Effect of fluctuations in the magnetic moments of microscopic particles on the time evolution of Mössbauer scattering

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The time evolution of the Mössbauer radiation scattered by Fe^{57} nuclei, which are part of a superparamagnetic easy-axis particle, is derived. It is shown in the Kubo–Anderson model that the shape of the curves is determined by the average frequency of the magnetic-moment fluctuations of a microparticle.

The selective-excitation double-Mössbauer (SEDM) technique in Mössbauer spectroscopy[1] has been used to study fluctuation processes in superparamagnetic particles of geothite,[2] FeOOH. This technique consists basically of the selective excitation of a Mössbauer transition of the hyperfine structure of a nucleus and a frequency analysis of the scattered γ rays. It has been demonstrated by this technique that the Kubo–Anderson fluctuation model is the most satisfactory one for describing geothite microparticles.

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Information of no less importance regarding the nature of fluctuation processes in a superparamagnetic sample can be extracted from a temporal analysis of the scattered radiation in the SEDM geometry. Existing experimental methods make such an analysis possible.[3–5] As a first step in the present paper, we examine the effect of fluctuation processes on the time evolution of the scattered radiation according to the Kubo–Anderson model. This model is valid for an easy-axis superparamagnetic particle. To find the time evolution, we use the density-matrix formalism in the Schrödinger picture. This formalism has been used previously to study the effect of fluctuation processes and of an external rf field on Mössbauer absorption and scattering spectra.[6,7] Along that approach, one analyzes an expression for the population of the final state of the system consisting of the nucleus, the electronic subsystem, and the γ ray:

$$P(\omega_1,t)=-2\mathrm{Im}\sum\rho^{ge(\pm)}_{n_{2g}n_{2e}}(\omega_2,\omega_1,t)V^{eg}_{n_{2e}n_{2g}}/h, \tag{1}$$

where ρ is the density-matrix operator of the system, the operator V represents the interaction of the nucleus with the γ ray, g and e specify sublevels of the nuclear hyperfine structure of the ground and excited states, and the $\pm$ specify states of the electronic subsystem associated with the direction of the vector magnetic moment of the microparticle. The matrix elements of the operator ρ are found by the Fourier-transform method from a system of eight Liouville equations averaged over states of the thermal reservoir,[8]

$$\frac{d}{dt}\rho^{ij(\pm)}_{n_{2i}n_{1i},n_{2j}n_{1j}}=-\frac{i}{h}[H,\rho]^{ij(\pm)}_{n_{2i}n_{1i},n_{2j}n_{1j}}-\frac{1}{\tau}\rho^{ij(\pm)}_{n_{2i}n_{1i},n_{2j}n_{1j}}+\frac{1}{\tau}\rho^{ij(\pm)}_{n_{2i}n_{1i},n_{2j}n_{1j}}, \tag{2}$$

under the initial conditions $\rho^{ij(\pm)}_{n_{2i}n_{1i},n_{2j}n_{1j}}(0)=0$. In Eqs. (1) and (2), n_1 and n_2 are the occupation numbers of the incident and scattered γ rays, and $1/\tau$ is the average frequency of fluctuations between states of the electronic subsystem with $S=\pm 1/2$.

Since the γ rays incident on the detector in a time-varying experiment are not distinguished by frequency, an average of $P(\omega_1,t)$ is taken over the phase of the scattered radiation. Consequently, the curves of the time evolution acquire oscillations due to a transition of the nucleus to a superposition state only during the absorption of the incident radiation. In addition, there is a change in the observed decay rate of the excited nuclear state as a result of the Poisson probability $\exp(-t/\tau)$.

Substituting $\rho^{ge(\pm)}_{n_{2g}n_{2e}}$ from (2) into (1), we find an expression for the population of the final state of the system:

$$P(\omega_1,t)\sim\mathrm{Re}\sum d^{(L)}_{r_1M}(\vartheta_1)\chi^{(in)}_{r_1r}d^{(L)}_{rM}(\vartheta_1)d^{(L)}_{s_1M_1}(\vartheta_2)\chi^{(sc)}_{s_1s}d^{(L)}_{sM_1}(\vartheta_2)$$

$$\times\ C^2(I_eLI_g,m_eM)C^2(I_eLI_g,m_eM_1)\Phi_{egg_1}(\omega_1,t). \tag{3}$$

Here $d^{(L)}$ are Wigner functions, $C(\ldots)$ are Clebsch–Gordan coefficients, and $\chi^{(in)}$ and $\chi^{(sc)}$ are the polarization density matrices of the incident and scattered radiation. The function Φ_{egg_1} is given by

$$\Phi_{egg_1} = \sum_{\alpha=\pm 1} F_1(\alpha\Delta, a_{eg_1})$$

$$+ \sum_{\beta=\pm 1} F_2(\alpha\Delta, \beta a_{eg_1}) + F_3(\Delta, \alpha a_{gg}, \beta a_{eg_1}) + F_4(\Delta, \alpha a_{eg}, \beta a_{eg_1}). \qquad (4)$$

