Theory of spin induction and conduction-electron echo in bulky metallic samples

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A theory is developed of the transient processes (spin induction and echo) of the conduction electrons in bulky metallic plates. The effect of a short microwave pulse on the conduction-electron spin system is investigated and attenuation of the transient signals observed under reflection and transmission conditions is calculated. The transmission coefficient of the metal is calculated for the transient processes.

1. We have previously reported[1, 2] observation of spin echo on conduction electrons (SECE) and constructed a theory of this phenomenon for samples whose characteristic dimensions (d) are smaller than the skin-layer depth (δ). Investigations of the stationary paramagnetic resonance of conduction electrons (CE) have shown that in a bulky metal a radical change takes place in the absorption line shape[3], and effects of the selective-transparency type appear[4]. It is therefore of interest to investigate theoretically and SECE experimentally in bulky samples ($d > \delta$), and in particular to investigate the transparency of a metal relative to transient processes.

A transient process of the type of free induction of the conduction electrons in a bulky sample was investigated by Taylor et al.[5] The theoretical description of this phenomenon is based in[5] on the line shape calculated by Dyson for stationary resonance with conduction electrons, the Fourier transform of the line shape being the envelope of the free-precession signal[6]. Kopvillem and Alekseev[8] have calculated the Fourier transform of the conduction-electron stationary resonance line shape in spherical samples. There is no proof that this approach is valid for the description of the damping of transients of the spin-echo type. Moreover, Doktorov and Burshteĭn[9] have shown that in a paramagnet the damping of the induction and echo signals follows different laws in most cases.

We develop below a theory of spin echo in the spin system of the conduction electrons in bulky metal samples.

2. In the calculations of the SECE we shall follow the procedure described in[2, 10]. We consider a sample placed in a constant magnetic field $\mathbf{H}_0(\parallel z)$ and in a microwave field $\mathbf{H}_1(t)$ perpendicular to $\mathbf{H}_0$. The behavior of the magnetization density $\mu(\mathbf{r}, t)$ of electrons will be described with the aid of the Bloch equations. In a coordinate system rotating at the frequency ω of the alternating field, the Bloch equations take the form

$$\begin{aligned} \dot{\mu}_x(\mathbf{r},t) &= -\Delta\omega(\mathbf{r})\mu_y(\mathbf{r},t) - \mu_x(\mathbf{r},t)/T_V, \\ \dot{\mu}_y(\mathbf{r},t) &= \Delta\omega(\mathbf{r})\mu_x(\mathbf{r},t) - \mu_y(\mathbf{r},t)/T_V - \gamma H_1(\mathbf{r})\mu_z(\mathbf{r},t), \\ \dot{\mu}_z(\mathbf{r},t) &= -[\mu_z(\mathbf{r},t)-\mu_0]/T_V + \gamma H_1(\mathbf{r})\mu_y(\mathbf{r},t). \end{aligned} \tag{1}$$

Here $\Delta\omega(\mathbf{r}) = \omega - \gamma H_0 - \gamma\Delta H_0(\mathbf{r})$, $\gamma\Delta H_0(\mathbf{r})$ is the spread of the resonance frequencies, $H_1(\mathbf{r})$ is the amplitude of the alternating field inside the sample and its coordinate dependence is determined by the skin effect, T_V is the time of the conduction-electron spin relaxation in the volume of the metal and $\mu_0 = \chi_0 H_0$ is the equilibrium density of the conduction-electron magnetization.

Owing to the diffusion motion of the conduction electrons, the coordinate-dependent parameters in (1) are random functions of the time. The stochastic character of these parameters will be taken into account by averaging the solutions of the Bloch equations over the trajectories of the conduction electrons. As usual, in the study of transient processes the solutions of (1) are obtained for the time interval $t_i \ll T_V$ during which the microwave field acts, and for the interval between the pulses. Combining these solutions, we can then describe the spin induction and spin echo.

