ISSN 1063-7761, Journal of Experimental and Theoretical Physics, 2014, Vol. 119, No. 3, pp. 573–578. © Pleiades Publishing, Inc., 2014.
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ELECTRONIC PROPERTIES OF SOLID

Partial Differential Cross Section of Inelastic Magnetic Neutron Scattering in the Paramagnetic Phase of $LaCoO_3$

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Received March 11, 2014

Abstract—The partial differential cross section of inelastic magnetic neutron scattering from the compound $LaCoO_3$ in the paramagnetic phase is studied theoretically. The contribution to scattering from the high-spin state of an ion in zero magnetic field and the modification of this contribution upon application of a magnetic field are calculated using the effective Hamiltonian for the ^5D term. The amplitude of the peak in the dependence of the scattering cross section on the energy of scattered neutrons, which corresponds to the transition from the low-spin to the intermediate-spin state, is estimated.

DOI: 10.1134/S1063776114090027

1. Cobaltites (as well as manganites and cuprates) have attracted particular attention in recent decades. The cobalt ion in cobaltites is in the trivalent state Co^{3+} with the $3d^6$ electron configuration of the outer shell. In accordance with the Hund rule, the cobalt ion must be in a high-spin state with spin moment $S = 2$ and orbital angular momentum $L = 2$. Conversely, in strong crystal fields with symmetry corresponding to the surroundings of the Co^{3+} in lanthanum cobaltites, all electrons should occupy t_{2g} orbitals, forming a singlet nonmagnetic low-spin state with $S = 0$. Therefore, competition between the intratomic and crystal fields leads to small splitting between the low-spin (LS), intermediate-spin (IS), and high-spin (HS) states (Fig. 1). This in turn leads to singularities in the magnetic, electric, and structural properties of cobaltites [1].

Despite the fact that a number of theoretical [2, 3] and experimental publications, including works devoted to electron paramagnetic resonance (EPR) [4], X-ray spectroscopy, and X-ray circular dichroism [5, 6], evidence the competition between the LS and HS states, some experiments (e.g., measurement of magnetization [7] and magnetic susceptibility [8]) point to possible mixing of these states with the IS state. A number of theoretical works taking into account the hybridization of $3d$ orbitals of cobalt and $2p$ orbitals of oxygen demonstrated the stabilization of the IS state [9, 10]. Additional information on this problem can be obtained from experiments on inelastic neutron scattering, which have proven effective in the study of magnetic ions [11] and, in particular, crystal field effects on these ions [12].

In this study, we consider the results of theoretically calculating the partial differential cross section of inelastic magnetic scattering of neutrons, which may help in formulating and subsequently interpreting the experiment.

2. The partial differential cross section of magnetic neutron scattering (per site) in the first Born approximation has the form [13, 14]

$$\frac{d^2\sigma}{d\Omega dE} = r_0^2\frac{k'}{k}S(\kappa, E), \tag{1}$$

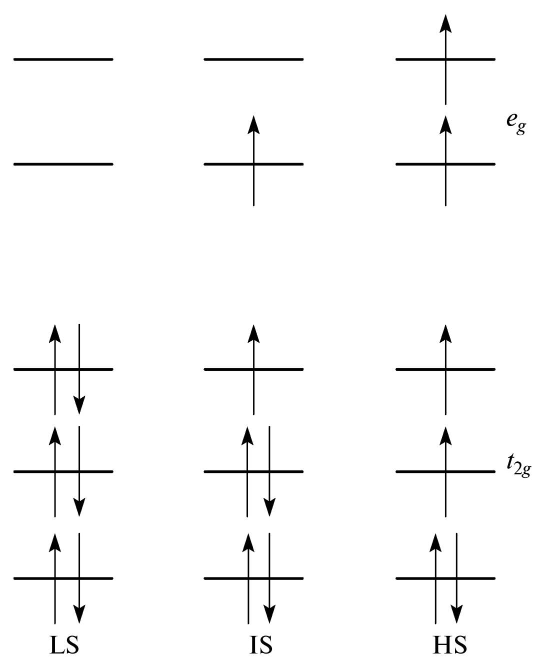

Fig. 1. Diagram of the electron configuration of the low-spin (LS), intermediate-spin (IS), and high-spin (HS) states of the Co^{3+} ion [1].