The function F_3, along with F_4 in expression (4), dominates the formation of the oscillations on the time evolution in (3). The former function is given by

$$F_3 = F_{30}(1 - \exp(ft)), \quad F_{30} = S/H, \quad H = -2ia_{eg}a_{eg_1}h_2h_3f, \quad S = h_1K + L,$$

$$h_1 = \Delta - a_{eg} - i\Gamma/2, \quad h_2 = h_1 - a_{gg_1}, \quad h_3 = h_1 + a_{gg_1},$$

$$f = i(\Delta - a_{eg_1}) + \Gamma/2 + 1/\tau,$$

$$L = \omega_{gg_1}(a_{eg}\omega_{eg_1} + \omega_{eg}a_{eg_1}) - i(\omega_{eg}^2 - \omega_{eg_1}\omega_{gg_1} - a_{eg}a_{eg_1})/\tau$$

$$+ (a_{eg_1} - a_{eg})/\tau^2 + i/\tau^3,$$

$$K = a_{eg}a_{eg_1} + \omega_{eg}\omega_{eg_1} - i(a_{eg} - a_{eg_1})/\tau + 1/\tau^2. \qquad (5)$$

In (4) and (5), Δ means the Doppler shift of the incident radiation, and ω_{ij} characterizes the Zeeman energy of the corresponding hyperfine transition of the nucleus. The quantity a_{ij} is related to ω_{ij} by $\sqrt{\omega_{ij}^2 - 1/\tau^2}$. It reflects the influence of the fluctuation processes on the hyperfine field at the nucleus. The quantity Γ is the natural linewidth of the γ radiation.

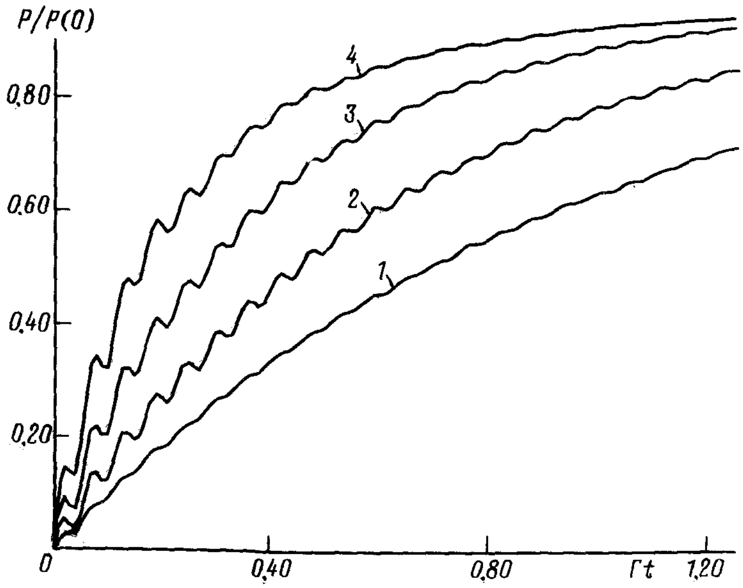

FIG. 1. Time evolution of the scattered radiation for several values of $1/\tau$. 1—0.1Γ; 2—Γ; 3—2Γ; 4—4Γ.

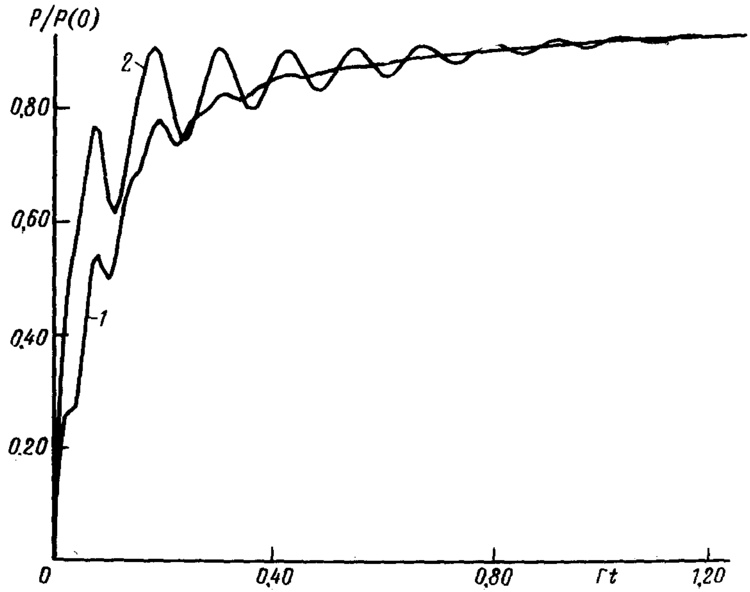

FIG. 2. Time evolution of the scattered radiation for two values of $1/\tau$. 1—8.6Γ; 2—17Γ.

