Applicability of Bloch model for describing the dynamics of magnetic-resonance signals

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The frequencies of transient oscillations in the EPR signals of systems with effective magnetic moments 1/2 and 1 were found to differ by a factor of 1.4. The investigations were performed on $P3$ and $P6$ centers in silicon irradiated by neutrons. On the basis of an analysis of results on the study of transient oscillations currently available, we show that the Bloch model is inapplicable for the description of dynamic magnetic-resonance signals. The experimental results are explained within the framework of a model which takes into account the difference between the properties of a single particle and an ensemble, as well as the resonance interactions of the magnetic particles with the enviroment.

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1. INTRODUCTION

The quantum-mechanical formalism developed by Bloch to describe the resonance interaction of radio-frequency radiation and magnetic nuclei[1–3] serves as the basis for the examination of the dynamics of spectroscopic transitions. In the case of two-level magnetic systems, this formalism leads to the well-known Bloch equation. Except for infralow temperatures, these same equations are also valid for systems with $I=1$ (Ref. 1). According to such a description, in the absence of relaxation processes the equation of motion for the magnetization vector of a system of non-interacting magnetic particles has the form

$$d\mathbf{M}/dt=\gamma[\mathbf{M}\times\mathbf{H}],$$

where H is the vector sum of the mutually perpendicular d. c. magnetic field $\mathbf{H}_0$ and the a. c. magnetic field $\mathbf{H}_1$, and γ is the gyromagnetic ratio. The solution of this equation indicates the existence of oscillations in the absorption and dispersion signals, with frequency

$$\Omega=[(\gamma H_1)^2+(\omega-\omega_0)^2]^{1/2}.$$

Here ω_0 and ω correspond to the resonance and the microwave frequencies. If at the instant when the field H_1 is turned on the macroscopic magnetization is oriented along the direction of the d.c. magnetic field and is equal to M_0, then the recorded absorption signal is given by $M_0\sin\Omega t$. This behavior of the signals can be observed after abruptly establishing the resonance conditions within times which are short compared with the relaxation times. Although the oscillatory character of the paramagnetic absorption signals was observed by Torrey as early as 1949,[4] a number of results of the Bloch model concerning the course of this process have not yet been confirmed.

Use of the EPR technique, with the establishment of resonance conditions by means of a pulse change in the polarizing magnetic field at a fixed microwave amplitude, has allowed us to investigate transient oscillations with a time resolution of about 80 ns; this has led to new results.[5,6] Comparison of these results with the results from the Bloch model reveals the following obvious discrepancies. First, the observed absorption signal has the form of a damped cosine curve, and not the sine curve obtained from the solution of the Bloch equation under the initial conditions corresponding to the experimental situation. Secondly, the predicted oscillations in the dispersion signals are absent while oscillations in the absorption signal are present under the same conditions. Thirdly, the frequency of the absorption-signal oscillations does not depend on the degree of detuning from resonance. Also, the secondary electron echo signals are not explained in this model.[7]

This paper is devoted to an experimental investigation of oscillations of the EPR signals of systems with differing magnetic moments, and to an analysis of the dynamics of resonant interaction of electromagnetic radiation with magnetic systems.

2. TECHNIQUE AND EXPERIMENTAL RESULTS

A study of the characteristic dynamics of transitions in multilevel systems requires that we ensure identical excitation and registration of signals in samples which contain centers with differing magnetic moments. In our work, comparative studies of the oscillations in the EQR signals are carried out in single crystals or irradiated silicon which simultaneously contain both $P6$ and $P3$ centers (respectively $I=\frac{1}{2}$ and $I=1$) and thus satisfy the condition of identical excitation and registration. Single crystals grown by one melting were irradiated by fast reactor neutrons with an integrated neutron flux of $5\cdot10^{18}-10^{19}$ cm^{-3} at a temperature not higher than 70 °C. After irradiation, the samples ($1\times2\times8$ mm rectangular prisms with (110) base plane) were treated in a polishing etchant CR-4. The investigations were performed with a superheterodyne 3-cm-band spectrometer which provided for recording both the spectral and the time dependences of the EPR signals.[6] The spectral characteristics were recorded with high-frequency modulation at 100 kHz. In the time measurements, the resonance conditions were established abruptly by a change of ~ 1 Oe in the polarizing magnetic field. We went from the spectral studies to the time studies without changing any of the other experimental conditions.