$$S(\kappa, E) = \frac{1}{N}\sum_{\alpha\beta} G_{\alpha\beta} \sum_{\lambda\lambda' l l'} p_\lambda \langle\lambda|\hat{F}^+_{l\alpha}|\lambda'\rangle\langle\lambda'|\hat{F}_{l'\alpha}|\lambda\rangle \times \delta(E - E_{\lambda'} + E_\lambda) e^{i\boldsymbol{\kappa}(\mathbf{l}' - \mathbf{l})}.$$

Here, the following notation has been introduced:

$$G_{\alpha\beta} \equiv \sum_\nu p_\nu \langle\nu|(\sigma_\alpha - e_\alpha(\boldsymbol{\sigma}\cdot\mathbf{e})) + (\sigma_\beta - e_\beta(\boldsymbol{\sigma}\cdot\mathbf{e}))|\nu\rangle, \quad (2)$$

$$\hat{F}_{l\alpha} = \frac{1}{2\mu_B}\int \hat{M}_{l\alpha}(\mathbf{r}) e^{i\boldsymbol{\kappa}(\mathbf{r} - \mathbf{l})} d\mathbf{r}, \quad (3)$$

where $\mathbf{M}_l$ is the magnetization associated with the spin moment and the orbital angular momentum of electrons of the ion in the **l** position, $r_0 = \gamma_n e^2/m_e c^2$, $\gamma_n = -1.91348$ is the gyromagnetic ratio for neutrons, $\boldsymbol{\kappa} = \mathbf{k} - \mathbf{k}'$ is the scattering vector, $\mathbf{e} = \boldsymbol{\kappa}/\kappa$, σ_α are the Pauli spin matrices acting in the space of spin states of a neutron, $|\nu\rangle$ are the spin parts of the neutron wavefunctions, p_ν is the statistical weight of the corresponding state in the beam, $|\lambda\rangle$ is the state of the scatterer, and p_λ is its statistical weight.

Compound $LaCoO_3$ is a paramagnetic dielectric in a wide temperature range [15]; at $T \longrightarrow 0$, it is transformed into a nonmagnetic state. This is due to the fact that the ground state of the Co^{3+} ion in lanthanum cobaltate is nonmagnetic and is separated from the magnetic states by activation energy $\tilde{\Delta} \sim 100$ K. Therefore, we should disregard the exchange interaction between magnetic ions at temperature $T < \tilde{\Delta}$ in view of the small population of the IS and HS states; at high temperatures, thermal fluctuations prevent the occurrence of magnetic order. Thus, at $T < 600$ K, the approximation of a paramagnet with noninteracting ions can be assumed to hold well for $LaCoO_3$.

In expression (1), λ and λ' describe the states of the entire scatterer and not of an individual ion. In this case, the product of matrix elements appearing in the sum in relation (1) for the paramagnet without interactions differs from zero only for $l = l'$ or $\lambda = \lambda'$. The latter condition corresponds to exclusively elastic neutron scattering, which is not considered here. For this reason, expression (1) can be written in a simpler form:

$$S(\kappa, E) = \sum_{\alpha\beta} G_{\alpha\beta} \sum_{nn'} p_n \langle n|\hat{F}^+_\alpha|n'\rangle \times \langle n|\hat{F}_\beta|n'\rangle \delta(E - E_{n'} + E_n), \quad (4)$$

where n and n' are the states of the cobalt ion. This expression corresponds to independent scattering from cobalt ions and holds for inelastic scattering from noninteracting ions in the paramagnetic phase (including the case when an external magnetic field is applied).

