TWO-PULSE SPIN ECHO IN TWO-LEVEL SYSTEMS INSIDE AMORPHOUS FERROMAGNETS

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The dependence of Mims echo on time, caused by the existence of two-level systems within 180° Bloch domain walls has been considered. It is shown that in the case of two-level systems, the two-pulse echo formation is characterized by some specific features due to the wide distribution of the two-level systems splitting energy.

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Amorphous systems with tunneling two-level systems (TLS) have been studied with great intensity [1]. The model of TLS was first developed for spin glasses, then for hydrogenous metals [2,3] and solid solutions [4,5]. Amorphous ferromagnetic materials with TLS were considered in [6,7].

Most of the materials as a rule are strongly inhomogeneous by a number of characteristics. In this connection, the development of various methods for investigation and control of inhomogeneities is of great interest.

The magnetic resonance is one of such methods, since some characteristics of spins are sensitive to inhomogeneities. Therefore, the method of spin echo the peculiarity of which in magnets is the enhancement factor of the domain walls [7–9], is widely used. In [10] the rotational echo was studied for amorphous ferromagnets without any consideration of domain walls. The spin echo caused by two-level systems was studied in [11–13].

The goal of this work is to study a two-pulse echo in amorphous ferromagnets in case of wide angles of rotation—the so called "Mims echo", caused by pseudo-spins of TLS located within 180° Bloch walls [14]. According to [6], we consider the case when magnetic atoms form TLS. Because of the strong anisotropy in domain walls the authors assume that the dipole field with account of the enhancement effect differently changes the TLS frequency for TLS located in walls and in domains:

 $\omega_{nw} = \omega_{nd} + \alpha \,,$

where $\omega_{nw} = \frac{E_{nw}}{\hbar}$ is the frequency of TLS in domain walls, $\omega_{nd} = \frac{E_{nd}}{\hbar}$ is the frequency in domains, E_{nw} is the splitting energy of the TLS located in domain walls, E_{nd} is the splitting energy of the TLS located in domains and \hbar is the reduced Planck constant. The value α must

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be limited by the condition $\gamma_n \eta_0 H_1 > |\omega_{nd} - \omega_{nw}| = \alpha$, which guarantees the TLS excitement along the whole Bloch walls. Here H_1 is the amplitude of radio frequency field, γ_n is the gyromagnetic ratio for nuclei, η_0 is the value of the enhancement coefficient $\eta(y)$ in the center of 180° Bloch wall. Let us explain our assumption briefly. As it was already mentioned, we consider the case when TLS atoms are magnetic. Therefore, the matter of particular interest for us is the dipole-dipole interaction between the spins of these atoms and the electron magnetic moments forming the ferromagnetic structure of the material (domains and Bloch walls). The fluctuation of the dipole-dipole interaction constant caused by atom transition between TLS states allows TLS pseudo-spin to experience the action of external magnetic field (static or variable). Let us consider that $\vec{M}_i = \gamma_s \vec{s}_i$ is the magnetic moment of the atom located in *i*-th TLS (γ_s is the gyromagnetic ratio and \vec{s}_i is the spin operator for electrons). The dipole-dipole interaction $H_{dd} = A\left(\frac{1}{r_{ij}^3}\right) \vec{M}_i \vec{M}_j$ between the magnetic moment \vec{M}_i and electronic magnetic moments located in domain wall changes due to the fluctuation of the inter-atomic distance r_{ij} caused by tunneling of *i*-th atoms from one state to another

$$H_{dd} = \begin{pmatrix} A\left(\frac{1}{\left(r_{ij} + \frac{d}{2}\right)^3}\right) & 0\\ 0 & A\left(\frac{1}{\left(r_{ij} - \frac{d}{2}\right)^3}\right) \end{pmatrix} \vec{M}_i \vec{M}_j \, .$$

where d is the distance between the states of *i*-th TLS, A is the dipole-dipole interaction constant. Expression for H_{dd} can be expanded in series in terms of the small parameter $\frac{d}{r_{ij}} \sim 0.1$. Taking into account the linear term of this expansion we obtain the following expression for the frequency of *i*-th TLS

