PULSED EXCITATION OF A SYSTEM OF WEAKLY INTERACTING PARTICLES

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The equations of motion of macroscopic quantities characterizing the nonequilibrium state of a many-particle system with a discrete energy spectrum and subjected to the strong action of a pulse generator are deduced by solution of the Schrödinger equation for the system. Simple algorithms are derived for the operator transformations related to pulse excitation. The general properties of pulse generators and of physical systems leading to the appearance of oscillatory and stationary response signals of the excited particle system are established. As an illustration of the application of the general relations, the nonequilibrium states of electron and nuclear spin systems produced by crossed magnetic-sound excitation are investigated. The prospects for the development of pulse techniques in the study of electric and orbital magnetic moment systems are discussed.

1. INTRODUCTION

PHYSICAL systems consisting of a large number of weakly interacting particles possessing a discrete energy spectrum and easily excited by any external action can be used as powerful coherent sources of radiation and as the amplifiers of magnetic [1] and light [2] energy or of the energy of phonons. [3] They are of great interest because of possible practical applications.

While the reaction of such systems to an arbitrary stationary external action has been studied in a number of researches, [4,5] the behavior of physical systems under the pulsed action from an external source has been considered only in the cases of magnetic, [6,7] acoustic, [8,9] and double-pulse [10] excitation of a nuclear spin-system.

The present paper is devoted to the theoretical investigation of the behavior of a system of weakly interacting particles, each of which interacts strongly for a short time interval with an external pulse generator. The concepts of the "weakness" of the internal interaction and of the "strength" of the external action mean that the external action succeeds in changing the state of the particles significantly in a time interval t which is much shorter than the characteristic time τ of interaction between the particles themselves, while the external action should not disrupt in this case the discreteness of the energy spectrum of the physical system or the possibility of selective excitation of the system..

If it is further assumed that the interaction operator of the system and the pulse generator is a one-particle one relative to the excited physical system, then a general solution for the time interval (0,t) can be constructed for the Schrödinger equation for the conservative combination of "system + pulse generator," which does not depend on the specific nature of the system and the generator.

This solution can be used for the calculation of the macroscopic value of an arbitrary physical quantity that is related to the systems considered by us. By the same token, the necessary general conditions can be found which must be satisfied by the physical systems and pulse generators suitable for the creation of coherently functioning quantum devices.

The broad generality of the solutions to problems of the pulse excitation of physical systems is due to the possibility of neglecting the effect of the form factor $g(\nu)$ of the spectral lines, which describes the internal interaction in the substance. To be precise, consideration of $g(\nu)$ makes the exploitation of the theory of the continuous excitation of the substance difficult and is responsible for the disparity between the experimental and theoretical data.

Strong interaction between the system and the pulse generator leads to the result that the energy absorbed by the system ceases to be a satisfactory characteristic of the excitation process, and the different oscillating macroscopic quantities describing the excited state of the system acquire a fundamental significance.

It should be kept in mind that the conditions

for obtaining oscillatory response signals from an excited system are much more severe than the conditions for absorption of energy of the system from the pulse generator. This is a distinguishing property of nonequilibrium states obtained by the pulse method.

It is easy to understand that the pulsed excitation method is more varied than the continuous one, since short-lived excited states of the system are not observed in the latter case. It suffices to show that the pulse method in magnetic spectroscopy allows us to study processes with a characteristic time $\tau \sim 10^{-8}$ sec.[11]

We shall illustrate the general formulas that we obtained for computation of the mean values of quantities relative to nonequilibrium physical systems by the example of magnetoacoustic excitation of electron and nuclear spin systems. However, these formulas also remain valid in the case of other sources of excitation of matter, as, for example, in the case of the excitation of paramagnetic crystals by a flux of cold neutrons or a coherent light beam from a laser.

2. SOLUTION OF THE SCHRÖDINGER EQUATION

Let us consider a closed system described by the Hamiltonian

$$\mathcal{H} = \mathcal{H}_S + \mathcal{H}_F + \mathcal{H}_{SF}, \qquad \mathcal{H}_{SF} = \sum_{\alpha,\gamma} A_S^{\alpha} B_F^{\gamma}, \tag{1}$$

where $\mathcal{H}_S$, $\mathcal{H}_F$, and $\mathcal{H}_{SF}$ describe the matter, pulse generator and their interaction, respectively. The indices S and F denote the fact that the given quantities depend only on the variables of the material and of the generator.

