$$

$$s = 5/2, \quad m_1 = 1/2, \quad j = 1$$

$$\langle m_1 = 1/2, m_2 = -1/2|\tilde{O}_{4j}^1/(-g\beta H^{\text{eff}})|m_1' = 3/2, m_2' = -1/2\rangle\langle m_1' = 3/2, m_2' = -1/2|-s_{z2}s_{y1}|m_1 = 1/2, m_2 = -1/2\rangle = 15/(g\beta H^{\text{eff}}),$$

$$\langle m_1 = 1/2, m_2 = -1/2|-s_{z1}s_{y1}|m_1' = 3/2, m_2' = -1/2\rangle \times\langle m_1' = 3/2, m_2' = -1/2|\tilde{O}_{4j}^1/(-g\beta H^{\text{eff}})|m_1 = 1/2, m_2 = -1/2\rangle = 15/(g\beta H^{\text{eff}}),$$

$$s = 5/2, \quad m_1 = 1/2, \quad j = 2$$

$$\langle m_1 = 1/2, m_2 = -1/2|\tilde{O}_{4j}^1/(-g\beta H^{\text{eff}})|m_1' = 1/2, m_2' = -3/2\rangle\langle m_1' = 1/2, m_2' = -3/2|s_{z1}s_{y2}|m_1 = 1/2, m_2 = -1/2\rangle = 15/(g\beta H^{\text{eff}}),$$

$$\langle m_1 = 1/2, m_2 = -1/2|s_{z1}s_{y2}|m_1' = 1/2, m_2' = -3/2\rangle \times \langle m_1' = 1/2, m_2' = -3/2|\tilde{O}_{4j}^1/(-g\beta H^{\text{eff}})|m_1 = 1/2, m_2 = -1/2\rangle = 15/(g\beta H^{\text{eff}}),$$

$$s = 5/2, \quad m_1 = -3/2, \quad j = 1$$

$$\langle m_1 = -3/2, m_2 = 3/2|\tilde{O}_{4j}^1/(-g\beta H^{\text{eff}})|m_1' = -1/2, m_2' = 3/2\rangle\langle m_1' = -1/2, m_2' = 3/2|-s_{z2}s_{y1}|m_1 = -3/2, m_2 = 3/2\rangle = -45/(g\beta H^{\text{eff}}),$$

$$\langle m_1 = -3/2, m_2 = 3/2|-s_{z2}s_{y1}|m_1' = -1/2, m_2' = 3/2\rangle \langle m_1' = 1/2, m_2' = 3/2|\tilde{O}_{4j}^1/(-g\beta H^{\text{eff}})|m_1 = -3/2, m_2 = 3/2\rangle = -45/(g\beta H^{\text{eff}}),$$

$$s = 5/2, \quad m_1 = -3/2, \quad j = 2$$

$$\langle m_1 = -3/2, m_2 = 3/2|\tilde{O}_{4j}^1/(-g\beta H^{\text{eff}})|m_1' = -3/2, m_2' = 1/2\rangle\langle m_1' = -3/2, m_2' = 1/2|s_{z1}s_{y2}|m_1 = -3/2, m_2 = 3/2\rangle = -45/(g\beta H^{\text{eff}}),$$

$$\langle m_1 = -3/2, m_2 = 3/2|s_{z1}s_{y2}|m_1' = -3/2, m_2' = 1/2\rangle \langle m_1' = -3/2, m_2' = 1/2|\tilde{O}_{4j}^1/(-g\beta H^{\text{eff}})|m_1 = -3/2, m_2 = 3/2\rangle = -45/(g\beta H^{\text{eff}}),$$

$$s = 5/2, \quad m_1 = 3/2, \quad j = 1$$

$$\langle m_1 = 3/2, m_2 = -3/2|\tilde{O}_{4j}^1/(g\beta H^{\text{eff}})|m_1' = 1/2, m_2' = -3/2\rangle\langle m_1' = 1/2, m_2' = -3/2|-s_{z2}s_{y1}|m_1 = 3/2, m_2 = -3/2\rangle = -45/(g\beta H^{\text{eff}}),$$

$$\langle m_1 = 3/2, m_2 = -3/2|-s_{z2}s_{y1}|m_1' = 1/2, m_2' = -3/2\rangle \langle m_1' = 1/2, m_2' = -3/2|\tilde{O}_{4j}^1/(g\beta H^{\text{eff}})|m_1 = 3/2, m_2 = -3/2\rangle = -45/(g\beta H^{\text{eff}}),$$