The solutions are averaged by integration, in functional space, of the trajectories of the diffuse motion of the conduction electrons with a Wiener measure $D^n(\mathbf{r})$[11], which is connected by the relation

$$D^n(\mathbf{r}) = \prod_{k=1}^{n} P(\mathbf{r}_k t_k/\mathbf{r}_{k-1} t_{k-1})\, d\mathbf{r}_{k-1}$$

with the conditional probabilities P for the displacement of the conduction electrons from the point $\mathbf{r}_{k-1}$ to the point $\mathbf{r}_k$ within the time interval $t_k - t_{k-1}$, these being the solutions of the diffusion equation.

We consider a metallic plate of thickness d and infinite dimensions in the (xy) plane. From symmetry considerations we can use the solutions of the one-dimensional diffusion equation for the probabilities P. It is obvious that in a bulky sample the relaxation of the conduction-electron spins on the surface should not greatly influence the damping of the transients. We therefore consider the case when the boundary conditions of the diffusion equation correspond to the absence of surface spin scattering[3, 2].

To perform concrete calculations it is necessary to take into account the following additional considerations: The transients constitute the response of the spin system to a certain sequence of microwave pulses, the durations t_i of which are much shorter than the interval τ between the pulses. The theory contains in natural manner two parameters, $L_{t_i} = \sqrt{2Dt_i}$ and $L_\tau = \sqrt{2D\tau}$, which have the meaning of the diffusion displacements of the conduction electrons during the time of action of the pulse and during the interval between the pulses (D is the diffusion coefficient of the conduction electrons). The derivation of the equations for the transients depends significantly on the ratio of the parameters δ, d, L_{t_i} and L_τ, and in our case we always have $d \gg \delta$ and $L_\tau \gg L_{t_i}$. In addition, at the typical values $t_i \sim 10^{-8}$ sec, $\delta \sim 10^{-4}$ cm, and $D \sim 30\ \text{cm}^2/\text{sec}$, the relation $L_{t_i}^2 \gg \delta^2$ is satisfied. When these inequalities are taken into account, the following situations can be realized:

$$\begin{aligned} &\delta^2 \ll L_{t_i}^2 \ll L_\tau^2 \ll d^2, \\ &\delta^2 \ll L_{t_i}^2 \ll d^2 \ll L_\tau^2, \\ &\delta^2 \ll d^2 \ll L_{t_i}^2 \ll L_\tau^2, \end{aligned} \tag{2}$$

and will be considered below[1)].

Assuming that the conduction-electron spin system is at equilibrium prior to the first pulse that excites the transient ($\mu(z, t) = \mu_0 k$), and assuming a classical penetration of the alternating field into the metal ($H_1(t) = H_1(0)e^{-z/\delta}$), we can calculate the magnetization of the conduction electrons after the action of one microwave pulse, and in particular, the component $\mu_y(z, t)$ that generates the observable free-induction signal. In cases (2a) and (2b), it is given by the expression

$$\mu_y(z,t)=\mu_0\gamma h_1 A_1(t_1)\exp\left\{-\frac{t}{T_V}\right\}\sum_{n=-\infty}^{\infty}\cos\frac{\pi n}{d}z\exp\left\{-\frac{\pi^2 D}{d^2}n^2(t-t_0)\right\},$$

$$A_1(t_1)=\int_0^{t_1}\exp\left\{-\frac{\gamma^2 h_1^2 d^2}{D}t_1\right\}dt_1,\quad h_1=\frac{\delta}{d}H_1(0). \tag{3}$$

The calculation of the function $\mu_y(z, t)$, which determines the spin-echo signal after the conduction-electron system is acted upon by two microwave pulses separated by an interval τ, leads in cases (2a) and (2b) to the form

$$\mu_y(z,t)=-\frac{1}{2}\mu_0\gamma^2 h_1 h_2 A_1(t_1)A_2(t_2)\exp\left\{-\frac{t-t_0}{T_V}\right\}$$

$$\times\theta(t-t_0-2\tau)\sum_{n=-\infty}^{\infty}\cos\frac{\pi n}{d}z\exp\left\{-\frac{\pi^2 D}{d^2}n^2(t-t_0-\tau)\right\}$$

$$\times\sum_{n_0=-\infty}^{\infty}\exp\left\{-\frac{\pi^2 D}{d^2}n_0^2(\tau-t_0)\right\}, \tag{4}$$

