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

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In the case of applied distorted pulse, the single pulse echo in amorphous magnets, caused by the tunneling two-level systems located inside 180° Bloch walls has been studied. It is shown that in case of TLS the single-pulse echo formation is characterized by some specificity due to the wide distribution of the TLS splitting energy.

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This paper is devoted to the presentation of a theoretical model of the single-pulse spin echo stemming from two-level systems (TLS) inside 180° Bloch domain walls in an amorphous ferromagnets.

Amorphous systems with tunneling two-level systems TLS are widely studied [1]. The TLS model was first developed for spin glasses and then for hydrogenous metals and solid solutions [2-5]. Amorphous ferromagnetic materials with TLS are considered in [6,7]. Excitations observed in experiments can be described by TLS [1]. According to the TLS model, it is assumed that TLS are formed by atoms or atom groups which can occupy two equivalent positions with almost equal probability in a double asymmetric well with splitting energy $E = \sqrt{\Delta_0^2 + \Delta^2}$ (Δ is the asymmetry parameter and Δ_0 is the TLS tunneling energy). The spin-echo method is very effective for TLS investigations. It allows establishing the relation between the experiment and the theory [8]. Polarized echo, caused by TLS in spin glasses, was investigated in [8] and in amorphous ferromagnetic in [9]. It is assumed that by "pulse methods" the two-pulse echo is meant. Recently, a single-pulse echo the mechanism of formation of which is not simple has provoked an interest [10]. This article deals with the investigation of single-pulse echo in amorphous ferromagnets (caused by TLS located inside domain walls) within the frames of the distorted pulse model. Atoms forming the TLS can be magnetic or non-magnetic. If atoms are magnetic they will be connected with the modulation of dipole-dipole, exchange and anisotropy energies. The dipole-dipole interaction between the magnetic moments of the atoms forming TLS and the electronic magnetic moments forming the ferromagnetic structure of the material (domains and 180° Bloch walls) is of great interest. The fluctuations of dipole-dipole interaction constant caused by the atom transition between TLS states allow TLS pseudo-spin to experience

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the action of external magnetic field (static or variable) through the influence of this field on magnetic moments of electronic spins located in the domain walls. To explain the above-mentioned mechanism we can apply the method used in [11]. Let us assume that $\vec{M_i} = \gamma_s \vec{S_i}$ is the magnetic moment of the atom located in the *i*-th TLS, where γ_s is the gyromagnetic ratio for the electrons, $\vec{S_i}$ is the electron spin operator. The dipole-dipole interaction $H_{dd} = A(1/r_{ij}^3)\vec{M_i}\vec{M_j}$ between magnetic moment $\vec{M_i}$ and electronic magnetic moments located in domain wall varies due to the fluctuation of the interatomic distance r_{ij} (as a result the tunneling of *i*-th atom from one state to another)

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

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 a series over $d/r_{ij} \approx 0.1$. Retaining the linear term of this expansion we obtain the following expression for the frequency of *i*-th TLS

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

where d_i^z is the pseudo-spin operator of TLS, \hbar is the Planck constant, E is the TLS splitting energy, ω_i' and E_i' are the TLS frequency and splitting energy with taking into account fluctuations of dipole-dipole interaction constant. When the line width of TLS caused by Klauder-Anderson mechanism equals $\Delta \omega \sim 10^6$ Hz the magnitude of $(3d/\bar{r})\bar{A}\gamma_s^2 \sim 0.3 \cdot 10^9$ Hz shows that the given mechanism is effective. The influence of external magnetic field on the TLS frequency is not direct. The influence of the external magnetic field on the magnetic moment of the *i*-th atom presented in the *i*-th TLS does not affect directly the frequency of this *i*-th TLS. This influence is displayed on other TLS, when the magnetic moment of the given *i*-th atom plays the role of external electronic moment. Let us consider the influence of external variable field on the frequency of a two-level system in more detail. The application of a variable field causes the change of δM electric magnetic moments orientation. These moments take the orientation of the effective field $H = H_0 + h(t)$, where H_0 is the static magnetic field and h(t) is the variable magnetic 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 the magnetic moments with equal quasi-Zeeman frequencies, included in the sum $A_{ij}M_iM_j$. As M_i and M_j differ from each other the variable field cannot be in resonance with both packets. Neglecting variable field influence on the nonresonant moments, we can see that above-mentioned effect must be linear in δM

$$\delta\omega' = \frac{3d}{\bar{r}}\bar{A}M\delta M,$$

where $\delta \omega'$ is the TLS frequency variation, \bar{A} is the average value of the dipole-dipole interaction constant \bar{r} is the average value of the interatomic distance. Let us imagine that δM has the following form: $\delta M = \lambda h(t)$, where λ is the magnetic susceptibility of electric system (the tensor of magnetic susceptibility $\lambda^{\alpha\beta}$, as well as the tensor of enhancement $\eta^{\alpha\beta} \approx \lambda^{\alpha\beta}$, have only one component, which is not zero [12] in the 180° Bloch strip domain structure). We compare two kinds of changes when the radio-frequency field is applied. The first change of TLS frequencies is caused by electron spins existing in domain walls, and the second one is caused by spins in domains. As $\lambda_w > \lambda_\alpha$, where λ_w and λ_α are the magnetic susceptibilities for walls and domains [12], we can conclude that the interaction of TLS with domain walls prevails over the interaction with domains:

$$\frac{\delta \omega'_w}{\delta \omega'_\alpha} \sim \frac{\lambda_w}{\lambda_\alpha} \sim 10^3,$$

where $\delta \omega'_{w}$ is the change of TLS frequency caused by domain walls and $\delta \omega'_{\alpha}$ is the change caused by domains. To obtain the dependence of echo amplitude on time, we use the method described in [13] and the method used for studying the nuclear single-pulse echo within the frame of the distorted pulse model [14]. In contrast to the single-pulse echo caused by nuclear spins, the single-pulse echo caused by TLS shows the wide distribution of the TLS parameters and a constant TLS state density. As it is known, the influence of pulse with the phase changed at the beginning and at the end of the pulse is equivalent to the influence of distorted pulse in the rotating system

$$h^{+} = h \begin{cases} \exp(i\varphi_{e}) & 0 < t' < \tau_{e} \\ \exp(i\varphi) & \tau_{e} < t' < \tau_{e} + \tau \\ \exp(i\varphi_{t}) & \tau_{e} + \tau < t' < \tau_{e} + \tau + \tau_{t} \end{cases}$$

where $h^+ = h_x + ih_y$, τ_e and τ_t are the time intervals of distorted parts, τ is the interval of non-distorted part, φ is the phase of the part of non-distorted pulse, and h is the amplitude of radio frequency (rf.) field.

Let us write the Bloch equations for pseudo-spins of TLS. Taking into account the interaction with the domain walls and introducing the notations $s = \mu_+/\mu_0$, $m = \mu_z/\mu_0$, $\delta = (E/h) - \omega$, $q = \gamma h^+$ we obtain:

$$\begin{split} \dot{s} + i\,\delta\,s &= iqm,\\ \dot{s}^* - i\,\delta\,s &= -iqm,\\ \dot{m} &= 1/2iq(s-s^*) \end{split}$$

where μ_0 is the equilibrium value of TLS state polarization [8], μ_+ and μ_z are the longitudinal and transverse components, ω is the frequency of the rf. field, E is the TLS splitting energy, the star (*) denotes the complex conjugation and $\gamma = (3d/r)\bar{A}\gamma_s\lambda_w$ is the constant describing the influence of variable field on TLS by means of electric magnetic moments. This constant is of the order of $\gamma \sim 10^3$ Hz M/A.

Using the Laplace transform

$$\hat{s} = \hat{s}(p) = \int_{0}^{\infty} S(t) \mathrm{e}^{-pt} \mathrm{d}t$$

and also the transform for m, Bloch's differential equations take algebraic form:

$$\begin{array}{l} (p+i\delta)\hat{s} - iq\hat{m} = s_{0}, \\ (p-i\delta)\hat{s}^{*} + iq\hat{m} = s_{0}^{*}, \\ p\hat{m} - \frac{i}{2}q(\hat{s} - \hat{s}^{*}) = m_{0}. \end{array}$$

Taking into account the first equation, we can express \hat{s} , \hat{s}^* by means of \hat{m} , then introducing the obtained expression in the third equation we obtain:

$$\hat{m} = \frac{m_0(p^2 + \delta^2) + \frac{i}{2}q[s_0(p - i\delta) - s_0^*(p + i\delta)]}{p(p^2 + \delta^2 + q^2)}.$$

Making the inverse Laplace transform

$$m(t) = \frac{1}{2i\pi} \int_{x-i\infty}^{x+i\infty} \hat{m}(p) e^{pt} dp$$

and taking into account that $\hat{s} = \frac{s_0 + iq\hat{m}}{p + i\delta}$, for the transverse component of the magnetization we obtain by solving the Bloch's equations the following:

$$\mu^{+}(t, E, \eta) = \mu_{0} \left[A_{1}^{e} \left(A_{2} A_{2}^{t} + A_{3} A_{1}^{t} + A_{4}^{*} A_{4}^{t} \right) + A_{1}^{*e} \left(A_{4} A_{2}^{e} + A_{2}^{*} A_{4}^{t} + A_{3}^{*} A_{1}^{t} \right) + A_{5}^{e} \left(A_{1} A_{2}^{t} + A_{5} A_{1}^{t} + A_{1}^{*} A_{4}^{t} \right) \right] \exp(i\delta t),$$

where

$$A_{1} = \frac{\chi}{\varphi} \left(-\frac{2\delta}{\varphi} \sin^{2} \frac{\varphi\tau}{2} + i \sin \varphi\tau \right) \exp(i\varphi),$$

$$A_{2} = \cos\varphi\tau + \left(\frac{\chi}{\varphi}\right)^{2} \sin^{2} \frac{\varphi\tau}{2} + i \frac{\delta}{\varphi} \sin\varphi\tau,$$

$$A_{3} = \frac{A_{1}}{2} \exp(-2i\varphi),$$

$$A_{4} = \left(\frac{\chi}{\varphi}\right)^{2} \sin \frac{\varphi\tau}{2} \exp(2i\varphi),$$

$$A_{5} = 1 - 2\left(\frac{\chi}{\varphi}\right)^{2} \sin^{2} \frac{\varphi\tau}{2},$$

$$\varphi = (\delta^{2} + \chi^{2})^{1/2}, \ \delta = \frac{E}{\hbar} - \omega, \ \chi = \eta \gamma \hbar.$$
(1)