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The $P3$ and $P6$ centers were the predominant paramagnetic defects in the studied samples. They are respectively: {110}—tetravacancy in a neutral charge state[8] and a positively charged cleavage ⟨100⟩—binary interstice.[9] The spectral characteristics of these centers have been rather well studied[8,9]: both centers have principal g-tensor values close to $g=2$, and the lines of the $P6$ center fall entirely within a broader range of fields occupied by the $P3$-center spectrum, and they overlap. The different saturation character of the $P3$ and $P6$ signals was used to separate them and to estimate the contribution of each to the spectral density. We tried to decrease the overlap of the lines of the studied centers by a choice of crystal orientation. At an angle between the [001] direction and the H_0 direction equal to 67° (the plane of rotation of the sample was (110)) we found a field value for which the ratio of the amplitudes of the $P6$ center signal to the amplitude of the $P3$ center signal was a maximum.

The time dependence of the EPR signal was studied in the crystallographic orientation indicated above. The spectral dependence of the nonequilibrium signal obtained at maximum microwave power (~40 mW) is shown on Fig. 1. At field points where only the $P3$ center is recorded under steady-state conditions, we observed the signal represented by curve (a). It can be described by the expression

$$f_1(H_0, t) = B_1(H_0, t)\cos\Omega(P3)t, \qquad (1)$$

where $B_1(H_0, t)$ characterize the damping of the signal detected at the point H_0, and $\Omega(P3) = 40.5\cdot 10^5$ rad/s. Curve (b) is obtained at the point in the spectrum with comparable $P3$ and $P6$ center signal amplitudes and is described by the expression

$$f(H_0, t) = f_1(H_0, t) + B_2(H_0, t)\cos\Omega(P6)t. \qquad (2)$$

Curve (c) corresponds to the point of maximum $P6$ center signal and also is described by an expression of the type (2), but with $B_1(H_0, t) \ll B_2(H_0, t)$. This condition allows us to determine the value of $\Omega(P6)$, equal to $28.6\cdot 10^5$ rad/s.

It was established that the oscillation frequencies

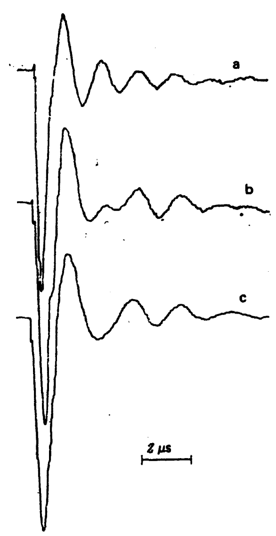

FIG. 1. Oscillations of EPR signals.

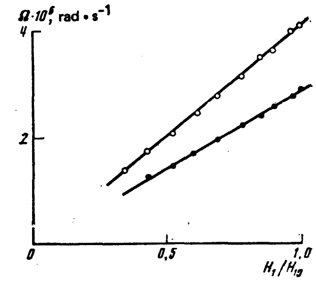

FIG. 2. Dependence of the frequency of the oscillations on the magnitude of the component H_1 of the microwave field for the Si–P3 centers (open circles) and the Si–P6 centers (filled circles); H_{10} is the maximum value of H_1.

$\Omega(P3)$ and $\Omega(P6)$ depend linearly on H_1 (Fig. 2). The ratio is $\Omega(P3)/\Omega(P6) \approx 1.4$ and stays the same for all values of H_1. We did not observe a temperature dependence for the frequencies Ω in the range 77–300 °K.

Since the value of $\gamma(P3)/\gamma(P6) = g(P3)/g(P6)$ does not exceed 1.01 for all the field points, the observed difference in oscillation frequency for the two types of centers cannot be due to a difference in γ. Thus, on the basis of the known models it is impossible to explain the difference in oscillation frequencies for systems of particles with different values of the magnetic moment, nor their cosine character.