$$A_2(t_2)=i\int_0^{t_2}\exp\left\{-\frac{\gamma^2 h_2^2 d^2}{D}t\right\}\Phi\left(i\frac{\gamma h_2 d}{\sqrt{D}}\sqrt{t}\right)dt,$$

where $\Phi(x)$ is the error integral, while h_1, t_1 and h_2, t_2 are the amplitudes and durations of the first and second pulses, respectively. In the derivation of (4) we discarded terms describing the free precession signals after each pulse, and averaged over the spread $\gamma\Delta H_0(z)$ of the constant magnetic field, the result of which was the appearance of the factor $\theta(t - t_0 - 2)$, which determined the shape of the echo signal. The form of the function θ depends on the character of the inhomogeneity of the magnetic field and, for example in the case of a normal distribution of the quantity $\gamma\Delta H_0(z)$, we have

$$\theta(t-t_0-2\tau)=\exp\{-(t-t_0-2\tau)^2/T_3^2\},$$

where T_3^2 is the second moment of the distribution.

In case (2c), the conduction electron criss-crosses the sample many times during the time of action of the microwave. From intuitive considerations, which are confirmed by calculation, the conduction electron "feels" in this case an average field

$$h_1=\int_0^d H_1(z)\,dz=\frac{\delta}{d}H_1(0)$$

and the action of the pulses is described by the expressions for dielectrics in a microwave field h_1.

3. Let us consider the main distinguishing features of transient processes in bulky metallic samples. The strong spatial inhomogeneity of the alternating field and the high mobility of the conduction electrons lead to a unique type of damping of the observed signals and to a non-standard dependence of their amplitude on the power and on the duration of the microwave pulses.

A. Action of Microwave Pulse

In a dielectric paramagnet, the free-induction signal depends on the amplitude H_1 of the alternating field and

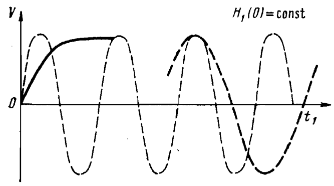

FIG. 1. Induction signal amplitude V at the instant when the field is turned on vs. the pulse duration t_1: solid curve—bulky metallic sample, cases (2a) and (2b); thick dashed curve—bulky metallic sample, case (2c). Light dashed line—dielectric.

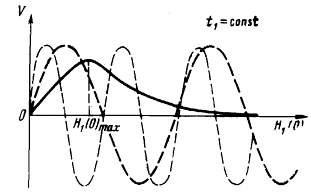

FIG. 2. Induction signal amplitude V at the instant when the field is turned off vs. the microwave field $H_1(0)$ on the surface of the metal: solid curve—bulky metallic sample, cases (2a) and (2b); thick dashed curve—bulky metallic sample, case (2c), thin dashed curve—dielectric.

on the duration t_1 of the microwave pulse like $\sin(\gamma H_1 t_1)$. According to (3), the corresponding expression in a bulky metallic sample is (see cases (2a) and (2b))

$$\gamma h_1 A_1(t_1)=\frac{D}{\gamma h_1 d^2}\left[1-\exp\left\{-\frac{\gamma^2 h_1^2 d^2}{D}t_1\right\}\right]. \tag{5}$$

If the amplitude of the microwave field on the surface of the metal is $H_1(0) \lesssim 10$ Oe, and if the durations t_1 are such that the inequality $L_{t_1}^2 < d^2$ is satisfied, then the argument of the exponential in (5) is small. Expanding the exponential, we represent (5) in the form

$$\gamma h_1 A_1(t_1)\approx\gamma H_1(0)\,\delta t_1/d. \tag{6}$$

Thus, the amplitude of the induction signal turns out to be a linear function of both $H_1(0)$ and t_1.

With increasing t_1, the inequality $L_{t_1}^2 < d^2$ no longer holds and the situation corresponds to the case (2c), in which the signal amplitude is proportional to $\sin(\gamma h_1 t_1)$. This expression should go over into (6) (in the region $L_{t_1}^2 \sim d^2$). The dependence of the induction signal on the duration of the pulse in a bulky metal is shown in Fig. 1 as compared with the standard expression for a dielectric.

