Relaxation Mössbauer spectra in cubic systems

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A method is proposed for calculating, within the framework of stochastic theory, the relaxation hyperfine Mössbauer spectra in the presence of cubic symmetry. An adequate physical object is, for example, superparamagnetic particles of cubic symmetry. Two examples with different orientations of the easy magnetization axes with respect to the cube symmetry axes are considered. A simple analytic expression is obtained which describes the spectrum throughout the relaxation-parameter range.

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1. INTRODUCTION

In the analysis of the experimental relaxation Mössbauer spectra one uses as a rule formulas based on the simplest relaxation-process model, according to which the electron spin producing the magnetic hyperfine field reverses randomly its direction (see, e.g., [1,2]). It is clear that by far not all experimental situations correspond to such a model. The theory developed by now makes it possible in principle to take into account relaxation processes that differ greatly both in complexity and in character (see, e.g., [3–8]), but the final results are represented in a form that is not suitable for a direct analysis of the experimental spectra. In fact, it is necessary to carry out additional cumbersome calculations involving inversion of matrices of rather high rank. It is therefore urgent at present to find cases that admit of a simpler description of their relaxation spectra. One such example was considered recently—the so-called strong-collision model—and the obtained formulas for the calculation of the spectra call for inversion of much smaller matrices.

We consider in this paper models describing the relaxation of superparamagnetic particles of cubic symmetry. The presence of cubic symmetry makes it possible to obtain rather simple expressions for the description of the relaxation spectra.

The formulation of the problem and the derivation of the basic equations is contained in Sec. 2. The following section is devoted to a brief analysis of the results.

2. FORMULATION OF PROBLEM AND DERIVATION OF GENERAL FORMULAS

We consider magnetic matter containing nuclei of the Mössbauer type. The magnetic electronic system produces at the nucleus a hyperfine magnetic field $\mathbf{H}_{hf} = \mathbf{n}H_{hf}$, the direction $\mathbf{n}$ of which is assumed to vary in time as a result of relaxation processes. The change of $\mathbf{n}$ with time will be specified by a stochastic model, according to which the vector $\mathbf{n}$ can assume N different positions $\mathbf{n}_a$ ($a = 1, \ldots, N$), between which random transitions occur with transition probabilities p_{ab} per unit time. If we choose these directions to be a certain family of equivalent symmetry axes of the cube, then we obtain a certain relaxation-process model for spherical supermagnetic particles of cubic symmetry.

The theory of relaxation Mössbauer spectra was developed by Blume, [7] and we shall use his results. According to [7], the resonance absorption spectrum $\varphi_{abs}(\omega)$ is given, apart from the inessential constant, by the formula

$$\varphi_{abs}(\omega) = -\operatorname{Im}\{(\mathbf{j}\eta)\rho\hat{G}(\omega+i\Gamma/2)(\mathbf{j}^{+}\eta^{*})E\}, \tag{1}$$

where η is the polarization vector of the incident γ radiation, $\mathbf{j}$ is the operator of the nuclear current responsible for the transitions between the ground ($|m_g\rangle$) and excited ($|m_e\rangle$) states of the nucleus, E is a unit column, and ρ is a row having the dimensionality N of the space of the electronic spin system. The elements ρ_a ($a = 1, \ldots, N$) are the relative probabilities of the populations of the electronic states $|a\rangle$; $\hat{G}(\omega + i\Gamma/2)$ is a superoperator acting in the space of the function $|m_e\rangle\langle m_g||a)$, and its form is

$$\hat{G}^{-1}(\omega+i\Gamma/2) = \omega - \hat{\Omega}_{hf} - i\hat{P} + i\Gamma/2. \tag{2}$$

Here $\hat{\Omega}_{hf}$ is the superoperator responsible for the hyperfine splitting of the nuclear levels and it is obviously diagonal in the variables of the electronic system a, that is,

$$(a|\hat{\Omega}_{hf}|b) = \hat{\omega}(a)\delta_{ab} \tag{3}$$

and the matrix elements of the superoperator $\hat{\omega}(a)$ are determined by the formulas

$$\hat{\omega}(a)_{m_e'm_g', m_em_g} = \mu_e H_{hf}(\mathbf{n}_a\mathbf{I}_e)_{m_e'm_e} - \mu_g H_{hf}(\mathbf{n}_a\mathbf{I}_g)_{m_gm_g'}, \tag{4}$$

where μ_g, I_g, μ_e, and I_e are the magnetic moments and spin operators of the nuclei in the ground and excited states, respectively.