At the time t = 0, let the state of the system $\mathcal{H}$ be described by the function $\Psi(0)$. Then, at a later time t we have

$$\Psi(t) = L(t)\,\Psi(0), \tag{2}$$

where the evolution operator L(t) is found by a solution of the Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} L(t) = \mathcal{H} L(t) \tag{3}$$

for the boundary condition L(0) = 1. It follows from (3) that

$$L(t) = \exp[-it\hbar^{-1}\mathcal{H}]. \tag{4}$$

For simplicity of subsequent calculations, it is convenient to expand the exponential in (4) by the Feynman formula:[12]

$$\exp[-it\hbar^{-1}\mathcal{H}] = \exp[-it\hbar^{-1}\mathcal{H}_F]\exp[-it\hbar^{-1}\mathcal{H}_S] \times \exp\left[-i\hbar^{-1}\int_0^t \mathcal{H}_{SF}(t')\,dt'\right], \tag{5}$$

where

$$\mathcal{H}_{SF}(t) = \exp[i\hbar^{-1}t(\mathcal{H}_S + \mathcal{H}_F)]\,\mathcal{H}_{SF} \times \exp[-it\hbar^{-1}(\mathcal{H}_S + \mathcal{H}_F)].$$

The mean value of the operator Q at the moment of time t is computed from the formula

$$\langle Q(t)\rangle = \mathrm{Sp}\{q_0 L(t)^{-1} Q L(t)\}, \qquad q_0 = e^{-\beta\mathcal{H}}\{\mathrm{Sp}\, e^{-\beta\mathcal{H}}\}^{-1}, \tag{6}$$

where β is an operator describing the populations of the energy levels of the system $\mathcal{H}$ at the time t = 0. If Boltzmann statistics are valid, and there exists a unique temperature T for the entire system $\mathcal{H}$, then $\beta = 1/kt$, where k is the Boltzmann constant.

For calculation of transformations of the type $L(t)^{-1}QL(t)$, the following formulas are useful:

$$Q(t) = \exp\left\{i\hbar^{-1}t\sum_v K_v P K_v\right\} Q \exp\left\{-i\hbar^{-1}t\sum_v K_v P K_v\right\} = \sum_{a,b} K_a Q K_b e^{i\omega_{ab}t},$$
$$\omega_{ab} = \hbar^{-1}\{R_a K_a P K_a - R_b K_b P K_b\}, \tag{7}$$

where K_V are projection operators, P is the diagonal operator, $K_V P K_V$ is an operator, the nonvanishing part of which is a multiple of the unit operator, and $R_a = [\mathrm{Sp}\, K_a]^{-1}$.

In the general case, we have

$$Q(t) = \exp[i\hbar^{-1}tP]\,Q\exp[-i\hbar^{-1}tP] = \sum_{n=0}^{\infty}\frac{(it\hbar^{-1})^n}{n!}\sum_{v=0}^{n}(-1)^v C_n^v P^{n-v} Q P^v = \sum_{n=0}^{\infty}(-1)^n\frac{(t\hbar^{-1})^{2n}}{(2n)!}\sum_{v=0}^{2n}(-1)^v C_{2n}^v P^{2n-v} Q P^v + i\sum_{n=0}^{\infty}(-1)^n\frac{(t\hbar^{-1})^{2n+1}}{(2n+1)!}\sum_{v=0}^{2n+1}(-1)^v \times C_{2n+1}^v P^{2n+1-v} Q P^v. \tag{8}$$

Let $P = P_{ab} + P_{ba}$, where $P_{ab} = K_a P' K_b$, and $P_{ba} = K_b P' K_a$. Then

$$(P_{ab} + P_{ba})^{2n} = 1_{aa}(P_{ab}P_{ba})^n + 1_{bb}(P_{ba}P_{ab})^n, \tag{9}$$

$$(P_{ab} + P_{ba})^{2n+1} = 1_{ab}(P_{ab}P_{ba})^n P_{ab} + 1_{ba}(P_{ba}P_{ab})^n P_{ba} = 1_{ab}P_{ab}(P_{ba}P_{ab})^n + 1_{ba}P_{ba}(P_{ab}P_{ba})^n, \tag{10}$$

where the symbol $1_{\alpha\beta}$ is introduced for the designation of the subspace separated out by the projection operators K_α and K_β, while this subspace is filled up with the nonvanishing part of some matrix $K_\alpha B K_\beta$.

Substituting (9) and (10) in (8), we get

$$Q(t) = \sum_{n=0}^{\infty} (-1)^n \frac{(t\hbar^{-1})^{2n}}{(2n)!} \Big\{ \sum_{k=2}^{2n-2} C_{2n}^{k} [1_{aa} A^{(2n-k)/2} Q_{aa} A^{k/2}$$

$$+ 1_{ab} A^{(2n-k)/2} Q_{ab} B^{k/2} + (a \leftrightarrow b)] + \sum_{\gamma} [1_{a\gamma} A^n Q_{a\gamma}$$

$$+ 1_{\gamma a} Q_{\gamma a} A^n + (a \leftrightarrow b)] - \sum_{r=3}^{2n-3} C_{2n}^{r}$$

$$\times [1_{aa} P_{ab} B^{(2n-r-1)/2} Q_{bb} B^{(r-1)/2} P_{ba}$$

$$+ 1_{ab} P_{ab} B^{(2n-r-1)/2} Q_{ba} A^{(r-1)/2} P_{ab} + (a \leftrightarrow b)]$$