$$\omega_i^{'} = \frac{E_i^{'}}{\hbar} = \left(E_i/\hbar + \frac{3d}{\overline{r}}\sum_j A_{ij}\vec{M}_i\vec{M}_j\right) d_i^z,$$

where d_i^z is the pseudo-spin operator of TLS, E is the splitting energy. When the line width of TLS caused by Klauder-Anderson mechanism is $\Delta \omega \sim 10^6$ Hz, the magnitude of $\frac{3d}{\overline{\tau}} \overline{A} \gamma_s^2 \sim 0.3 \cdot 10^9$ Hz shows that the given mechanism is effective. External magnetic field influences the TLS frequency by means of the interaction with magnetic moments of electron spins. (The direct influence of the external magnetic field on the magnetic moments of *i*-th atom, presented in the *i*-th TLS, does not affect the frequency of this *i*-th TLS. This influence on frequency becomes apparent in other TLS, when the magnetic moments of the given *i*-th TLS play the role of external electron moments.) Let us consider the influence of external variable field on the frequency of two-level system in detail. The variable field changes the orientation of δM electron magnetic moments. These moments take the orientation along the effective field

$$H = H_0 + h(t)$$

where H_0 is the static magnetic field and h(t) is the variable field.

We can qualitatively estimate the change of the TLS frequency caused by this effect. Let us suppose that the variable field is in resonance with one of the packets of magnetic moments with equal quasi-Zeeman frequencies included in the sum $A_{ij}M_iM_j$. As the frequencies of M_i and M_j differ from each other the variable field can not be in resonance with both packets. Neglecting the influence on the non-resonant moments, we can see that the above-mentioned effect is linear to δM

$$\delta \omega^{'} = \frac{3d}{\overline{r}} \overline{A} M \delta M \,.$$

Let us imagine that δM has the following form $\delta M = \lambda h(t)$, where λ is the magnetic susceptibility of the electron system (the tensor of magnetic susceptibility $\lambda^{\alpha\beta}$ like the tensor of enhancement $\eta^{\alpha\beta} \approx \lambda^{\alpha\beta}$ has only one component, which is nonzero for 180° Bloch strip domain structure). We compare two kinds of changes when the radio-frequency field is applied. The first change of TLS frequencies $\delta \omega'_w$ is caused by electron spins, existing in domain walls, and the second one $\delta \omega'_{\alpha}$ is the change caused by the spins in domains. As $\lambda_w > \lambda_{\alpha}$, where λ_w and λ_{α} are the magnetic susceptibilities for walls and domains respectively,

$$\frac{\delta \omega_w'}{\delta \omega_\alpha'} \sim \frac{\lambda_w}{\lambda_\alpha} \sim 10^3,$$

we can conclude that the interaction of TLS with domain walls will prevail over the interaction with domains.

For calculation of echo amplitude from the TLS pseudo-spin located in domain walls, we use the method offered in [14]

$$|\Delta M(\omega, t)| = \left| \int_{-\delta\omega/2}^{\delta\omega/2} \mathrm{d}(\Delta\omega) g_w(\Delta\omega) \eta(\Delta\omega) m^+(\Delta\omega, t) \right|,\tag{1}$$

here $\Delta \omega = \omega_n - \omega$ is the detuning of radio-frequency field relative to the centre of passband of the receiver ω_n with the width $\delta \omega$; $g_w(\Delta \omega)$ is the function which takes into account the variations of the TLS frequency in the walls, and for 180° Bloch walls has form:

$$g_w(\Delta\omega) = \left(2D\frac{\mathrm{d}\omega_n(y)}{\mathrm{d}y}\right)^{-1} = \left[4(\Delta\omega_d - \Delta\omega_w)^{-1/2}(\Delta\omega_d - \Delta\omega)(\Delta\omega - \Delta\omega_w)^{1/2}\right]^{-1}$$

 $\Delta\omega_d = \omega_{nd} - \omega; \ \Delta\omega_w = \omega_{nw} - \omega, D$ is the width of the walls, $\eta(\Delta\omega) = \eta_0 [(\Delta\omega_d - \Delta\omega)/(\Delta\omega_d - \Delta\omega_w)]^{1/2}$ is the enhancement coefficient. We neglect the dispersion of η_0 for the centers of the walls.