Introducing the mean values of the beam polarization projection [14] $P_\alpha = \sum_\nu p_\nu \langle\nu|\sigma_\alpha|\nu\rangle$, we can write coefficients $G_{\alpha\beta}$ in explicit form:

$$G_{\alpha\beta} = \delta_{\alpha\beta} - e_\alpha e_\beta + i\varepsilon_{\alpha\beta\gamma} e_\gamma (\mathbf{P}\cdot\mathbf{e}), \quad (5)$$

where $0 \le P \le 1$ and $\varepsilon_{\alpha\beta\gamma}$ is the Levi-Chivita symbol.

In many cases, formula (4) can be simplified. For example, if the spin–orbit interaction considerably exceeds the splitting in the crystal field, the vectorial form factor **F** can be written in the total momentum representation. Conversely, when the crystal field considerably exceeds the spin–orbit interaction, the orbital angular momentum is "frozen," and we can take into account only inelastic scattering from the spin moment of the ion. In the case of lanthanum cobaltate under investigation, both interactions are of the same order of magnitude, and the contribution to the inelastic magnetic scattering of neutrons comes both from the scattering from the spin moment and the orbital angular momentum. In this connection, we must find exact wavefunctions to calculate the matrix elements $\langle n|\hat{F}_\alpha|n'\rangle$.

3. In the case of the $LaCoO_3$ compound, the singlet level of the LS state, which makes zero contribution to magnetic scattering of neutrons with low energies, possesses the lowest energy. According to EPR measurements, the triplet split into singly and doubly degenerate levels by the crystal field of the nearest surroundings possesses the lowest energy in the HS state. Since the splitting is $\Delta E \sim 7.2$ K, the contribution to the magnetic neutron scattering associated with transitions between these energy levels must be significant.

To determine the wavefunctions of the cobalt ion in $LaCoO_3$, we used the model proposed in [16]. In this case, the Hamiltonian was represented in the form

$$\hat{H} = H_{\text{cub}} + \lambda\mathbf{L}\cdot\mathbf{S} + B_2^0 O_2^0(L, L_z) - \mu_B\mathbf{H}\cdot(2\mathbf{S} + \mathbf{L}),$$
$$H_{\text{cub}} = -\frac{2}{3}B_4(O_4^0(L, L_z) - 20\sqrt{2}O_4^3(L, L_z)), \quad (6)$$

where the first term describes the crystal field with cubic symmetry, the second term describes the spin–orbit interaction, and the third term corresponds to the crystal field with trigonal symmetry, and the fourth term describes the interaction of an ion with the external magnetic field; the z axis is directed along the trigonal axis. The characteristic values obtained in EPR experiments are $B_4 = 200$ K, $\lambda = -185$ K, and $B_2^0 = 7.2$ K.

Taking into account all terms of Hamiltonian (6) sequentially and calculating each time the wavefunctions in perturbation theory for a degenerate level in the zeroth approximation, we obtained the wavefunctions characterizing the lower triplet of the HS state in zero magnetic field:

$$\begin{aligned}|\tilde{0}\rangle &= -\frac{1}{\sqrt{5}}|2, 1\rangle + \frac{1}{\sqrt{10}}|-1, 1\rangle + \sqrt{\frac{4}{10}}|0, 0\rangle \\ &\quad + \frac{1}{\sqrt{5}}|-2, -1\rangle + \frac{1}{\sqrt{10}}|1, -1\rangle, \\ |\tilde{1}\rangle &= -\sqrt{\frac{2}{5}}|2, 2\rangle + \frac{1}{\sqrt{5}}|-1, 2\rangle + \sqrt{\frac{3}{10}}|0, 1\rangle \\ &\quad + \frac{1}{\sqrt{15}}|-2, 0\rangle + \frac{1}{\sqrt{30}}|1, 0\rangle, \\ |\widetilde{-1}\rangle &= -\sqrt{\frac{2}{5}}|-2, -2\rangle + \frac{1}{\sqrt{5}}|1, -2\rangle + \sqrt{\frac{3}{10}}|0, -1\rangle \\ &\quad - \frac{1}{\sqrt{15}}|2, 0\rangle + \frac{1}{\sqrt{30}}|-1, 0\rangle.\end{aligned} \tag{7}$$

The energy gap between states $|\tilde{0}\rangle$ and $|\widetilde{\pm 1}\rangle$ in this case is $\Delta E_0 = 9B_2^0/10 \approx 7$ K. Here, the wavefunctions of the triplet are expressed in terms of the wavefunctions of a free ion, $|L_z, S_z\rangle$.