$$- [1_{aa} P_{ab} Q_{bb} B^{n-1} P_{ba} + 1_{aa} P_{ab} B^{n-1} Q_{bb} P_{ba}$$

$$+ 1_{ab} P_{ab} Q_{ba} A^{n-1} P_{ab}$$

$$+ 1_{ab} P_{ab} B^{n-1} Q_{ba} P_{ab} + (a \leftrightarrow b)] \Big\} + i \sum_{n=0}^{\infty} (-1)^n \frac{(t\hbar^{-1})^{2n+1}}{(2n+1)!}$$

$$\times \Big\{ \sum_{k=2}^{2n-2} C_{2n+1}^{k} [1_{aa} P_{ab} B^{(2n-k)/2} Q_{ba} A^{k/2}$$

$$+ 1_{ab} P_{ab} B^{(2n-k)/2} Q_{bb} B^{k/2} + (a \leftrightarrow b)]$$

$$+ [1_{aa} P_{ab} Q_{ba} A^n + 1_{ab} P_{ab} Q_{bb} B^n - 1_{aa} A^n Q_{ab} P_{ba}$$

$$- 1_{ab} A^n Q_{aa} P_{ab} + (a \leftrightarrow b)]$$

$$- \sum_{r=3}^{2n-1} C_{2n+1}^{r} [1_{aa} A^{(2n+1-r)/2} Q_{ab} B^{(k-1)/2} P_{ba}$$

$$+ 1_{ab} A^{(2n+1-r)/2} Q_{aa} A^{(r-1)/2} P_{ab} + (a \leftrightarrow b)]$$

$$+ \sum_{\gamma} [1_{a\gamma} P_{ab} B^n Q_{b\gamma} - 1_{\gamma a} Q_{\gamma b} B^n P_{ba} + (a \leftrightarrow b)] \Big\},$$

$$A = P_{ab} P_{ba}, \qquad B = P_{ba} P_{ab}, \tag{11}$$

where k is an even number, r is an odd number, and the symbol $(a \leftrightarrow b)$ denotes that one must add an expression to the foregoing which is obtained from it by replacing a by b.

1. We shall now discuss some general properties of the transformations (7) and (11). It follows from (7) that Q(t) can have oscillating matrix blocks if the operator Q possessed nondiagonal matrix blocks prior to the transformation, i.e.,

$$K_a Q K_b \neq 0, \quad a \neq b. \tag{12}$$

2. Let the indices a and b take on the values $1, 2, \ldots, n$. We shall assume that for the operator Q the only such non-zero blocks $K_a Q K_b$ are those for which $|a-b| = \eta$, while the corresponding condition for the operator P has the form $|a'-b'| = \xi$. It is easy to see from Eq. (8) that Q(t) can have diagonal blocks if η is divisible by ξ, a fact which we denote by

$$\eta : \xi. \tag{13}$$

In particular, $\eta = \xi = 0$ corresponds to the identity transformation.

For $\xi > \eta$, the operator Q(t) does not have diagonal blocks. For $\eta = 0$ and arbitrary ξ, the operator Q(t) possesses diagonal blocks.

3. If Eq. (8) does not depend on k or (11) on k and r, then Q(t) is expressed in terms of the sine and cosine of the matrix argument, which, in the case of an argument in the form of a diagonal matrix, reduces to the usual trigonometric functions.

4. Equation (11) divides into two parts, which differ essentially from each other. The first part, which depends on the matrix cosine, describes the change in the nonvanishing matrix blocks of the operator already present, but does not produce new, nonvanishing submatrices. The second part, which depends on the matrix sine, characterizes new properties acquired by the matrix Q(t) resulting from the transformation $L(t)\,QL(t)^{-1}$. For example, diagonal matrix blocks produce nondiagonal blocks and, vice versa, nonzero nondiagonal matrix blocks appear between the new subspaces. At the same time, the very structure of the blocks $K_a Q K_b$ can change, i.e., blocks with diagonal structure can transform into blocks with nondiagonal structure.

The foregoing properties of the transformations (7) and (11) characterize interesting physical transformations, which come about for a system of many particles in its pulse excitation by an external source. We shall illustrate these properties by the example of the single particle model studied in the present research.

3. SINGLE PARTICLE APPROXIMATION

Let

$$\mathcal{H}_S = \mathcal{H}_0 + \mathcal{H}_1 = \sum_i \mathcal{H}_0^i + \sum_{i>j} \mathcal{H}_1^{ij},$$

$$Q = \sum_i Q^i, \quad A_S^{\alpha} = \sum_i A_S^{\alpha i}, \tag{14}$$

where $\mathcal{H}_0$ is the fundamental Hamiltonian, $\mathcal{H}_1$ is the perturbation; $i, j = 1, \ldots, N$ are the indices for designating the particles, and the time τ characterizes the relaxation process brought about by the interactions $\mathcal{H}_1$. For sufficiently high temperatures T and sufficiently short times t of the pulse action, characterized by the inequalities