The superoperator $\hat{P}$ describes relaxation processes in the electronic system. It is diagonal in the nuclear variables:

$$\hat{P}_{m_e'm_g'a', m_em_ga} = p_{a'a}\delta_{m_e'm_e}\delta_{m_g'm_g} \tag{5}$$

and the quantities $p_{a'a}$ (at $a' \neq a$) are the probabilities of the transition per unit time from the state $|a'\rangle$ to the state $|a\rangle$. The diagonal elements are determined by the relation

$$P_{aa} = -\sum_{a'\neq a} p_{aa'}. \tag{6}$$

Formulas (1)–(6) determine completely the absorption spectrum, but in concrete calculations it is necessary to deal with inversion of matrices of rather high rank. Indeed, in the case of the Fe^{57} nucleus ($I_g = \frac{1}{2}$ and $I_e = \frac{3}{2}$),

taking into account the fact that the smallest possible value of the equivalent directions $\mathbf{n}_a$ in the cubic systems is equal to six, we are dealing with a matrix of order 48 or higher. On the other hand, it is clear that the presence of cubic symmetry should lead to simplification of formula (1), as will indeed by demonstrated below.

We shall assume that all the electronic states are equally populated, as is the case when no external magnetic field acts on the superparamagnetic particle.

Since it is clear that the absorption spectrum (1) in cubic systems does not depend on the polarization vector η of the incident radiation, we shall direct it along an arbitrary axis z and choose this axis as the quantization axis. We confine ourselves henceforth to Mössbauer nuclei of the type Fe^{57} with nuclear ground-state spin $I_g = \frac{1}{2}$ and with excited-state spin $I_e = \frac{3}{2}$. Recognizing that the nuclear current operator j_g has matrix elements between states with coinciding quantum numbers $m_e = m_g = m$, we rewrite (1), apart from a constant factor, in the form

$$\varphi_{abs}(\omega) = -\mathrm{Im} \sum_{ma} \hat{G}_{mma,mma}(\omega + i\Gamma/2). \tag{7}$$

We assume now that the possible directions of the hyperfine field at the nucleus $\mathbf{n}_a$ coincide with the fourfold axis of the cube (see Fig. 1). Such a model corresponds to the situation when these axes are the easiest magnetization axes of a superparamagnetic particle. Out of the 48 states $|m_e\rangle\langle m_g||a)$ that form the basis of the space in which the superoperators $\hat{\Omega}$ and $\hat{P}$ act, we can construct four half-spaces, the states of which are either invariant to the action of all the rotations about the fourfold axis z, or acquire factors -1 and $\pm i$. The superoperators $\hat{\Omega}$, P, and $\hat{G}$ have no matrix elements between states belonging to such different irreducible representations of the subgroup C_4. In turn, the invariant half-space breaks up into two aggregates of six states with different symmetry properties relative to the operation of time inversion. According to (7), we are interested only in that aggregate which contains a fully symmetrical combination of the states $|m\rangle\langle m||a)$:

$$|1\rangle = \frac{1}{2\sqrt{3}}(|1) + |2) + |3) + |\bar{3}) + |\bar{2}) + |\bar{1})) (|^1/_2\rangle\langle^1/_2| + |-^1/_2\rangle\langle-^1/_2|).$$

The remaining five functions are the following:

$$|2\rangle = \frac{1}{\sqrt{6}}(|1) - {}^1/_2|2) - {}^1/_2|3) - {}^1/_2|\bar{3}) - {}^1/_2|\bar{2}) + |\bar{1})) (|^1/_2\rangle\langle^1/_2| + |-^1/_2\rangle\langle-^1/_2|),$$

$$|3\rangle = \frac{1}{2\sqrt{2}}(|2) - |3) + |\bar{2}) - |\bar{3})) (|^3/_2\rangle\langle-^1/_2| + |-^3/_2\rangle\langle^1/_2|),$$

