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Translated by J. G. Adashko

Theoretical investigation of the effect of irreversible relaxation on signals such as photon echo following multipulse excitation

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(Submitted May 26, 1976)
Zh. Eksp. Teor. Fiz. 72, 2130–2140 (June 1977)

With irreversible relaxation taken into account, a relatively complete and detailed classification is presented for the sets of responses, such as optical induction or photon echo, which appear in large samples of matter as a result of multipulse excitation. Allowance is made for the responses that appear during the intermediate stages of the excitation process and for the responses after the end of the excitation process. The excited samples are regarded as quantum systems consisting of a large number of identical noninteracting particles with a discrete nondegenerate finite equidistant energy-level spectrum describable with the aid of energy-spin concepts. The operator of the interaction of the field of the pulses is linear in the energy spin. The transverse and longitudinal irreversible relaxations are accounted for by phenomenological spin operators defined by time-dependent differential equations whose averaging yields equations of the Bloch type. A new matrix method of investigating the solutions of these equations is developed. Some examples of possible technical applications of the relations obtained in this paper are discussed. These relations, in particular, can help choose the most convenient sequences of exciting pulses for the measurement of the times of transverse and longitudinal irreversible relaxation, and can also be useful in the development of spin, acoustic, and optical memory elements.

PACS numbers: 42.65.Gv

A natural continuation of Dicke's ideas[1] was the theoretical prediction of the photon-echo phenomenon.[2] Photon echo in ruby was observed experimentally in a number of studies.[3] A relatively detailed theory of photon echo was given in[4] on the basis of the formalism developed by Dicke.[1] A photon echo is a sharply directional coherent beam (spontaneous coherent emission) that contains information on the dynamics of optical quantum systems and on the external generators that illuminate the medium, and appears after two laser pulses are applied to a quantum system.[3,4] An analogous situation arises in the case when quantum systems are excited with hypersound and terasound ($\nu \sim 10^{12}$ sec^{-1}),[5] and also in the case of a combination of optical and acoustic exciting pulses.[6]

Multipulse spin-echo excitation has aroused considerable theoretical and experimental interest.[7–11] It appears that multipulse excitation should also be of interest in the region of photon and phonon[5] echo, all the more since experiments have already been performed on three-pulse excitation of signals of the photon-echo type.[12] In addition, the spiked structure of a giant laser pulse is a typical example of multipulse excitation.[13] This has made necessary a detailed theory of multipulse excitation, particularly with allowance for irreversible relaxation, or at least an initial treatment of the simplest and most graphic case. Such a theory is needed also for the analysis of problems connected with the development of spin memory elements.[14] Similar memory elements, using the phenomena of photon and phonon echo, are also possible.

Jaynes and Bloom[7] have considered different general matrix methods for the investigation of the solution of modified Bloch equations. They have also made the first attempt, to our knowledge, to analyze the general case of excitation of spin systems by an arbitrary number n pulses without allowance for irreversible relaxation. The methods considered in their papers, however,

 0038-5646/77/4506-1118$02.40

did not lead to concrete final results on n-pulse excitation and only preliminary remarks were made on this subject. Gryaznov and Chastnov[10] have considered theoretically multipulse excitation of photon echos with the aid of a single particular sequence of pulses, without allowance for irreversible relaxation. An approximate theory of irreversible damping of echo signals excited by certain specific multipulse sequences, with account taken of the Hamiltonian of the spin-spin interaction, was considered in[9].

The first indication that photon echo can be described by equations of the Bloch type is contained in[4]. It appears that it is precisely such a phenomenological theory of multiple excitation of echo signals, with allowance for irreversible relaxation, which should be the initial stage of the description, since it is simplest and most graphic, and admits of exact solutions, whereas an exact solution of this problem with allowance, say, for the Hamiltonian of the spin-spin interaction of a large number of particles, is utterly impossible. Besides, the Bloch equations and their solutions are highly symmetrical and are very simple.

It is obvious that to obtain detailed final results in the case of the excitation of spin systems by general sequences consisting of an arbitrary of n pulses it would be necessary either to supplement the methods considered in[7,9] or to use a different approach. In the present paper we develop a new general matrix method that leads to the indicated goal.

The results obtained in the present paper allow us to make an important physical conclusion: multipulse excitation of quantum systems is of great physical interest precisely from the point of view of the influence of the irreversible relaxation on the echo signals, since these numerous responses all depend in general differently on the time of the irreversible relaxation, and consequently each of them should carry new information on the processes and mechanisms of irreversible relaxation. In this paper we have adhered to the style of Jaynes and Bloom,[7] in order to make our results readily applicable also to spin echo.