$$kT \gg N^{-1} |\mathcal{H}_1|, \qquad \tau \gg t, \tag{15}$$

we obtain, with account of (5), (6), (14), and (15),

$$|\langle Q(t) \rangle = N \langle Q^i(t) \rangle = N \operatorname{Sp} \{ q_0^i L^i(t)^{-1} Q^i L^i(t) \}, \tag{16}$$

$$L^i(t) = \exp[-i\hbar^{-1} t \mathcal{H}_F] \exp[-it\hbar^{-1} \mathcal{H}_0^i]$$

$$\times \Big| \exp\Big[-i\hbar^{-1} \int_0^t \sum_{\alpha, \gamma} A_S^{\alpha i}(t') B_F^{\gamma}(t')\, dt' \Big], \tag{17}$$

$$q_0^i = \exp\left[-\beta\mathcal{H}_0^i\right]\{\mathrm{Sp}\exp\left[-\beta\mathcal{H}_0^i\right]\}^{-1}, \quad (18)$$

$$A_S^{\alpha i}(t) = \exp\left[i\hbar^{-1}t\mathcal{H}_0^j\right] A_S^{\alpha j} \exp\left[-i\hbar^{-1}t\mathcal{H}_0^j\right]. \quad (19)$$

The expression $\mathcal{H}_{SF}$ contains factors of two types: the A_S^α describe the internal parameters of the system $\mathcal{H}_S$ which can be changed by the external influence $\mathcal{H}_F$, and the B_F^γ characterize the external effect conjugate with the A_S^α. For example, in the case of paramagnetic resonance, the A_S^α and B_F^γ describe the components of the magnetic moment of the spin system and of the variable magnetic field, respectively.[6,7] For acoustic magnetic resonance on nuclei these quantities correspond to the components of the quadrupole moment tensor of the nucleus and the components of the deformation tensor, which are associated with the ultrasonic propagation.[8,9] While the A_S^α are usually operator quantities, the B_F^γ are described by classical functions. The latter is due to the fact that the pulse generator has a limited number of degrees of freedom and an extraordinarily high degree of excitation.[13]

Let E_α be the eigenvalues of the operator $\mathcal{H}_0^i = \mathcal{H}_0^j$ (i, j = 1, ..., N), where $\omega_{\alpha\beta} = \hbar^{-1}(E_\alpha - E_\beta)$ and

$$|\omega_{\alpha\beta}| - |\omega_{\gamma\delta}| \gg t^{-1}, \quad (20)$$

i.e., there are no equidistant pairs of levels in the spectrum of the operator $\mathcal{H}_0^j$.

With sufficient generality, we can write

$$\sum_{\alpha,\gamma} A_S^{\alpha j} B_F^\gamma(t) = R\cos\omega_{ab}t + D\sin\omega_{ab}t,$$

whence, with allowance for (20), we obtain

$$\int_0^t \sum_{\alpha,\gamma} A_S^{\alpha j}(t') B_F^\gamma(t')\,dt' = t(P_{ab} + P_{ba}), \quad (21)$$

$$P_{ab} = \tfrac{1}{2}K_a(R - iD)K_b, \qquad P_{ba} = \tfrac{1}{2}K_b(R + iD)K_a. \quad (22)$$

The relations (15)—(19) describe the single particle approximation, while the formulas (20)—(22) are the conditions for maximum selectivity of the excitation.

Let a succession of k-pulse generators of different type act on the system $\mathcal{H}_S$ over the time intervals $\sum_{n=1}^{k}\Delta t_n = t$. Application of Feynman's theorem to the expansion of the exponentials in (17) gives the result

$$L^i(t) = \exp\left[-i\hbar^{-1}t\mathcal{H}_F\right]\exp\left[-i\hbar^{-1}t\mathcal{H}_0^j\right] \times \prod_{n=1}^{k}\exp\left[-i\hbar^{-1}\Delta t_n(P_{a_nb_n} + P_{b_na_n})\right], \quad (23)$$

where the exponentials in the product are written from right to left in the order in which the pulses acting on the system occur.

4. CLASSIFICATION OF THE QUANTITIES CHARACTERIZING THE NONEQUILIBRIUM SYSTEM

We introduce the following terminology. If, after the pulse generator is shut off, the quantity $\langle Q(t+t_0)\rangle$ executes oscillations at a later instant of time $t+t_0$ with frequencies $\omega_{\alpha\beta}$ corresponding to intervals of energy in the unperturbed spectrum of particles j, then we shall call the corresponding signal, the free induction signal (IS). If the IS decays as the result of some reverse process and repeated pulse action leads to a regeneration of the signal, then this responding reaction of the system will be called the echo signal (ES). In the absence of oscillations with frequencies $\omega_{\alpha\beta}$, the mean value of $\langle Q(t+t_0)\rangle$ will characterize the polarization of the quantity Q. We can then write that

$$\langle Q(t+t_0)\rangle = \langle Q(t)\rangle f(t+t_0), \quad (24)$$

where $f(t+t_0)$ is a certain correlation function, which is equal to unity at $t_0 = 0$. If quantum transitions with a change in the unperturbed energy of the particles are required to disrupt the nonequilibrium value, then the parameter $\tau_{||}$ of the function $f(t+t_0)$ characterizes the longitudinal relaxation. In the opposite case, there is a single relaxation time, or the variation of the function $f(t+t_0)$ is described by a transverse relaxation time $\tau_\perp$, where $\tau_\perp \le \tau_{||}$ (see [7]).

