Mössbauer spectra of rf-modulated magnetic stochastic bistable systems

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The behavior of a superparamagnetic particle in an rf field is examined theoretically. Calculations predict the appearance of satellites in the Mössbauer spectrum of such a system. These satellites are interpreted on the basis of the concept of a stochastic resonance.

An "easy-axis" ferromagnetic superparamagnetic particle is essentially a stochastic bistable system. Its behavior in an rf field is of interest as a qualitatively new mechanism for dynamic magnetization reversal of such systems (finely disperse magnetic materials) in their bistability regime. In addition, a superparamagnetic particle is a particular model which obeys the conditions of a stochastic resonance under these conditions.[1] In this letter we calculate the Mössbauer spectrum of a superparamagnetic particle in an rf field. The shape of the Mössbauer spectrum reflects changes in the stochastic dynamics of the magnetic moments of this particle which stem from a coherent external perturbation.

The Mössbauer absorption spectra are represented by the cross section

$$\sigma(\omega_\gamma) \sim \mathrm{Re} \sum_{MM'mm'} \langle m' | H_\gamma^+ | M' \rangle U_{M'm'Mm}(p) \langle M | H_\gamma | m \rangle . \qquad (1)$$

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The evolution superoperator $U(t)$, whose Laplace transform appears in (1), obeys the equation[2] $dU(t)/dt = iL(\theta)U(t) + PU(t)$. Here P is a Fokker-Planck operator. This equation is written for a superparamagnetic particle whose free energy is, when we allow for the interaction with the rf field, $K\cos^2\theta - H_1 M_s v\cos(\omega t + \varphi)$, where K is the anisotropy constant, H_1 is the field amplitude, v is the volume, M_s is the magnetization of the particle, and $L(\theta)$ is the Liouville superoperator, which depends on the random variable θ (the polar angle of the magnetization). We are assuming an averaging over the fast precession around the easy axis.[2] In the model of discrete orientations,[3] the latter expression can be reduced to a control equation

$$\frac{dU(t)}{dt} = iFU(t) + W(t)U(t), \tag{2}$$

where $W(t)$ is the matrix of modulated rates of Kramers transitions between discrete orientations[4] $(\theta = 0,\pi)$,

$$C_{kk'}(t) = C_0 \exp\left[-\frac{v(K - M_s H_1 \cos(\omega t + \varphi))}{k_B T}\right], \tag{3}$$

and the matrices F and $W(t)$ are given by

$$W(t) = \sum_{n=0,\pm 1,\pm 2} W_n \exp(in(\omega t + \varphi)),$$
$$W_n = I_n(\delta) C_0 \exp\left(-\frac{vK}{k_B T}\right) \begin{vmatrix} -(-1)^n E & E \\ (-1)^n E & -E \end{vmatrix}, \quad F = \begin{vmatrix} L(\theta = 0) & 0 \\ 0 & L(\theta = \pi) \end{vmatrix}. \tag{4}$$

Here $\delta = H_1 M_s v/(k_B T)$, and E is a unit matrix which has the same dimensionality as $L(\theta)$.

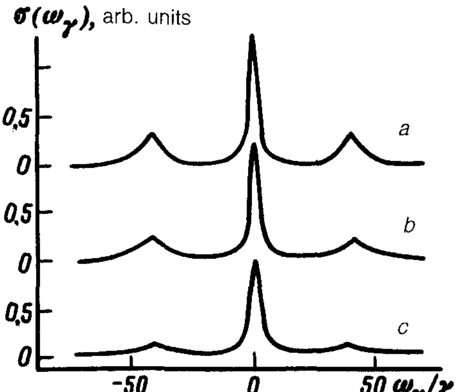

FIG. 1. The absorption cross section in (1), for the absorption of unpolarized γ radiation by ^{57}Fe nuclei, versus H_1. The easy axis of the superparamagnetic particle is directed parallel to the observation direction. $\alpha_0 = 3\times 10^7\ s^{-1}$, $\omega = 40$ MHz. *a*—$H_1 M_s/K = 0.1$; *b*—0.2; *c*—0.3.

For rf fields of small amplitude ($\delta < 1$), the Laplace transform of Eq. (2) becomes

$$(pE' - iF - W_0)U(p) = E' + \sum_{n=\pm 1} W_n U(p - in\omega), \tag{5}$$

where E' is a unit matrix of dimensionality $U(p)$. We will solve this equation by the method of chain fractions,[5] under the assumption that ω is large. Taking an average of $U(p)$ over the random discrete variable, and substituting the result into (1), we find the part of the Mössbauer spectrum corresponding to a γ transition Mm. For brevity, we write the expression derived in the limit of large W_0:

$$\sigma^{Mm}(\omega_\gamma) \sim \left(1 - \frac{2L^2 I_1^2(\delta)/I_0^2(\delta)}{\omega'^2}\right) \mathrm{Re}\left(\frac{1}{p}\right) + \frac{L^2 I_1^2(\delta)/I_0^2(\delta)}{\omega'^2} \mathrm{Re}\left(\frac{1}{p - i\omega'} + \frac{1}{p + i\omega'}\right),$$

$$\omega'^2 \doteq \omega^2 + \frac{2L^2 I_1^2(\delta)}{I_0^2(\delta)}, \qquad p = -i\omega_\gamma + \gamma/2, \quad L = L_{MmMm}. \tag{6}$$