The application of a strong magnetic field may substantially change the pattern especially when the field is directed at an angle to the trigonal axis:

$$H_z = H\cos\theta, \quad H_x = H\sin\theta\cos\phi,$$
$$H_y = H\sin\theta\sin\phi.$$

Field $H \sim 3$ T can be treated as strong. The set of energies (in the first approximation in B_2^0/H) and the wavefunctions (in the zeroth approximation) in this case is written in the form

$$\begin{aligned}E_0 &= \frac{3}{10}B_2^0(1 - 3\cos^2\theta) \\ \psi_0 &= -\frac{e^{-i\phi}}{\sqrt{2}}\sin\theta|\tilde{1}\rangle + \cos\theta|\tilde{0}\rangle + \frac{e^{i\phi}}{\sqrt{2}}\sin\theta|\widetilde{-1}\rangle,\end{aligned} \tag{8}$$

$$\begin{aligned}E_1 &= \frac{3}{10}B_2^0\left(1 - \frac{3}{2}\sin^2\theta\right) - g\mu_B H, \\ \psi_1 &= \frac{e^{-i\phi}}{2}\frac{\sin^2\theta}{\cos\theta - 1}|\tilde{1}\rangle - \frac{\sin\theta}{\sqrt{2}}|\tilde{0}\rangle - \frac{e^{i\phi}}{2}\frac{\sin^2\theta}{\cos\theta + 1}|\widetilde{-1}\rangle,\end{aligned} \tag{9}$$

$$\begin{aligned}E_{-1} &= \frac{3}{10}B_2^0\left(1 - \frac{3}{2}\sin^2\theta\right) + g\mu_B H, \\ \psi_{-1} &= \frac{e^{-i\phi}}{2}\frac{\sin^2\theta}{\cos\theta + 1}|\tilde{1}\rangle - \frac{\sin\theta}{\sqrt{2}}|\tilde{0}\rangle - \frac{e^{i\phi}}{2}\frac{\sin^2\theta}{\cos\theta - 1}|\widetilde{-1}\rangle,\end{aligned} \tag{10}$$

where $g = 3.5$, which is close to the value of $g = 3.35$ obtained in the EPR experiment.

In the case of magnetic scattering of neutrons characterized by small vectors $\boldsymbol{\kappa}$ of scattering from the cobalt ion, we can confine our analysis to the dipole approximation. This approximation may lead to considerable errors in calculating the amplitudes of scattering from one-electron shells in strong magnetic fields; however, in the case of near half-filling of the shell, this approximation gives an insignificant error. In this case, we can write

$$\begin{aligned}\mathbf{F} &= \langle j_0(\kappa)\rangle\mathbf{S} + \frac{1}{2}(\langle j_0(\kappa)\rangle + \langle j_2(\kappa)\rangle)\mathbf{L}, \\ \langle j_K(\kappa)\rangle &= \int r^2 R_{nl}^2 j_K(\kappa r)dr,\end{aligned} \tag{11}$$

where $j_K(x)$ is the spherical Bessel function and R_{nl} is the radial part of the hydrogen-like wavefunction of an electron on the orbital with principal quantum number n and orbital quantum number l. In the case of $3d$ electrons, this function has the form

$$R_{3d} = \sqrt{\left(\frac{2}{3a_0}\right)^3\frac{1}{6!}}\left(\frac{2r}{3a_0}\right)^2 e^{-r/3a_0}. \tag{12}$$