Amplitude $m^+(\Delta\omega, t)$ after the action of two pulses is described by [15]

$$m^+(\Delta\omega, t) = m_0 \cdot 2\alpha_1^* \beta_1^* \beta_2^2 \exp(-i\Delta\omega(t - 2\tau_{12}))$$

where τ_{12} is the time interval between the pulses, t is the time measured from the first pulse, m_0 is the equilibrium magnetization, α_1 , β_1 and β_2 are the Kelly-Klein parameters, expressed by the pulse characteristics [15–16]:

$$\begin{aligned} \alpha_1 &= \cos\left[\Omega_1(\Delta\omega)t_1/2\right] + i\cos\left[\theta_1(\Delta\omega)\right]\sin\left[\Omega_1(\Delta\omega)t_1/2\right],\\ \beta_1 &= \sin\left[\theta_1(\Delta\omega)\right]\sin\left[\Omega_1(\Delta\omega)t_1/2\right],\\ \beta_2 &= \sin\left[\theta_2(\Delta\omega)\right]\sin\left[\Omega_2(\Delta\omega)t_2/2\right],\\ \Omega_{1,2}(\Delta\omega) &= \left[\gamma_n^2\eta^2(\Delta\omega)(h^{(1,2)})^2 + \Delta\omega^2\right]^{1/2},\\ tn[\theta_{1,2}(\Delta\omega)] &= \gamma_n h^{(1,2)}\eta(\Delta\omega)/\Delta\omega. \end{aligned}$$

Here $h^{(1,2)}$ is the radio-frequency field amplitude, t_1 and t_2 denote the duration of radio-frequency field.

When we have wide angles of rotation $(\Omega_{1,2}t_{1,2} >> 2\pi)$ for the same radio-frequency pulses, the integration by $\Delta \omega$ removes fast oscillating factors $\sin \Omega t$ and $\cos \Omega t$ down to zero. Therefore, the formula (1) takes the simplest form:

$$|\Delta M(\omega,t)| = \frac{3m_0}{16} \left| \int_{-\delta\omega/2}^{\omega/2} d(\Delta\omega)(\omega_{nd} - \omega_{nw})^{-1/2} (\Delta\omega_d - \Delta\omega)^{-1} (\Delta\omega - \Delta\omega_w)^{-1/2} \cdot \Delta\omega \eta^4 (\Delta\omega)(\gamma_n h)^3 [\Delta\omega^2 + (\gamma_n \eta(\Delta\omega)h)^2]^2 \exp\left[-i\Delta\omega(t - 2\tau_{12})\right] \right|.$$
(2)

The peculiarity of TLS is the dispersion of splitting energy E requiring the obtained expression to be averaged by distribution function. The further simplification of (2) also depends on E. When $E/\hbar \sim \omega_n$ where ω_n is the carrier frequency of the receiver, $\delta\omega >> \gamma_n h\eta(\Delta\omega)$ is valid $\delta\omega$ is the TLS line shape width caused by Klauder-Anderson mechanism [17], equals to the passband width of the receiver). In this case the expression

$$\Delta\omega\exp\left[-i\Delta\omega(t-2\tau_{1,2})\right]\div\left[\Delta\omega^2+(\gamma_nh\eta(\Delta\omega))^2\right]^2$$

is the fast-oscillating factor under the integral (2). Therefore, for $\Delta \omega = 0$ other factors can be removed from under the sign of the integral and thereafter the limits of integration tend to infinity. As a result, taking into account the averaging by the TLS distribution function, we have

$$\bar{p} \int_{E=0}^{E_{\max}} \int_{\Delta_0=0}^{E} \frac{E dE d\Delta_0}{\Delta_0 \sqrt{E^2 - \Delta_0^2}},$$