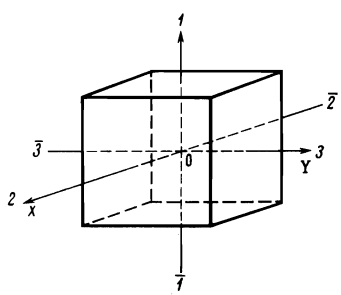

FIG. 1.

$$|4\rangle = \frac{1}{2\sqrt{2}}[(|2) + i|3) - |\bar{2}) - i|\bar{3})) |-^3/_2\rangle\langle-^1/_2| +$$
$$+ (|2) - i|3) - |\bar{2}) + i|\bar{3})) |^3/_2\rangle\langle^1/_2|],$$

$$|5\rangle = \frac{1}{2\sqrt{2}}[(|2) + i|3) - |\bar{2}) - i|\bar{3})) |-^1/_2\rangle\langle^1/_2| +$$
$$+ (|2) - i|3) - |\bar{2}) + i|\bar{3})) |^1/_2\rangle\langle-^1/_2|],$$

$$|6\rangle = {}^1/_2(|1) - |\bar{1})) (|^1/_2\rangle\langle^1/_2| - |-^1/_2\rangle\langle-^1/_2|) \tag{8}$$

Thus, the problem of inverting a matrix of 48th order has been reduced to inversion of a matrix of 6th order only. In addition, according to (7) and (8), the absorption spectrum is determined only by the only matrix element of the superoperator G, namely G_{11}.

In the subspace formed by the functions (8), the matrix G^{-1} takes the following form:

$$\hat{G}^{-1}(\omega + i\Gamma/2) = \begin{pmatrix} (\omega + i\Gamma/2)E + i\hat{P}_1 & \hat{\Omega} \\ \hat{\Omega} & (\omega + i\Gamma/2)E + i\hat{P}_1 \end{pmatrix}, \tag{9}$$

where $\hat{\Omega}$ is a 3×3 matrix with matrix elements

$$\hat{\Omega} = \begin{pmatrix} -\frac{A_e}{\sqrt{2}} & \frac{-2A_e + A_g}{\sqrt{6}} & \frac{-A_e + A_g}{2\sqrt{3}} \\ \frac{A_e}{2} & \frac{2A_e - A_g}{2\sqrt{3}} & \frac{-A_e + A_g}{\sqrt{6}} \\ \frac{A_g}{2} & -\frac{\sqrt{3}}{2}A_e & 0 \end{pmatrix},$$

$\hat{\Omega}'$ is its transpose, and the parameters $A_{e,g}$ are connected with a hyperfine field: $A_{e,g} = \mu_{e,g} H_{hf}$. The relaxation matrices P_1 and P_2 are diagonal and take the form

$$\hat{P}_1 = \begin{pmatrix} 0 & & \\ & 6p & \\ & & 6p \end{pmatrix}, \quad \hat{P}_2 = (4p + 2q)E.$$

Here E is a unit matrix, p is the probability of transition per unit time between the states $|a)$ with change of direction of the vector $\mathbf{n}_a$ by an angle $\pi/2$, and q is the probability of a transition with reversal of the direction of $\mathbf{n}_a$.

According to the foregoing, the absorption spectrum will be determined by the formula

$$\varphi_{abs}(\omega) = -\mathrm{Im}\,(\Delta_{11}/\Delta), \tag{10}$$

where Δ is the determinant of the matrix (9) and Δ_{11} is its principal minor.