$$s = 5/2, \quad m_1 = 3/2, \quad j = 2$$

$$\langle m_1 = 3/2, m_2 = -3/2|\tilde{O}_{4j}^1/(g\beta H^{\text{eff}})|m_1' = 3/2, m_2' = -1/2\rangle\langle m_1' = 3/2, m_2' = -1/2|s_{z1}s_{y2}|m_1 = 3/2, m_2 = -3/2\rangle = -45/(g\beta H^{\text{eff}}),$$

$$\langle m_1 = 3/2, m_2 = -3/2|s_{z1}s_{y2}|m_1' = 3/2, m_2' = -1/2\rangle \langle m_1' = 3/2, m_2' = -1/2|\tilde{O}_{4j}^1/(g\beta H^{\text{eff}})|m_1 = 3/2, m_2 = -3/2\rangle = -45/(g\beta H^{\text{eff}}),$$

$$s = 5/2, \quad m_1 = -3/2, \quad j = 1$$

$$\langle m_1 = -3/2, m_2 = 3/2|\tilde{O}^1_{4j}/(g\beta H^{\mathrm{eff}})|m_1' = -5/2, m_2' = 3/2\rangle\langle m_1' = -5/2, m_2' = 3/2|-s_{z2}s_{y1}|m_1 = -3/2, m_2 = 3/2\rangle = -45/(2g\beta H^{\mathrm{eff}}),$$

$$\langle m_1 = -3/2, m_2 = 3/2|-s_{z2}s_{y1}|m_1' = -5/2, m_2' = 3/2\rangle\langle m_1' = -5/2, m_2' = 3/2|\tilde{O}^1_{4j}/(g\beta H^{\mathrm{eff}})|m_1 = -3/2, m_2 = 3/2\rangle = -45/(2g\beta H^{\mathrm{eff}}),$$

$$s = 5/2, \quad m_1 = -3/2, \quad j = 2$$

$$\langle m_1 = -3/2, m_2 = 3/2|\tilde{O}^1_{4j}/(g\beta H^{\mathrm{eff}})|m_1' = -3/2, m_2' = 5/2\rangle\langle m_1' = -3/2, m_2' = 5/2|s_{z1}s_{y2}|m_1 = -3/2, m_2 = 3/2\rangle = -45/(2g\beta H^{\mathrm{eff}}),$$

$$\langle m_1 = -3/2, m_2 = 3/2|s_{z1}s_{y2}|m_1' = -3/2, m_2' = 5/2\rangle\langle m_1' = -3/2, m_2' = 5/2|\tilde{O}^1_{4j}/(g\beta H^{\mathrm{eff}})|m_1 = -3/2, m_2 = 3/2\rangle = -45/(2g\beta H^{\mathrm{eff}}),$$

$$s = 5/2, \quad m_1 = 3/2, \quad j = 1$$

$$\langle m_1 = 3/2, m_2 = -3/2|\tilde{O}^1_{4j}/(-g\beta H^{\mathrm{eff}})|m_1' = 5/2, m_2' = -3/2\rangle\langle m_1' = 5/2, m_2' = -3/2|-s_{z2}s_{y1}|m_1 = 3/2, m_2 = -3/2\rangle = -45/(2g\beta H^{\mathrm{eff}}),$$

$$\langle m_1 = 3/2, m_2 = -3/2|(-s_{z2}s_{y1}|m_1' = 5/2, m_2' = -3/2\rangle\langle m_1' = 5/2, m_2' = -3/2|\tilde{O}^1_{4j}/(-g\beta H^{\mathrm{eff}})|m_1 = 3/2, m_2 = -3/2\rangle = -45/(2g\beta H^{\mathrm{eff}}),$$

$$s = 5/2, \quad m_1 = 3/2, \quad j = 2$$

$$\langle m_1 = 3/2, m_2 = -3/2|\tilde{O}^1_{4j}/(-g\beta H^{\mathrm{eff}})|m_1' = 3/2, m_2' = -5/2\rangle\langle m_1' = 3/2, m_2' = -5/2|s_{z1}s_{y2}|m_1 = 3/2, m_2 = -3/2\rangle = -45/(2g\beta H^{\mathrm{eff}}),$$