Here η is the enhancement factor for the domain walls, ω is the frequency of the applied pulse, t and l subscripts correspond to $\tau_{l,t}$ and $\varphi_{l,t}$. The effect of domain walls makes it necessary to take into account enlarging the effect using the enhancement factor distribution function [15]

$$F_{\omega}(\eta) = \eta^{-1} \int_{0}^{\infty} \exp\left(-\ln 2 \frac{\left[(\varepsilon^{2} + \eta^{2})^{1/2} - \bar{\eta}_{0}\right]^{2}}{\Delta \eta_{0}^{2}}\right) \mathrm{d}\varepsilon, \qquad (2)$$

where ε , $\bar{\eta}_0$, and $\Delta \eta_0$ are parameters described in [15]. After averaging (1) by the TLS distribution function p ($\Delta_0 E$) [1], and using (2) we obtain:

$$M(t) = \left| \int_{E=0}^{E_{\text{max}}} \int_{\Delta_0=0}^{E} \frac{\bar{p}E \mathrm{d}E \mathrm{d}\Delta_0}{\Delta_0 \sqrt{E^2 - \Delta_0^2}} \int_{0}^{\infty} F_{\omega}(\eta) \eta \mathrm{d}\eta \mu^+(t, E, \eta) \right|,$$
(3)

where \bar{p} is the TLS state density.



Fig. 1. Curve of the free induction decay and echo amplitude as a function of time

To analyze the obtained results we need the numerical estimates. The result of numerical calculations for the parameters $\tau_l = \tau_t = 1 \cdot 10^{-6}$ s, $\gamma \hbar = 10^4$ s⁻¹, $\tau = 8 \cdot 10^{-6}$ s, $\omega \approx 10^6$ Hz, $\varphi_l = \pi/6$, $\varphi_t = \pi/3$, $\varphi = 0$, $\Delta \eta_0 = 100$ [12, 13], is shown in Fig. 1.

A comparison of the single-pulse echo formation in case of TLS and nuclear spins is worthwhile. Since in contrast to nuclear spins the pseudo-spins of the TLS are not directly affected by the variable field, the mechanism of single-pulse echo formation, suggested in this paper, can be considered as original and different from that in [14]. There is another significant difference in the value of δ . For nuclear spins, $\delta = \omega_0 - \omega_n$, where ω_0 is the frequency of nuclear magnetic relaxation (NMR) (the pulse frequency coincides with it), ω_n is the frequency of a specific nucleus (distribution of nuclear frequencies is considered to have the Gaussian form). In our case $\delta = (E/\hbar) - \omega$, where ω is the pulse frequency, E is the TLS splitting energy. At the same time, the limits of E variation are sufficiently large $0.01 \text{ K} \leq E \leq 10 \text{ K}$, and we use the TLS distribution function $P(E, \Delta)$ to average the obtained result (see eq. (3)). Thus, both the resonant and nonresonant TLS take part in echo formation. Since there is no other difference in our opinion, the fact that echo signal formed by TLS (Fig. 1) decays slower in time than the signal formed by nuclear spins (ref. [14]) can be explained by the above-described peculiarities.

By this specific feature in the low-temperature experiments, one can distinguish the subsystem of TLS from the nuclear subsystem.

Because of the lack of published experimental results on the single-pulse spin echo in amorphous magnets, we cannot present experimental data on (Fig. 1) to compare them with the theory. As it was mentioned above, the model of the amorphous ferromagnets was studied in [6], but only theoretically. Then in ref. [9] the transverse relaxation process was studied and the formula for TLS pseudo-spins transverse relaxation time T_2 was obtained. We hope that the present investigation will be useful for experimental determination of relaxation time T_2 and accordingly of the interaction constants between two-level systems calculated in [6]. For experimental determination of relaxation time T_2 we think it reasonable to use the method described in [16]. According to eq. (4.1.2) in [16], for T_2 we have

$$T_2 = -4 \frac{\tau - \tau'}{\ln \mu(\tau) / \ln \mu(\tau')},$$

where $\mu(\tau)$ is the intensity of the echo signal. Using the single-pulse echo signal for different time intervals of non-distorted pulse τ , one can determine T_2 , which, in its turn, gives the value of the interaction between TLS in amorphous ferromagnets.

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