Formula (10) can be easily transformed into

$$\varphi_{abs}(\omega) = -\mathrm{Im}\,\frac{1}{6}\frac{\varphi_0(\omega) + \varphi_1(\omega)}{1 - ip\varphi_0(\omega)}, \tag{11}$$

where

$$\varphi_0(\omega) = \frac{\omega_1}{\omega_1\omega_2 - \beta_3^2} + \frac{2\omega_1}{\omega_1\omega_2 - \beta_2^2} + \frac{3\omega_1}{\omega_1\omega_2 - \beta_1^2},$$
$$\varphi_1(\omega) = -\frac{16q(p-q)}{(\omega_1\omega_2 - \beta_1^2)(\omega_1\omega_2 - \beta_2^2)(\omega_1\omega_2 - \beta_3^2)},$$
$$\omega_1 = \omega + i(4p + 2q + \Gamma/2), \quad \omega_2 = \omega + i(6p + \Gamma/2),$$
$$\beta_1 = {}^1/_2(-3A_e + A_g), \quad \beta_2 = {}^1/_2(-A_e + A_g), \quad \beta_3 = {}^1/_2(A_e + A_g).$$

Here β_i are the frequencies of the hyperfine transitions. We have thus obtained in our case a simple analytic expression for the form of the absorption spectra.

The method developed above can be used also in the analysis of more complicated situations. We consider

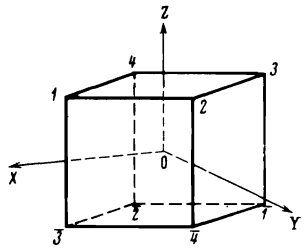

FIG. 2.

by way of a second example the case when the possible directions of the magnetic field at the nucleus coincide with threefold axes of the cube (see Fig. 2).

This model corresponds to the situation wherein the easiest-magnetization axes are the principal diagonals of the cube. The number of directions of the vector $\mathbf{n}_a$ is now eight, and the matrix $\hat{G}$ is of order 64. To construct a subspace that is invariant to the action of the elements of the group C_4, we introduce combinations of stochastic states $|a)$ such that they acquire a factor $c_k = i^k$ $(k = 0, 1, 2, 3)$ when rotated about the z axis through the angle $\pi/2$

$$K_k = |1) + c_k|2) + c_k^2|3) + c_k^3|4).$$

The analogous construction consisting of inverse states ($\bar{1}$ etc.), will be designated $\bar{K}_k$.

The subspace of interest to us is formed by the following functions:

$$|1\rangle = {}^1/_4(K_0 + \bar{K}_0)(|{}^1/_2\rangle\langle{}^1/_2| + |-{}^1/_2\rangle\langle-{}^1/_2|),$$
$$|2\rangle = {}^1/_4[(K_3 + \bar{K}_3)|{}^3/_2\rangle\langle{}^1/_2| - (K_1 + \bar{K}_1)|-{}^3/_2\rangle\langle-{}^1/_2|],$$
$$|3\rangle = {}^1/_4[(K_3 + \bar{K}_3)|{}^1/_2\rangle\langle-{}^1/_2| - (K_1 + \bar{K}_1)|-{}^1/_2\rangle\langle{}^1/_2|],$$
$$|4\rangle = {}^1/_4(K_2 + \bar{K}_2)(|{}^3/_2\rangle\langle-{}^1/_2| + |-{}^3/_2\rangle\langle{}^1/_2|),$$
$$|5\rangle = {}^1/_4(K_0 - \bar{K}_0)(|{}^1/_2\rangle\langle{}^1/_2| - |-{}^1/_2\rangle\langle-{}^1/_2|),$$
$$|6\rangle = {}^1/_4[(K_1 - \bar{K}_1)|-{}^3/_2\rangle\langle-{}^1/_2| + (K_3 - \bar{K}_3)|{}^3/_2\rangle\langle{}^1/_2|],$$
$$|7\rangle = {}^1/_4[(K_1 - \bar{K}_1)|-{}^1/_2\rangle\langle{}^1/_2| + (K_3 - \bar{K}_3)|{}^1/_2\rangle\langle-{}^1/_2|],$$
$$|8\rangle = {}^1/_4(K_2 - \bar{K}_2)(|{}^3/_2\rangle\langle-{}^1/_2| - |-{}^3/_2\rangle\langle{}^1/_2|).$$