Let Q depend only on the variables of the system $\mathcal{H}_S$. Then the factor $\exp\left[-i\hbar^{-1}t\mathcal{H}_F\right]$ in (23) does not act on Q and the oscillations of the quantity Q(t) can be produced only by the diagonal operator $\exp\left[-i\hbar^{-1}t\mathcal{H}_0^j\right]$. Therefore, the induction and echo signals can be observed only for the values of Q which satisfy the condition (12). For example, in the case of axial symmetry of the spin Hamiltonian of paramagnetic particles $\mathcal{H}_0$ the magnetic IS and ES can be observed only on the components of the magnetic moment which are directed perpendicular to the axis of axial symmetry.

In order that the quantity $\langle Q(t)\rangle$ exist, the operator Q(t) ought to have nonzero diagonal blocks. Therefore, in correspondence with the structure of the operator Q, the type of exciting pulse $P_{ab} + P_{ba}$ should be selected on the basis of the rule (13). For example, in the case of axial symmetry of $\mathcal{H}_0^j$, the transverse components of the magnetic moment have matrix elements only

between states with a change in quantum number of $\Delta m = \pm 1$, i.e., $\eta = 1$. Therefore, the ultrasonic excitation which produces transitions $\Delta m > \pm 1$ ($\eta > 1$ and ξ is not divisible by η), does not excite IS and ES.[8,9] At the same time, for the operator of the electric quadrupole moment of the nucleus $\Delta m = \pm 2$, $\eta = 2$, and pulse excitation for $\xi = 1$ and 2 produces corresponding electric quadrupole induction and echo signals.[8] For the longitudinal component of the magnetic moment $\eta = 0$, and the mean value of this quantity can be changed by pulse interaction for arbitrary values $\xi > 0$.[9]

Interesting phenomena are observed for multiple pulse excitation of the system $\mathcal{K}_S$. Let η_0, referred to the operator Q at $t = 0$, be equal to unity and the system $\mathcal{K}_S$ be excited successively by pulses of the type (23), where $\xi_n = n$ ($n = 1, \ldots, k$) for the n-th pulse. In accord with (13), the first pulse can produce the signal $\langle Q(\Delta t_1)\rangle \neq 0$, while, simultaneously, the blocks $\eta_1 = 2$ appear for the operator $Q(\Delta t_1)$. Therefore, after the second pulse, one can obtain the signal from the blocks $\eta_1 = 2$. After the n-th pulse, one can obtain the signal from the blocks $\eta_{n-1} = n$ and in the same way study the properties of the interaction of the pulse generator ($\xi = n$) and the system $\mathcal{K}_S$. For example, an ultrasonic pulse cannot bring about the appearance of IS and ES, but "magnetic + ultrasonic" pulses can.[14] Such a "recurrence" method can be used for the study of interactions with large ξ.

5. MAGNETIC-SOUND EXCITATION OF A SPIN-SYSTEM IN THE ABSENCE OF A CONSTANT MAGNETIC FIELD

In dielectric paramagnetic crystals, discreteness of the energy spectrum of the spin system $\mathcal{K}_S$ frequently arises from the Stark energy of the magnetic particles in the internal crystalline electric field. Such substances, are characterized by a wide range of dynamic properties and if used in quantum devices they call for strong static magnetic fields. We shall investigate the possibility of excitation of such a system by a double pulse.

Let

$$\mathcal{H}_0^j = D\left[s_z^{j2} - \tfrac{1}{3}s(s+1)\right], \tag{25}$$

where s is the effective spin and D is a constant of the axial crystalline electric field.[15]

Except for the eigenvalue $E_{m=0}$ all the E_m are doubly degenerate for even s and correspond to the eigenfunctions $|\pm m\rangle$, where m is the modulus of the magnetic quantum number. In what follows, we shall exclude from consideration all cases in which the matrix elements $\langle \alpha | m = 0\rangle$ play an important role.

Let the spin system be acted on successively by the pulsed variable magnetic field

$$R\cos\omega_{ab}t + D\sin\omega_{ab}t = g_\perp\beta\,[s_xH_x\cos\omega_{m+1}t + s_yH_y\sin\omega_{m+1}t] \tag{26}$$

and the sound field

$$R\cos\omega_{ab}t + D\sin\omega_{ab}t = (A_1Q_{\pm1} + B_1v_{\pm1} + A_2Q_{\pm2} + B_2v_{\pm2})\cos\omega_m t, \tag{27}$$

where

$$\omega_{m+1} = \hbar^{-1}(E_{m+2} - E_{m+1}), \qquad \omega_m = \hbar^{-1}(E_{m+1} - E_m),$$
$$Q_{\pm1} = s_xs_z + s_zs_x, \qquad v_{\pm1} = s_ys_z + s_zs_y,$$
$$Q_{\pm2} = s_x^2 - s_y^2, \qquad v_{\pm2} = s_xs_y + s_ys_x,$$

β is the magneton, $g_\perp$ is the spectroscopic splitting factor, H_α are the amplitudes of the variable magnetic field along the α axes.