The nucleus Fe^{57} ($I_g = 1/2$, $I_e = 3/2$) in a superparamagnetic sample has been used as an example to analyze the time evolution of the scattered radiation. It was assumed that the experimental curves were found by coincidence Mössbauer spectroscopy. Accordingly, an integration was carried out over ϑ_1 and ϑ_2 in (3), and an

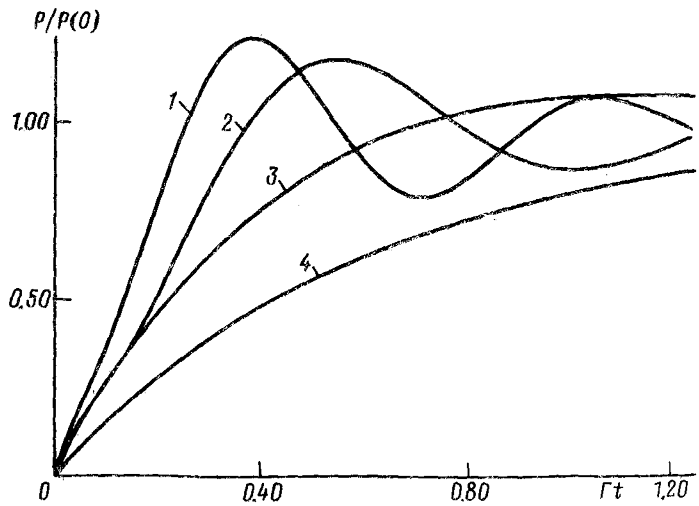

FIG. 3. Time evolution of the scattered radiation in case (2) for several values of $1/\tau$. 1—54Γ; 2—54.4Γ; 3—54.7Γ; 4—55Γ.

average was taken over a Lorentzian distribution of the incident radiation. It was also assumed that the incident radiation and the scattered radiation were unpolarized.

We assumed that the γ transition $-1/2 \to -3/2$ was excited selectively. Two cases were selected for the numerical calculations: (1) $\Delta = \omega_{-3/2-1/2}$; (2) $\Delta = a_{-3/2-1/2}$. Analysis of the time evolution shows that it is essentially the same for cases (1) and (2) up to $1/\tau \simeq 17\Gamma$, since in this interval the changes in the average fluctuation frequency are $a_{-3/2-1/2} \simeq \omega_{-3/2-1/2}$. Beginning at $1/\tau \simeq 0.1\Gamma$, oscillations with a frequency $\Omega_1 = 2\omega_{-3/2-1/2}$ appear on the time evolution of the scattered radiation. The reason for this result is that the Fe^{57} nucleus undergoes a transition from a superposition state with $m_g = \pm 1/2$ to a superposition state with $m_e = \pm 3/2$, because of fluctuations in the direction of the magnetic moment of the superparamagnetic particle during the absorption of the γ ray. These fluctuations are most obvious at $1/\tau \simeq \Gamma$; with a further increase in $1/\tau$, they are distorted by the increase in the observed decay rate of the excited nuclear state (Fig. 1).

At $1/\tau \geqslant |\omega_{1/2-1/2}| = 8.6\Gamma$ the decrease in the hyperfine field at the nucleus causes the frequency of the γ transition $-1/2 \to 1/2$ to become equal to the frequency of the γ transition in the absence of a field. For this reason, oscillations with a frequency $\Omega_2 = \omega_{-3/2-1/2}$ can appear on the curves of the time evolution. These oscillations are insignificant at $8.6\Gamma \leqslant 1/\tau \simeq 10\Gamma$, since they stem from the nonresonant nature of the excitation and the large width of the γ-transition line. With a further increase in $1/\tau$, the linewidth decreases ($\Gamma_1 = \Gamma + 1/\tau - \sqrt{1/\tau^2 - \omega_{1/2-1/2}^2}$), and the oscillations at the frequency Ω_2 become comparable in magnitude to those at the frequency Ω_1. At $1/\tau \simeq 17\Gamma$ they become predominant (Fig. 2). At $1/\tau > 17\Gamma$, the frequency Ω_2 does not change in case (1). In case (2), the frequency $\Omega_2 = a_{-3/2-1/2}$ gradually decreases. After the hyperfine structure of the Fe^{57} nucleus has completely disappeared ($1/\tau \gg \omega_{-3/2-1/2}$), we find the ordinary time evolution in the absence of fluctuation processes (Fig. 3).

Note that the results of these calculations and their analysis are valid for superparamagnets with a uniform size distribution of the microparticles. A method for preparing such particles is described in Ref. 9.

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