here \bar{p} is the TLS state density and Δ_0 is the TLS tunneling parameter. For the resonant TLS, the energy of which is within the interval $\hbar\omega_n - \hbar\delta\omega < E < \hbar\omega_n + \hbar\delta\omega$, we have

$$|M(\omega,t)| \sim \bar{p} \int_{\hbar\omega_n - \hbar\delta\omega}^{\hbar\omega_n + \hbar\delta\omega} \int_{\Delta_0 = 0}^{E} \sqrt{\left|\frac{E/\hbar - \omega_n}{E/\hbar - \omega}\right|} |t - 2\tau_{12}| \exp\left[-\frac{(|E/\hbar - \omega_n|)^{1/2}}{|t - 2\tau_{12}|}\right] \frac{E \mathrm{d} E \mathrm{d} \Delta_0}{\Delta_0 \sqrt{E^2 - \Delta_0^2}}$$
(3)



In the opposite case $\delta \omega \ll \gamma_n \eta(\Delta \omega)h$ we can neglect $\Delta \omega$ with respect to $\gamma_n \eta(\Delta \omega)h$ in the integral expression (2). Finally we have:

$$|M(\omega,t)| = \bar{p} \int_{E=0}^{\hbar\omega_n - \hbar\delta\omega} \int_{\Delta_0=0}^{E} \frac{EdEd\Delta_0 \left| \sin\frac{\delta\omega(t-2\tau_{12})}{2} - \frac{\delta\omega(t-2\tau_{12})}{2} \cos\frac{\delta\omega(t-2\tau_{12})}{2} \right|}{\sqrt{E^2 - \Delta_0^2} \left| 1 - (E/\hbar - \omega) \right| \sqrt{|E/\hbar - \omega|} \left(\frac{\delta\omega(t-2\tau_{12})}{2} \right)^2 + \frac{\bar{p}}{\hbar\omega_n + \hbar\delta\omega} \int_{\Delta_0=0}^{E} \frac{EdEd\Delta_0 \left| \sin\frac{\delta\omega(t-2\tau_{12})}{2} - \frac{\delta\omega(t-2\tau_{12})}{2} \cos\frac{\delta\omega(t-2\tau_{12})}{2} \right|}{\Delta_0 \sqrt{E^2 - \Delta_0^2} \left| 1 - (E/\hbar - \omega) \right| \sqrt{|E/\hbar - \omega|} \left(\frac{\delta\omega(t-2\tau_{12})}{2} \right)^2$$
(3")

The expression for echo amplitude (3") can not be solved analytically and needs the use of numerical methods. The echo amplitude dependence on time is shown in Fig. 1. As the graphic shows, the echo amplitude has a split summit with two maxima that are set in time shifted from $2\tau_{12}$ that is completely consistent with the Mims [11] theory. The graphic is plotted by the numerical integration for the following parameters: $\tau_{12} \sim 10^{-6}$ s, $h \sim 1$ A/m, $\eta_0 \sim 10^6$, $\gamma_n/2\pi \sim 1$ Hz·m/A, $\omega \sim 10^6$ Hz, $\delta\omega/2\pi \sim 10^5$ Hz, $T \sim 0.1$ K, $E_{\rm max} \sim 10$ K, $\omega_n \sim 10^{10}$ Hz.

As it was already mentioned, the Mims echo in amorphous magnets must be specific due to the wide dispersion of TLS frequencies. It is also well known that Mims echo from nuclear spins depends significantly on detuning (i.e. whether the influence is resonant or not with respect to spins) and has a sharp maximum if the influence is resonant.

In our case the TLS, for which the pulse frequency is resonant as well as the TLS the frequency of which differs from the pulse frequency take part in echo formation (see formula (3")). Since there is no other principal difference, the authors think that the fact that echo signal amplitude formed by TLS is less-dependent on pulse frequency (see Fig. 2) could be explained by the above-described peculiarities.



Fig. 2. Echo amplitude dependence on rf field frequency. The solid line corresponds to nuclear spins is taken from [14]. The dashed line corresponds to TLS.

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