The phenomenological coefficients A_1, A_2, B_1, and B_2 depend on the configuration of the acoustic field, the sound intensity, the elastic constants of the crystal and the coupling between the energy of the system $\mathcal{K}_S$ and the deformations of the crystal, brought about by the propagation of the ultrasound. In the case of cubic crystals, explicit expressions for these coefficients are well known (see [8]), while the general case is discussed in [16].

In accord with (22), (26), and (27), the non-zero blocks of the matrix have the form

$$P_{m,m+1} = -m\begin{vmatrix} -(m+1) & (m+1) \\ l_1 & l_2 \\ m & l_3 & l_4 \end{vmatrix};$$

$$P_{m+1,m} = -(m+1)\begin{vmatrix} -m & m \\ l_1^* & l_3^* \\ (m+1) & l_2^* & l_4^* \end{vmatrix}. \tag{28}$$

In the case of circular polarization of the magnetic field and the transition $|m\rangle \longleftrightarrow |m+1\rangle$,

$$l_1 = l_2 = l_3 = 0,\ l_4 = 2\alpha = \tfrac{1}{2}g_\perp\beta H\,[(s+m+1)(s-m)]^{1/2},$$
$$\omega_m = \hbar^{-1}D\,(2m+1). \tag{29}$$

For linear polarization ($H_y = 0$) of the magnetic field, we have

$$l_1 = l_4 = \alpha, \qquad l_3 = l_2 = 0. \tag{30}$$

In the case of sonic excitation, we have

$$l_1 = -\alpha_1^*, \quad l_2 = \alpha_2^*, \quad l_3 = \alpha_2, \quad l_4 = \alpha_1,$$
$$\alpha_1 = \langle s, m|A_1Q_{\pm1} + B_1v_{\pm1}|s, m+1\rangle,$$
$$\alpha_2 = \langle s, m|A_2Q_{\pm2} + B_2v_{\pm2}|s, -(m+1)\rangle, \tag{31}$$

where $\langle s, m | s, m+1\rangle$ denotes the matrix element between the states $|s, +m\rangle$ and $|s, +(m+1)\rangle$.

Let an ultrasonic pulse of frequency ω_m act on a spin system with magnetic moment μ over the time interval $(t_{m+1}, t_{m+1} + t_m)$, and a pulse of the variable magnetic field of circular polarization and frequency ω_m over the interval $(0, t_{m+1})$. Applying Eqs. (6), (18), (23), (29), and (31), we find that at the time $t_m + t_{m+1}$ the macroscopic value of the magnetic moment of the spin system is described by the formula

$$\langle \mu (t_m + t_{m+1}) \rangle^{BH}_{m, m+1; m+1, m+2} = N\beta R_{m+1, m+2} [A_1 z + (A_{21} + A_{22}) x + (A_{31} + A_{32}) y], \quad (32)$$

$$R_{\alpha\beta} = (e^{-E_\alpha/kT} - e^{-E_\beta/kT}) \Big[\sum_\gamma^{2s+1} e^{-E_\gamma/kT} \Big]^{-1},$$

$$A_1 = \frac{1}{2} g_\parallel [1 - \cos 4\theta_{m+1} t_{m+1}] \Big\{ 1 + \frac{1}{2} [1 - \cos 2\theta_m^{(4)} t_m] \times \Big[m + 1 + \frac{m(|\alpha_2|^2 - |\alpha_1|^2)}{|\alpha_1|^2 + |\alpha_2|^2} \Big] \Big\}, \quad (33)$$

$$A_{21} = -\langle m+1 | m+2 \rangle |g_\perp| \times \sin 4\theta_{m+1} t_{m+1} \cos 2\theta_m^{(4)} t_m \sin \omega_{m+1} (t_m + t_{m+1}) + \langle m | m+1 \rangle (|\alpha_1|^2 + |\alpha_2|^2)^{-1/2} \sin 2\theta_m^{(4)} t_m \times \{ \cos \omega_m (t_m + t_{m+1}) [\mathrm{Im}(\alpha_1^*) [2g_\perp + \tfrac{1}{2}(1 - \cos 4\theta_{m+1} t_{m+1}) |g_\perp|]] + \sin \omega_m (t_m + t_{m+1}) \times [\mathrm{Re}(\alpha_1^*) \tfrac{1}{2} (\cos 4\theta_{m+1} t_{m+1} - 1) |g_\perp|] \}, \quad (34)$$

$$A_{22} = \delta(m - \tfrac{1}{2}) \tfrac{1}{2} |g_\perp| (|\alpha_1|^2 + |\alpha_2|^2)^{-1} (1 - \cos 4\theta_{m+1} t_{m+1}) \times (\cos 2\theta_m^{(4)} t_m - 1) \mathrm{Re}(\alpha_1^* \alpha_2^*), \quad (35)$$