Using this approximation, we can easily obtain the sought expression for the partial differential cross section of inelastic magnetic scattering of neutrons in $LaCoO_3$. In zero magnetic field, function $S(\kappa, E)$ (4) has the form

$$\begin{aligned}S(\kappa, E) &= \mathcal{A}_0[p_0\delta(E - \Delta E_0) + p_1\delta(E + \Delta E_0)], \\ \mathcal{A}_0 &= 2K(1 + e_z^2), \quad \Delta E_0 = \frac{9}{10}B_2^0, \\ K &= \frac{(7\langle j_0(\kappa)\rangle + \langle j_2(\kappa)\rangle)^2}{32}.\end{aligned} \tag{13}$$

In strong fields $H > 3$ T, we have

$$\begin{aligned}S(\kappa, E) &= \mathcal{A}_+[p_0\delta(E - \Delta E_1) + p_{-1}\delta(E + \Delta E_{-1})] \\ &\quad + \mathcal{A}_-[p_0\delta(E - \Delta E_{-1}) + p_1\delta(E + \Delta E_1)], \\ \mathcal{A}_\pm &= K(1 + e_\parallel^2) \pm 2Ke_\parallel(\mathbf{P}\cdot\mathbf{e}), \\ \Delta E_1 &= \frac{9}{10}B_2^0\left(\cos^2\theta - \frac{\sin^2\theta}{2}\right) - g\mu_B H, \\ \Delta E_{-1} &= \frac{9}{10}B_2^0\left(\cos^2\theta - \frac{\sin^2\theta}{2}\right) + g\mu_B H,\end{aligned} \tag{14}$$

$e_\parallel$ is the projection of vector $\mathbf{e}$ onto the magnetic field direction, $p_0 \sim \exp(-\tilde{\Delta}/T)$, $p_{\pm 1} \sim \exp(-\tilde{\Delta}/T) \times \exp(\mp\Delta E_{\pm 1}/T)$, where $\tilde{\Delta} \approx 140$ K is the activation energy of the transition between the LS and HS states.

Comparison of the calculated curves for the polycrystalline sample and experimental data is illustrated in Fig. 2. In our calculations, we assumed that the z axis of cobalt is oriented arbitrarily relative to the directions of the magnetic field and the neutron beam

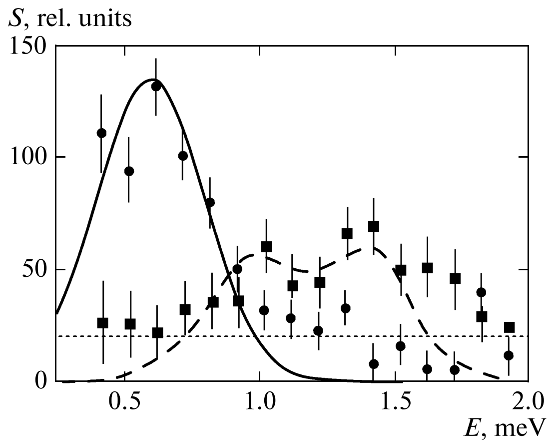

Fig. 2. Partial differential cross section of inelastic scattering of neutrons from $LaCoO_3$ in relative units at $T = 50$ K. The solid curve corresponds to calculations for a polycrystalline sample fpr $H = 0$; the dashed curve corresponds to $H = 6$ T; symbols correspond to experimental data from [17].

and that the broadening of δ-peaks was described by the Gaussian function with $\sigma = 0.1$ meV for scattering in the presence of a magnetic field and with $\sigma = 0.2$ meV in zero magnetic field; this agrees with the experimental error indicated in [17]. An insignificant discrepancy is associated with the problem of directly singling out the inelastic peaks against the background of a weak signal due to elastic scattering, which was mentioned in [17], as well as with the lack of data on the experimental geometry, such as the scattering angle and the angle between the magnetic field and the incident beam.