It can be seen from (6) that the spectrum contains a central line, which is the result of a collapse, but this central line is accompanied by satellites at a distance $\pm\omega' \sim \pm\omega$ from the center. These satellites appear as a result of coherent processes which are induced in the stochastic bistable system by oscillations in the populations of the two discrete orientations of the magnetization. We have developed a method for calculating the Mössbauer spectra in the general case (for arbitrary δ). This new method is based on a numerical integration of the evolution equations[6] for periodic perturbations of the system. Figures 1 and 2 show Mössbauer spectra calculated by this method for $\alpha_0 = C_0 \exp(-vK/k_B T) \sim 3\times10^7\ \mathrm{s}^{-1}$. We see that even at $\delta > 1$ the Mössbauer spectra contains satellites, whose intensity increases with increasing field amplitude (with increasing δ; Fig. 1) and with decreasing field frequency (Fig. 2). This behavior can be explained by following the dynamics of the populations of the minima of the superparamagnetic particle (Fig. 3).

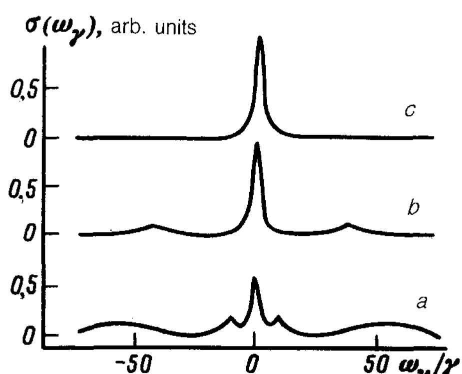

FIG. 2. The absorption cross section in (1) versus ω. $\alpha_0 = 3\times10^7\ \mathrm{s}^{-1}$, $H_1 M_s/K = 0.1$. a—$\omega = 10$ MHz; b—40 MHz; c—70 MHz.

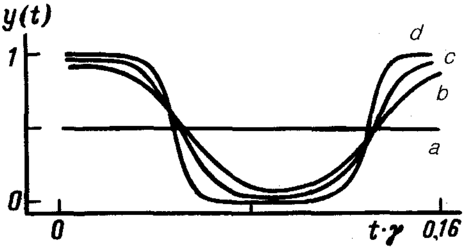

FIG. 3. Time evolution of the population, $y_1(t) = 1 - y_2(t)$, of one of the energy minima of the superparamagnetic particle. $\alpha_0 = 3\times10^7\ s^{-1}$, $\omega = 40$ MHz. *a*—$H_1M_s/K = 0$; *b*—0.1; *c*—0.2; *d*—0.3.

The results (Figs. 1–3) can be interpreted as confirmation of the concept of a stochastic resonance (see Ref. 1, where the amplitude and frequency dependence of the intensity of the output signal in a modulated bistable system are given).

What are the experimental possibilities for observing these results? The parameter values corresponding to Figs. 1–3 correspond to the assumption $\delta > 1$. Using the explicit expression[4] for $C_0(K,M_s)$, we thus find

$$H_1 > \frac{K}{M_s}\left[\ln\left(\frac{2Kg\alpha}{\sqrt{2\pi}M_s(1+\alpha^2)\alpha_0}\right)\right]^{-1},$$

where g is the gyromagnetic ratio, and $\alpha(\sim 1)$ is the dissipation coefficient in the Landau-Lifshitz equation. With $\alpha_0 = 3\times10^7\ s^{-1}$ and values of K and M_s typical of iron, we find $H_1 \sim 5\times10^{-3}\ T$.

The condition of a high potential barrier becomes

$$\frac{Kv}{k_B T} = \left[\ln\left(\frac{2Kg\alpha}{\sqrt{2\pi}M_s(1+\alpha^2)\alpha_0}\right)\right] > 1.$$

From this condition we can find the size of the superparamagnetic particle. For iron at $T = 300$ K, for example, we find $v \sim 1000\ nm^3$.

The transformation to control equation (2) with a modulated Kramers-transition matrix presupposes satisfaction of the condition for a quasiadiabatic behavior,[4] $\omega \ll \omega_l$, where ω_l is the rate of the local relaxation of the magnetization near a minimum. For iron we have $\omega_l \sim 10^{10}\ s^{-1}$, so the values of ω used in the calculations satisfy this condition.

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