$$A_{31} = \langle m+1 | m+2 \rangle |g_\perp| \times \sin 4\theta_{m+1} t_{m+1} \cos 2\theta_m^{(4)} t_m \cos \omega_{m+1} (t_m + t_{m+1}) + \langle m | m+1 \rangle (|\alpha_1|^2 + |\alpha_2|^2)^{-1/2} \times \sin 2\theta_m^{(4)} t_m \{ \cos \omega_m (t_m + t_{m+1}) \mathrm{Re}(\alpha_1^*) [2g_\perp + \tfrac{1}{2}(1 - \cos 4\theta_{m+1} t_{m+1}) |g_\perp|] + \sin \omega_m (t_m + t_{m+1}) \times [\mathrm{Im}(\alpha_1^*) \tfrac{1}{2} (1 - \cos 4\theta_{m+1} t_{m+1}) |g_\perp|] \}, \quad (35')$$

$$A_{32} = \delta(m - \tfrac{1}{2}) \tfrac{1}{2} |g_\perp| (|\alpha_1|^2 + |\alpha_2|^2)^{-1} (1 - \cos 4\theta_{m+1} t_{m+1}) \times (\cos 2\theta_m^{(4)} t_m - 1) \mathrm{Im}(\alpha_1^* \alpha_2^*), \quad (36)$$

where $\langle \alpha | \beta \rangle = \langle \alpha | s_x | \beta \rangle$, $\delta(m - 1/2)$ is the Kronecker symbol, α^* and α are complex conjugate numbers.

The parameters of "rotation" of the components of the magnetic moment under the action of the pulse generator are determined by the formulas

$$\theta_\varepsilon = \hbar^{-1} |\alpha| = \left| \tfrac{1}{4} \hbar^{-1} g_\perp \beta H_x [(s + \varepsilon + 1)(s - \varepsilon)]^{1/2} \right|, \quad (37)$$

$$\theta_\varepsilon^{(4)} = \hbar^{-1} (|\alpha_1|^2 + |\alpha_2|^2)^{1/2}, \quad (38)$$

where α_1 and α_2 are computed from (31).

If the variable pulsed magnetic field of circular polarization acts in the time interval $(t_m, t_m + t_{m+1})$ and the ultrasonic field in the time interval $(0, t_m)$, then we get in place of (32)

$$\langle \mu (t_{m+1} + t_m) \rangle^{HB}_{m+1, m+2; m, m+1} = N\beta [A_3 z + (A_{41} + A_{42}) x + (A_{51} + A_{52}) y], \quad (39)$$

$$A_3 = \tfrac{1}{2} g_\parallel [1 - \cos 4\theta_{m+1} t_{m+1}] \times [\tfrac{1}{2}(1 - \cos 2\theta_m^{(4)} t_m) R_{m, m+1} + R_{m+1, m+2}], \quad (40)$$

$$A_{41} = -\langle m+1 | m+2 \rangle \sin 4\theta_{m+1} t_{m+1} \sin \omega_{m+1} (t_{m+1} + t_m) |g_\perp| \times [\tfrac{1}{2}(1 - \cos 2\theta_m^{(4)} t_m) R_{m, m+1} + R_{m+1, m+2}], \quad (41)$$

$$A_{42} = 2 \langle m | m+1 \rangle g_\perp (|\alpha_1|^2 + |\alpha_2|^2)^{-1/2} \sin 2\theta_m^{(4)} t_m \cos \omega_m \times (t_{m+1} + t_m) \cos 2\theta_{m+1} t_{m+1} \mathrm{Im}(\alpha_1^*) R_{m, m+1}, \quad (42)$$

$$A_{51} = \langle m+1 | m+2 \rangle \sin 4\theta_{m+1} t_{m+1} \cos \omega_{m+1} (t_{m+1} + t_m) |g_\perp| \times [\tfrac{1}{2}(1 - \cos 2\theta_m^{(4)} t_m) R_{m, m+1} + R_{m+1, m+2}], \quad (43)$$

$$A_{52} = 2 \langle m | m+1 \rangle g_\perp [|\alpha_1|^2 + |\alpha_2|^2]^{-1/2} \times \sin 2\theta_m^{(4)} t_m \cos \omega_m (t_{m+1} + t_m) \times \cos 2\theta_{m+1} t_{m+1} \mathrm{Re}(\alpha_1^*) R_{m, m+1}. \quad (44)$$

After the pulse generator is turned off, the signals and the polarization described by Eqs. (32) and (40) will be damped according to (24).

We shall stop to consider the physical content of Eqs. (32)—(44).

1) Comparison of (32) and (40) shows that the result of the action of two pulses depends essentially on the order of the pulse sequence. This follows directly from Eq. (23), and is brought about by the fact that, in a change of order of alternation of the pulses, the magnetic and sound fields "see" different initial conditions of the spin system. Therefore, the information obtained by the pulse technique is proportional to the factorial of the number of successive pulses.