The induction signal as a function of the amplitude of the alternating field $H_1(0)$ at a fixed pulse duration is shown in Fig. 2. Expression (5) has a maximum at

$$H_1(0)=H_{1\,max}=[\sqrt{2}\gamma t_1\delta/L_{t_1}]^{-1}.$$

Further increase of the field amplitude on the surface of the metal leads to a decrease of the induction signal, which is described asymptotically by the function $H_1^{-1}(0)$.

The physical reason why the induction-signal amplitude has a maximum in terms of $H_1(0)$ can be explained in the following manner. In a rotating coordinate system, the action of the alternating field reduces to a rotation through the angle $\varphi = \gamma H_1 t_1$ (H_1 is the amplitude of the field and t_1 is the duration of the action of the field). The maximum rotation through the angle $\varphi_0 = \gamma H_1(0)t_1$ is executed by the magnetic moment of a conduction-electron that is permanently located in the skin layer. Most electrons, on the other hand, leave the skin layer and return to it many times during the time t_1, and consequently they experience the action of the field for a shorter time. So long as $H_1(0)$ is smaller than or of the order of H_{1max}, the transverse components of the moments of the individual electrons have the same sign and the induction signal increases with increasing

$H_1(0)$ (see expression (6)). When this field value is exceeded, the projections of the magnetic moments will have opposite signs and will in part cancel each other. This leads to a decrease in the induction signal.

A similar comparison of the echo-signal amplitude on the characteristics of the pulses reveals the same situation. The echo signal in a bulky metal sample is proportional to the quantity $\gamma^2 h_1 h_2 A_1(t_1) A_2(t_2)$, which under the conditions formulated in the derivation of (6) can be regarded as equal to

$$\gamma^2 h_1 h_2 A_1(t_1) A_2(t_2) \approx \frac{4}{3\sqrt{\pi}} \gamma^3 H_1(0) H_2^2(0) t_1 t_2^{3/2} \frac{d}{\sqrt{D}} \left(\frac{\delta}{d}\right)^3. \quad (7)$$

The dependence of the signal on the values of $H_1(0)$ and $H_2(0)$ at fixed durations of the pulses remains qualitatively the same. However, only a numerical calculation H_{2max} is possible in this case. An increase in the pulse duration, just as in the case of the induction, leads to the case of the average field (2c), where the echo signal depends on the parameters of the pulses like $\sin(\gamma h_1 t_1)\sin^2(\gamma h_2 t_2/2)$.

B. Damping of Transients in Metals

It was shown above (sec. 1.2) that owing to the skin effect the magnetization induced by a microwave pulse in a bulky metallic plate is spatially inhomogeneous. To determine the magnitudes of the observed transient signals it is necessary to obtain the magnetization on the surface of the metal, and signals can be produced also on the opposite side of the plate ($z = d$), where no microwave pulses penetrate. The transport of phase memory in the transient processes, just as the transfer of the spin information in the well-investigated stationary spin transparency of metals, is due to the diffusion of the conduction electrons. To describe the transport processes, we introduce the transparency coefficient η, defined as the ratio of the amplitudes of the signals of the transient processes on opposite sides of the plates. According to expressions (3) and (4) we have

$$\eta(t) = \sum_{n=-\infty}^{\infty} \exp\left\{-\frac{\pi^2 D}{d^2} n^2 t\right\} \Big/ \sum_{n=-\infty}^{\infty} (-1)^n \exp\left\{-\frac{\pi^2 D}{d^2} n^2 t\right\}. \quad (8)$$

In samples with dimensions $d^2 \ll L^2$ (case (2b)), after a time τ following the determination of the last pulse, a homogeneous magnetization is established, and the transient signals are determined by the expressions

$$V_{\text{ind}}(0, \tau) = V_{\text{ind}}(d, \tau) = \mu_0 \gamma h_1 A_1(t_1) \exp\{-\tau/T_V\},$$
$$V_{\text{echo}}(0, 2\tau) = V_{\text{echo}}(d, 2\tau) = {}^1/_2 \mu_0 \gamma^2 h_1 h_2 A_1(t_1) A_2(t_2) \exp\{-2\tau/T_V\}. \quad (9)$$

Thus, in the case (2b), the transparency coefficient is equal to unity.