$$\langle m_1 = 3/2, m_2 = -3/2|s_{z1}s_{y2}|m_1' = 3/2, m_2' = -5/2\rangle\langle m_1' = 3/2, m_2' = -5/2|\tilde{O}^1_{4j}/(-g\beta H^{\mathrm{eff}})|m_1 = 3/2, m_2 = -3/2\rangle = -45/(2g\beta H^{\mathrm{eff}}),$$

$$s = 5/2, \quad m_1 = -5/2, \quad j = 1$$

$$\langle m_1 = -5/2, m_2 = 5/2|\tilde{O}^1_{4j}/(-g\beta H^{\mathrm{eff}})|m_1' = -3/2, m_2' = 5/2\rangle\langle m_1' = -3/2, m_2' = 5/2|-s_{z2}s_{y1}|m_1 = -5/2, m_2 = 5/2\rangle = 75/(2g\beta H^{\mathrm{eff}}),$$

$$\langle m_1 = -5/2, m_2 = 5/2|-s_{z2}s_{y1}|m_1' = -3/2, m_2' = 5/2\rangle\langle m_1' = -3/2, m_2' = 5/2|\tilde{O}^1_{4j}/(-g\beta H^{\mathrm{eff}})|m_1 = -5/2, m_2 = 5/2\rangle = 75/(2g\beta H^{\mathrm{eff}}),$$

$$s = 5/2, \quad m_1 = -5/2, \quad j = 2$$

$$\langle m_1 = -5/2, m_2 = 5/2|\tilde{O}^1_{4j}/(-g\beta H^{\mathrm{eff}})|m_1' = -5/2, m_2' = 3/2\rangle\langle m_1' = -5/2, m_2' = 3/2|s_{z1}s_{y2}|m_1 = -5/2, m_2 = 5/2\rangle = 75/(2g\beta H^{\mathrm{eff}}),$$

$$\langle m_1 = -5/2, m_2 = 5/2|s_{z1}s_{y2}|m_1' = -5/2, m_2' = 3/2\rangle\langle m_1' = -5/2, m_2' = 3/2|\tilde{O}^1_{4j}/(-g\beta H^{\mathrm{eff}})|m_1 = -5/2, m_2 = 5/2\rangle = 75/(2g\beta H^{\mathrm{eff}}),$$

$$s = 5/2, \quad m_1 = 5/2, \quad j = 1$$

$$\langle m_1 = 5/2, m_2 = -5/2|\tilde{O}^1_{4j}/(g\beta H^{\mathrm{eff}})|m_1' = 3/2, m_2' = -5/2\rangle\langle m_1' = 3/2, m_2' = -5/2|s_{z2}s_{y1}|m_1 = 5/2, m_2 = -5/2\rangle = 75/(2g\beta H^{\mathrm{eff}}),$$

$$\langle m_1 = 5/2, m_2 = -5/2|-s_{z2}s_{y1}|m_1' = 3/2, m_2' = -5/2\rangle\langle m_1' = 3/2, m_2' = -5/2|\tilde{O}^1_{4j}/(g\beta H^{\mathrm{eff}})|m_1 = 5/2, m_2 = -5/2\rangle = 75/(2g\beta H^{\mathrm{eff}}),$$

$$s = 5/2, \quad m_1 = 5/2, \quad j = 2$$

$$\langle m_1 = 5/2, m_2 = -5/2|\tilde{O}^1_{4j}/(g\beta H^{\mathrm{eff}})|m_1' = 5/2, m_2' = -3/2\rangle\langle m_1' = 5/2, m_2' = -3/2|s_{z1}s_{y2}|m_1 = 5/2, m_2 = -5/2\rangle = 75/(2g\beta H^{\mathrm{eff}}),$$

$$\langle m_1 = 5/2, m_2 = -5/2|s_{z1}s_{y2}|m_1' = 5/2, m_2' = -3/2\rangle\langle m_1' = 5/2, m_2' = -3/2|\tilde{O}^1_{4j}/(g\beta H^{\mathrm{eff}})|m_1 = 5/2, m_2 = -5/2\rangle = 75/(2g\beta H^{\mathrm{eff}}).$$

Here $H^{\mathrm{eff}} = H_1^{\mathrm{eff}} = -H_2^{\mathrm{eff}}$.

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