4. A characteristic feature of $LaCoO_3$ is small splitting between the ^5D energy level, which gives the HS state ^1H corresponding to the LS state and probably ^3G energy level corresponding to the IS state. In spite of its advantages, the EPR has the following significant limitation: it permits the measurement of energy gaps between the states with different projections of the total angular momentum, but does not reveal the peaks associated with transitions between the states with different values of the total angular momentum. In contrast to EPR, no limitation on the initial and final states is imposed in the case of magnetic scattering of neutrons, and the amplitudes of peaks in the partial differential cross section of inelastic magnetic scattering of neutrons, which correspond to transitions between different energy levels, may differ from zero. In this connection, it is important to estimate the peak amplitudes in the partial differential cross section of inelastic magnetic scattering of neutrons, which correspond to transitions $LS \longrightarrow HS$ and $LS \longrightarrow IS$ because this will allows us to find out whether these peaks can be detected in experiment.

Since the energy levels considered above differ in the values of spin moments, the contributions from the orbital terms appearing in the matrix elements $\langle n'|\hat{F}_\alpha|n\rangle$ are equal to zero if $|n\rangle$ and $|n'\rangle$ correspond to the states for different energy levels. In this case, the expression for the matrix elements acquires the simple form

$$\langle n'|\mathbf{F}|n\rangle = \left\langle n'\left|\sum_j \mathbf{s}_j e^{i\boldsymbol{\kappa}\cdot\mathbf{r}_j}\right|n\right\rangle, \quad (15)$$

where the sum over j is carried out over all electrons of the partly filled shell of the ion and $\mathbf{s}_j$ are one-electron spin operators.

In the case of interest to us, when the electron is on the $3d$ orbital, matrix elements $\langle m'|e^{i\kappa r}|m\rangle$ can be calculated exactly:

$$\begin{aligned}
&\langle 0|e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}|\mp 1\rangle \\
&= \mp\frac{8}{\sqrt{6}}\frac{\tilde{\kappa}^2 e_z\sqrt{1-e_z^2}}{(1+\tilde{\kappa}^2)^6}[1+6\tilde{\kappa}^2-15\tilde{\kappa}^2e_z^2]e^{\mp i\phi_\kappa}, \\
&\langle \pm 2|e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}|\pm 1\rangle \\
&= \pm\frac{4\tilde{\kappa}^2 e_z\sqrt{1-e_z^2}}{(1+\tilde{\kappa}^2)^6}[2-3\tilde{\kappa}^2+5\tilde{\kappa}^2e_z^2]e^{\mp i\phi_\kappa}, \\
&\langle \pm 1|e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}|\mp 1\rangle \\
&= \pm\frac{4\tilde{\kappa}^2(1-e_z^2)}{(1+\tilde{\kappa}^2)^6}[1+\tilde{\kappa}^2-10\tilde{\kappa}^2e_z^2]e^{\mp 2i\phi_\kappa}, \\
&\langle \pm 2|e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}|0\rangle \\
&= \frac{4}{\sqrt{6}}\frac{\tilde{\kappa}^2(1-e_z^2)}{(1+\tilde{\kappa}^2)^6}[2-3\tilde{\kappa}^2+15\tilde{\kappa}^2e_z^2]e^{\mp 2i\phi_\kappa}, \quad (16)\\
&\langle \pm 1|e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}|\mp 2\rangle = \mp\frac{20\tilde{\kappa}^4 e_z\sqrt{1-e_z^2}^3}{(1+\tilde{\kappa}^2)^6}e^{\mp 3i\phi_\kappa}, \\
&\langle \pm 2|e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}|\mp 2\rangle = \frac{10\tilde{\kappa}^4 e_z(1-e_z^2)^2}{(1+\tilde{\kappa}^2)^6}e^{\mp 4i\phi_\kappa}, \\
&e^{\mp i\phi_\kappa} = \frac{\tilde{\kappa}_x \mp i\tilde{\kappa}_y}{|\tilde{\kappa}_x \mp i\tilde{\kappa}_y|} = \frac{e_x \mp ie_y}{\sqrt{1-e_z^2}}, \\
&\tilde{\kappa} = \kappa\frac{3}{2a_0}.
\end{aligned}$$

It should be noted that the dipole approximation is inapplicable in principle for describing the peak associated with scattering with a change of the energy level because the corresponding matrix elements vanish in this approximation.