The superoperator $\hat{G}^{-1}$ is a matrix of eighth order of the form (9). The 4×4 matrix $\hat{\Omega}$ now takes the form

$$\hat{\Omega} = \begin{pmatrix} \frac{-A_e+A_g}{2\sqrt{3}} & -\frac{1}{\sqrt{2}}A_e & \frac{-2A_e+A_g}{\sqrt{6}} & 0 \\ -\frac{1}{\sqrt{2}}A_e & \frac{-3A_e+A_g}{2\sqrt{3}} & 0 & \frac{1}{\sqrt{6}}A_g \\ \frac{2A_e+A_g}{\sqrt{6}} & 0 & -\frac{A_e+A_g}{2\sqrt{3}} & -\frac{1}{\sqrt{2}}A_e \\ 0 & \frac{1}{\sqrt{6}}A_g & -\frac{1}{\sqrt{2}}A_e & -\frac{3A_e+A_g}{2\sqrt{3}} \end{pmatrix} \quad (12)$$

The relaxation matrices $\hat{P}_1$ and $\hat{P}_2$ are diagonal, as before:

$$\hat{P}_1 = \begin{pmatrix} 0 & & & \\ & 4(p+q) & & \\ & & 4(p+q) & \\ & & & 4(p+q) \end{pmatrix},$$

$$\hat{P}_2 = \begin{pmatrix} 2(p+2q+r) & & & \\ & 2(p+2q+r) & & \\ & & 2(p+2q+r) & \\ & & & 2(3p+r) \end{pmatrix}.$$

Here p and q are the probabilities, per unit time, of the transitions with the direction of the hyperfine field turned through the angles between neighboring and opposite diagonals of the cube, respectively, and r is the probability of the transition with reversal of the direction of the hyperfine field.

The absorption spectrum will be given by Eq. (10), where Δ is the determinant of the matrix (9), (12) while Δ_{11} is its principal minor.

3. DISCUSSION AND ANALYSIS OF THE RESULTS

We were thus able to simplify considerably the cumbersome problem of finding the form of the relaxation spectra in a cubic system. In the first example discussed above, when the possible directions of the hyperfine field at the nucleus coincide with fourfold axes of the cube (Fig. 1), the situation reduced to inversion of a sixth-order matrix containing two relaxation parameters. In the second example, when the hyperfine field varies between directions specified by the principal diagonals of the cube, an eighth-order matrix containing three relaxation parameters must be inverted to find the form of the spectrum. By varying the values it is possible to describe a large number of experimental spectra.

We note that in the strong-collision model, when all the relaxation frequencies are equal, the relaxation matrix differs from the unit matrix in the absence of the first element, and therefore formula (10) leads directly to an expression (11) in which φ_1 must be set equal to zero, in both considered examples. This result was recently obtained by Dattagupta and Blume.[9] We believe, however, that a more realistic situation from the experimental point of view is one in which the relaxation parameters are different.

With the aid of (10), and particularly (11), it is easy to analyze the dynamics of the variation of the hyperfine spectra with increasing relaxation parameters. We shall stop to consider here only the limiting values for slow and fast relaxation.

If $p,\ q,\ r \ll \beta_i$, then the influence of the relaxation reduces to broadening of the hyperfine-spectrum lines. In the first example, the width of the i-th line $(i = 1, 2, 3)$, corresponding to a transition at the frequency $\pm\beta_i$, differs from the natural width by an amount $\Delta\Gamma_i$, equal to $\Delta\Gamma_1 = 7p + 2q$, $\Delta\Gamma_2 = 8p + 2q$, $\Delta\Gamma_3 = 9p + 2q$. In the second example, the corresponding corrections are of the form

$$\Delta\Gamma_1 = {}^{38}/_9 p + {}^{52}/_9 q + 2r, \quad \Delta\Gamma_2 = {}^{16}/_3 p + {}^{14}/_3 q + 2r, \quad \Delta\Gamma_3 = {}^{20}/_3 p + {}^{22}/_3 q + 2r.$$

In the opposite limiting case of rapid relaxation, when $p,\ q,\ r \gg \beta_i$, the spectrum consists of a single Lorentz line with width differing by $\Delta\Gamma$ from the natural width. In the two examples considered above we obtain for $\Delta\Gamma$ the values

$$\Delta\Gamma^{(1)} = \frac{1}{3(4p+2q)}\left(\frac{15}{2}A_e^2 + \frac{3}{2}A_g^2 - 5A_eA_g\right),$$

$$\Delta\Gamma^{(2)} = \frac{1}{3(2p+4q+2r)}\left(\frac{15}{2}A_e^2 + \frac{3}{2}A_g^2 - 5A_eA_g\right),$$

which coincide with the results of[10], where the general case of relaxation spectra was investigated in the fast relaxation limit.