3. ANALYSIS OF THE DYNAMICS OF THE MAGNETIC RESONANCE SIGNALS[1)]

The totality of the experimental data above is evidence of the unsuitability of the Bloch formalism for the description of the dynamics of the resonance interaction of microwave radiation with magnetic systems.

In the Bloch model, a change in angle between the directions of the d.c. field and the magnetic moment, both for an individual particle and for the total magnetic moment of the sample, is accomplished at a common frequency equal to Ω and usually small compared with the microwave frequency ω. Consequently, we would predict a zero absorption signal at the instant that the resonance conditions are established.

The experimentally observed maximum absorption signal at the initial instant is evidence that the processes occur in a different manner. An explanation of this fact requires the assumption that the reorientation of the magnetic moment of an individual particle occurs at the frequency ω of the inducing microwave field, and consequently, the oscillations could not be due to the reorientation rate of a single moment but are a collective effect. From this it follows that the absorbed microwave energy recorded at any instant is determined by the difference in the number of flipped magnetic moments; or in other words, to the difference in the populations of the Zeeman levels.

As indicated previously,[6] the reason for oscillations in the absorption signal is the presence of resonance exchange of energies between the magnetic particles and the lattice. During a time short compared with the relaxation processes, such an exchange leads to an accumulation in the lattice of adsorbed energy of the radio-frequency field in the form of resonant phonons. Since resonant phonons are created an annihilated with a change in the state of the magnetic moments, the magnetic system and the system of resonant phonons are interrelated. This relation can be described by the following system of equations:

$$dm/dt = Wn - m/\tau, \tag{3}$$
$$dn/dt = -Wm, \tag{4}$$

where n is the difference between the populations of the Zeeman levels, m and τ are respectively the number of nonequilibrium resonance phonons and their lifetime, W is the probability of stimulated transitions in the magnetic system per unit time. When we can neglect the relaxation, we obtain a solution of this system with equilibrium initial conditions $n(0) = n_0$ and $m(0) = 0$ in the form

$$m(t) = n_0 \sin Wt, \quad n(t) = n_0 \cos Wt, \tag{5}$$

In the case of a two-level system[2]

$$W = \pi (\gamma H_1)^2 f(\omega - \omega_0).$$

Since in order for oscillations to arise it is necessary that H_1 be greater than the homogeneous linewidth, in the case $f(\omega - \omega_0) = 1/\pi\gamma H_1$, and accordingly $W = \gamma H_1$. For a system with $I = 1$ we have $W = \sqrt{2}\gamma H_1$. Consequently, the oscillation frequency is determined only by the probability of stimulated resonance transitions and should not depend on the degree of detuning from resonance.

Since nonequilibrium phons are coherent, their presence does not lead to a change in the phase characteristics of the magnetic system. Accordingly, the dispersion signal remains constant in time, and its amplitude is determined by the number of stimulated transitions in the magnetic system.

The assumption necessary in the construction of the model of resonance transitions, that the absorption of microwave energy lead to a change in the orientation of the magnetic moment, practically ensures that it is impossible to say anything concerning the magnitude of this change. This is connected with the ambiguous notion concerning the orientation of the magnetic moment of the particles in a sample kept rather long time in a d.c. magnetic field. In the quantum mechanical treatment, the direction of the magnetic moment does not coincide with the field direction; therefore when the orientation of the magnetic moment changes only its corresponding projection is operated on and the resonance condition is written as $\omega = g\gamma H_0$, where $g \approx 2$.

From the classical viewpoint, in the situation under consideration the directions of the magnetic moment and the field coincide. As a result of the action of the microwave field, the magnetic moment flips; therefore, the condition for this interaction, in the same notation, has the form $\omega = 2g\gamma H_0$, where g is close to unity. Although this interpretation contradicts current quantum theory, it has a reliable physical interpretation and has the lucidity of a model. At present, sufficient grounds do not exist for peremptorily giving preference to one of these approaches.

From the considerations presented, we see why the Bloch model leads to an incorrect result. One of these reasons is that it is incorrect to the Larmor theorem to describe the behavior of a magnetic moment in an alternating resonant field. Another reason is that in a condensed sample the magnetic particles cannot be considered to be free and we must take into account the resonant interaction between the magnetic moments and the environment.

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