In the multielectron case, the problem can be reduced to the one-electron case by a transition to the second-quantization representation [18], in which the spin part of operator **F** has the form

$$F_- = \sum_{m'm} d^\dagger_{m'\downarrow} d_{m\uparrow} f_{m'm},$$

$$F_+ = \sum_{m'm} d^\dagger_{m'\uparrow} d_{m\downarrow} f_{m'm}, \quad (17)$$

$$F_z = \sum_{m'm\sigma} \sigma d^\dagger_{m'\sigma} d_{m\sigma} f_{m'm},$$

$$f_{m'm} = \langle m'|e^{i\boldsymbol{\kappa}\cdot\mathbf{r}}|m\rangle.$$

The features of the symmetry of the multielectron system are such that all matrix elements $\langle HS|\mathbf{F}|LS\rangle$ vanish; for this reason, it is impossible to observe the transition between these two states in experiments on inelastic neutron scattering. Matrix elements $\langle IS|\mathbf{F}|LS\rangle$ differ from zero, and if the IS state possesses an energy comparable with the energies of the LS and HS states, the corresponding peak can be observed in the partial differential cross section of the magnetic scattering of neutrons.

In evaluating the peak height, we assumed that all energy quantities appearing in Hamiltonian (6) are identical for the IS and HS states. In such a case, the expressions for the sought matrix elements (15) have the form

$$\langle IS|F_z|LS\rangle = -0.3062i|f_{2-1}|\sin\phi_\kappa,$$

$$\begin{aligned}\langle IS|F_x|LS\rangle = i(&-0.0722|f_{21}|\sin\phi_\kappa + 0.1768|f_{10}|\sin\phi_\kappa \\ &+ 0.1250|f_{20}|\sin 2\phi_\kappa - 0.1021|f_{1-1}|\sin 2\phi_\kappa \\ &+ 0.1021|f_{2-2}|\sin 4\phi_\kappa),\end{aligned} \quad (18)$$

$$\begin{aligned}\langle IS|F_y|LS\rangle = i(&0.0722|f_{21}|\cos\phi_\kappa + 0.1768|f_{10}|\cos\phi_\kappa \\ &+ 0.1250|f_{20}|\cos 2\phi_\kappa + 0.1021|f_{1-1}|\cos 2\phi_\kappa \\ &- 0.1021|f_{2-2}|\sin 4\phi_\kappa).\end{aligned}$$

For a polycrystalline sample, the peak amplitude in the partial differential cross section of inelastic magnetic scattering of neutrons corresponding to the $LS \longrightarrow IS$ transition can amount to 10^{-1} to 10^{-2} of the amplitude of the well-resolved experimental peak associated with transitions within the split triplet in the HS state. The detection of such a peak would solve the problem of realization of the IS state of Co in the $LaCoO_3$ compound.

5. In conclusion, we outline the main results of this study. Using the Hamiltonian proposed in [16] for describing the EPR experiment, we have calculated the amplitudes and positions of the peaks associated with transitions within the lower triplet of the HS state of the cobalt ion. Comparison with experiment demonstrates good agreement between the results of theoretical calculations and experimental data. It is shown that if the IS state possesses an energy comparable with the energies of the LS and HS states, the peak corresponding to the transition from the LS to the IS state should be observed. Under the assumption that the Hamiltonian describing the IS state contains the same energy parameters as the Hamiltonian of the HS state, we estimated the amplitude of the peak corresponding to the transition of the cobalt ion from the LS to the IS state.

ACKNOWLEDGMENTS

The author is grateful to V.V. Val'kov, E.S. Klement'ev, and P.A. Alekseev for numerous discussions and persistent interest in this work.

This study was supported by the Russian Foundation for Basic Research (project nos. 13-02-00523 A, 14-02-31280 mol_a, and 13-02-98013).

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Translated to N. Wadhwa