We note in the conclusion that the method developed

can be used to investigate relaxation spectra in more complicated situations, for example in the presence of quadrupole interaction or an external magnetic field, if the directions of these fields coincide with the symmetry axes of the cube. The orders of the matrices to be inverted, however, doubles in comparison with the examples analyzed above.

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Magnetic transitions in mutual solid solutions of lower iron and manganese silicides

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An investigation was made of the temperature dependences of the magnetization (4.2–900°K) and of the magnetic susceptibility (60–1200°K) of $(Fe_{1-x}Mn_x)_3Si$ solid solutions ($0 \leq x \leq 1$). The Mössbauer spectra of Fe^{57} in these solutions were recorded and a neutron diffraction study of their structure was made. The magnetic structures of the antiferromagnets Mn_3Si and $(Fe_{0.33}Mn_{0.67})_3Si$ were determined. Three ranges of compositions were distinguished in the magnetic phase diagram: 1) $0 \leq x < 0.33$, 2) $0.33 \leq x < 0.67$, 3) $0.67 \leq x \leq 1$, in which the low-temperature order was, respectively, ferromagnetic, ferrimagnetic, and antiferromagnetic.

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1. INTRODUCTION

Magnetic properties and magnetic structure of many transition-metal alloys (such as Ni–Mn, Ni–Fe, and others) are governed by the competition between exchange interactions of opposite signs. It is interesting to determine the laws governing the magnetic structure of multicomponent systems, the simplest of which are quasibinary solid solutions with an ordered distribution of transition-metal atoms. An example of such solutions is the quasibinary $(Fe_{1-x}Mn_x)_3Si$ system whose components Fe_3Si and Mn_3Si have the $D0_3$ structure and an unlimited mutual solubility.

In an earlier paper[1] we showed that an increase of x from 0 to 0.33 in this system results in the replacement of iron atoms with Mn only at the C sites, whose immediate environment includes eight A and B sites occupied by iron atoms. Further increase of x is accompanied by the replacement of iron atoms at the A and B sites whose immediate environment includes four C sites occupied by manganese atoms and four D sites occupied by silicon.

The magnetic properties of these solutions were investigated in the composition ranges $0 \leq x < 0.25$[2] and $0 \leq x < 0.6$[3] but their magnetic structure was not investigated in detail. All that is known is that Fe_3Si is a collinear ferromagnet [$\mu(Fe_C) = 2.2\mu_B$ and $\mu(Fe_{AB}) = 1.15\mu_B$][4] and Mn_3Si is an antiferromagnet (one of the variants of the magnetic structure of this antiferromagnet is described in[5]).

We investigated the influence of the composition ($0 \leq x \leq 1$) and temperature of $(Fe_{1-x}Mn_x)_3Si$ solid solutions on the magnetization (Sec. 3), magnetic susceptibility (Sec. 4), parameters of Mössbauer spectra (Sec. 5), and characteristics of magnetic scattering of neutrons (Sec. 6) with the aim of determining the conditions for the ferromagnetic–antiferromagnetic transition and the principal features of the magnetic phase diagram (Sec. 7).

2. SAMPLES AND INVESTIGATION METHODS

Our solid solutions were prepared from pure components (the impurity concentrations did not exceed 0.01% in iron, 0.02% in manganese, and 0.001% in silicon) in a hermetically sealed hf furnace and beryllium oxide crucibles. The melting and heat treatment conditions were described earlier.[1] A metallographic analysis demonstrated that samples of $(Fe_{1-x}Mn_x)_3Si$ with $0 \leq x \leq 0.6$ obtained in this way consisted of a single phase,