2) The terms (33) and (40) describe the polarization of the magnetic moment along the direction of the axis of axial symmetry of the crystalline electric field, which arises only as the result of the action of the pulse excitation, since for $t_m + t_{m+1} = 0$ we get $A_1 = A_3 = 0$. For low temperatures, in the region $kT \ll D$, the pulse method allows us, in the case (33) and especially in the case (40), when the mean value of the operators is proportional to $R_{m,m+1} + R_{m+1,m+2} = R_{m,m+2}$, to obtain the absolute polarization of the magnetic moment. Since $D\hbar^{-1} \sim 2000$ Mc for the spin system of I^{127} in certain diamagnetic crystals,[17] the absolution nuclear polarization p can be achieved even for $T \sim 0.1°K$, while the value of p does not depend on the character of the internal interaction and is preserved during the longitudinal

relaxation of the nuclear magnetic moment.

3) The terms A_{21} and A_{31} describe the macroscopic components of the magnetic moment of the spin system, oscillating in the xy plane. These components bring about the appearance of signals of free magnetic induction. The first component in A_{21} oscillates with the frequency ω_{m+1} and contains the factors $4\theta_{m+1}t_{m+1}$ and $\cos 2\theta_m^{(4)}t_m$, i.e., the appearance of this component is due to magnetic excitation and the value of the corresponding signal is modulated by the sound pulse. The second component is brought about by sonic excitation, where the character of the modulation of this signal by the variable magnetic field depends on the sign of the g factor. The components in A_{31} have a similar meaning.

4) The terms A_{22} and A_{32}, which are different from zero only for $m = 1/2$, describe the polarization of the magnetic moment in the xy plane and are produced by the combined action of the sound and magnetic pulses, since the separate pulses do not result in the appearance of nonoscillating macroscopic components of μ perpendicular to the symmetry axis of the crystalline field. In contrast to the polarization along z, the nonequilibrium values of A_{22} and A_{32} decay after the time of transverse magnetic relaxation and are therefore very unstable.

5) The terms (A_{41}, A_{51}) and (A_{42}, A_{52}) describe the free induction signals at frequencies ω_{m+1} and ω_m. Factors of the type $(aR_{m,m+1} + R_{m+1,m+2})$ characterize the intensification of the signals originating under double pulse excitation.

6) In view of the small wavelength of the hypersound, it is difficult to put the sample inside one loop of the standing wave. Therefore, in (32) and (39), all the terms containing the factor $\mathrm{Re}(\alpha_k)$ or $\mathrm{Im}(\alpha_k)$ explicitly vanish upon summation over the particles inside the entire sample if an even number of loops of the sound wave is contained inside the specimen. If the number of loops is odd, then the number N in this formula enumerates the magnetic particles over the sample inside a single loop of the standing sound wave.

On the other hand, terms of the type (A_{41}, A_{51}) and the first components in (A_{21}, A_{31}), for which the sound vibrations of the crystal play the role of modulator of the intensity of the signals from magnetic excitation, describe volume effects in which the corresponding signals possess high intensity, i.e., the intensity of the usual induction and echo signals in the magnetic excitation.[6,11]

Since the time of transverse magnetic relaxation in nuclear and dilute electron spin systems changes in the range 10^{-5}—10^{-2} sec and 10^{-8}—10^{-6} sec, respectively, the length of the acoustic pulse can be made much shorter than these times if the surface of the crystal is displaced a distance of 10^{-7}—10^{-8} cm under the action of the sound vibrations.[8] Different methods of calculations of the quantities α_k are shown in [16].

6. CONCLUSION

The nonequilibrium states of systems with discrete energy spectra are of interest from the point of view of obtaining "negative" temperatures, of investigation of relaxation processes and of pulse spectroscopy.

The possibility of obtaining nonequilibrium states of the system and in the same fashion, "negative" temperatures, reduces to the realization of the conditions

$$\tau \ll t, \quad \theta t = \pi, \tag{45}$$

where the further free development of the state of the system determines the dynamic constants of the system.

The task of pulse spectroscopy is to determine the constants θ of the interaction between the pulse generator and the different degrees of freedom of the system from the initial intensities of the induction and echo signals. Hitherto the "indicators" of the value of θ were the macroscopic components of the magnetic moment of the spin system, and the possibility of using magnetic "indicators" were far from exhausted. For example, a possibility of studying the interaction of the crystalline electric field with the electron and nuclear spin magnetic moment follows from (32) and (39). It follows from the results of Bloembergen[18] that application of pulsed variable electric fields makes it possible to study the covalent bond in molecules.

It follows from the general relations (5) and (11) that the application of the pulsed light beams radiated by lasers,makes it possible to investigate and use for practical purposes systems of electric moments in crystals in addition to the spin systems. On the other hand, the "magnetic indicators" of the system of orbital magnetic moments can be used for the measurement of the intensity of flow of cold neutrons passing through matter, while the nonequilibrium states of such systems of orbital moments can be used as quantum neutron accelerators.

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220