At $L_T^2/d^2 \ll 1$ (case (2a)), the amplitudes of the conduction and echo signals on opposite sides of the plate are greatly different. Accurate to quantities of the order of $\exp\{-d^2/L_T^2\}$, we obtain from (3) and (4)

$$V_{\text{ind}}(0, \tau) = \mu_0 \gamma h_1 A_1(t_1) \sqrt{\frac{d^2}{\pi D}} \frac{1}{\sqrt{\tau}} \exp\left\{-\frac{\tau}{T_V}\right\},$$
$$V_{\text{ind}}(d, \tau) = 2\mu_0 \gamma h_1 A_1(t_1) \sqrt{\frac{d^2}{\pi D}} \frac{1}{\sqrt{\tau}} \exp\left\{-\frac{\tau}{T_V} - \frac{d^2}{4D\tau}\right\},$$
$$V_{\text{echo}}(0, 2\tau) = \frac{1}{2} \mu_0 \gamma^2 h_1 h_2 A_1(t_1) A_2(t_2) \frac{d^2}{\pi D} \frac{1}{\tau} \exp\left\{-\frac{2\tau}{T_V}\right\},$$
$$V_{\text{echo}}(d, 2\tau) = \mu_0 \gamma^2 h_1 h_2 A_1(t_1) A_2(t_2) \frac{d^2}{\pi D} \frac{1}{\tau} \exp\left\{-\frac{2\tau}{T_V} - \frac{d^2}{4D\tau}\right\}. \quad (10)$$

The expression for $V_{\text{ind}}(0, \tau)$ coincides with the result of Taylor et al.[5], which describes well the conduction-electron free-induction signal observed in the reflection regime[5]. Under these conditions, the transparency coefficient is equal to

$$\eta(\tau) = 2 \exp\left\{-\frac{1}{2}\left(\frac{d}{L_\tau}\right)^2\right\} \quad (11)$$

(remember that we are considering the case $\tau \ll d^2/2D$).

As follows from (10), owing to the diffusion of the conduction electrons, the amplitudes of the signals on the opposite side of the plate increase with increasing time, since in-phase electron spins arrive in this case into the region of large z. On the other hand, the relaxation in the interior of the metal decreases the observed signal. It is easily seen that there exists an optimal observation interval τ_{max}, at which the signal has a maximum:

$$\tau_{max} = T_V d/2\delta_s, \quad \delta_s = \sqrt{2DT_V}, \quad (12)$$

where δ_S is the diffusion mean free path of the conduction electrons. The transparency coefficient under the optimal conditions is equal to

$$\eta = 2 \exp\{-d/2\delta_s\}.$$

At $d/\delta_S = 4$, for example, this results in a signal attenuation 10 dB.

In[8] they give a theoretical expression for the transparency coefficient of spin-echo signals, which differ strongly from our results both in the functional dependence and in the numerical estimates. Thus, the results of[8] do not show the increase that takes place in the transparency coefficient with increasing τ as a result of the diffusion propagation of the coherent spins in the interior of the sample. The numerical estimates given in[8] at $d \sim \delta_S$ lead to a signal damping ~95 dB, whereas the physical meaning of the considered conditions for the magnetization to be uniform in the entire sample. (For example, according to formulas (8) under the same conditions, the attenuation obtained at $\tau = T_V$ is ~2 dB.)

The SECE was observed experimentally in samples with small dimensions, and an intermediate situation ($d \gtrsim \delta$) was obtained under certain conditions[1, 2]. One can hope to perform similar experiments on bulky samples in the nearest future. Transient processes under conditions of spin transparency of the metal have not been observed so far, although the undertaking of similar experiments has been reported[12].

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1) In the case of an induction signal, τ has the meaning of the interval between the instants of signal observation and turning off the microwave pulse.

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