1. SOLUTION OF FUNDAMENTAL EQUATIONS

The object of the investigation in the exposition that follows will be a system of N ($N=1, 2, \ldots$) identical noninteracting particles with a discrete multilevel equidistant nondegenerate energy spectrum. A generalization of the Dicke theory[1] was carried out by Solovarov and Nagibarov.[15] In this paper we investigate only one case out of all those considered in[15], namely transitions of the dipole type between neighboring levels of each particles. Just as in[15], we shall describe the spectrum of each particle with the aid of the concept of the energy spin[16] of the corresponding quantity R ($R = \frac{1}{2}, 1, \frac{3}{2}, 2, \ldots$). Using the energy-spin concept, we describe the considered system with the aid of a static Hamiltonian in the Schrödinger representation

$$\mathcal{H}_R=\mathcal{H}_0+\mathcal{H}', \quad \mathcal{H}_0=\sum_j \hbar\omega_0 R_{j3}, \quad \mathcal{H}'=\sum_j \hbar\Delta\omega_j R_{j3}, \qquad (1)$$
$$\omega_0>0, \quad j=1, 2, \ldots, N.$$

Here ω_0 is the angular frequency corresponding to a transition between neighboring levels of unperturbed particles, $\omega_0+\Delta\omega_j$ is the frequency of the perturbed j-th particle, while R_{j1}, R_{j2}, and R_{j3} are the operators of the cartesian components of the energy spin of the j-th particle, or more simply speaking, the usual well-known and well-investigated spin operators. In the exposition that follows we shall use the following universally employed complex linear combinations of spin operators:

$$R_{j(\gamma)}=|\gamma|R_{j(1)}+i\gamma R_{j2}+(1-|\gamma|)R_{j3}, \quad \gamma=0, \pm1. \qquad (2)$$

It is obvious that at the instant of time t the system (1) can be in a thermodynamic-equilibrium state described by a density matrix ρ in the Schrödinger representation

$$\rho=\prod_j \rho_j, \quad \rho_j=\exp[\zeta R_{j(0)}][\mathrm{Sp}\exp\zeta R_{j(0)}]^{-1}, \qquad (3)$$
$$j=1, 2, \ldots, N,$$

where $\zeta=-\hbar\omega_0/k_B T$, k_B is the Boltzmann constant, and T is the temperature. Assume that, starting with the instant of time $t=0$, the system (1) is acted upon after certain time intervals by n periodic coherent resonant pulses from several external generators of different type (optical, acoustic, etc.) with frequency ω_0.

To describe the system (1) on which n pulses act we introduce phenomenological spin operators $R_{j(\gamma)\,\mathrm{phen}}(t)$, specified by the following fundamental equations

$$\frac{\partial R_{j(\gamma)\,\mathrm{phen}}(t)}{\partial t}=\sum_{\gamma'=0,\pm1} H^{(j)}_{\gamma,\gamma'\,\mathrm{phen}}(t)R_{j(\gamma')\,\mathrm{phen}}(t)$$
$$+\left[1-\sum_{m=1}^{n}U_0(m)\right]\frac{1-|\gamma|}{T_{(\gamma)}}R_{j(\gamma)\,\mathrm{phen}}(0), \quad t\geq0, \qquad (4)$$
$$R_{j(\gamma)\,\mathrm{phen}}(0)=R_{j(\gamma)}, \quad \gamma=0,\pm1, \quad T_{(\pm1)}=T_2, \quad T_{(0)}=T_1,$$

where T_2 and T_1 are certain numerical parameters, $U_0(m)=U_-(t-t_{0m})-U_+(t-t_m)$, t_{0m} and t_m are the respectively instants of the beginning and end of the action of the m-th pulse on the system (1) (with $t_{01}=0$), and $U_-(x)$ $U_+(x)$ are asymmetrical unit step functions of real variable.[17] The matrix elements $H^{(j)}_{\gamma,\gamma'\,\mathrm{phen}}(t)$ contain quantities that take into account the wave character and the amplitude of the field, the constants that characterize the interaction of the field with the particles, the parameters T_1 and T_2, as well as the sums of the step functions that describe the multiplicity of the pulses.

We call attention to the fact that averaging (taking the trace) of the left-hand and right-hand parts of Eqs. (4) with the density matrix (c) yields equations of the Bloch type[18] for the mean values of the components of the energy spin. We note in this connection that it follows from the general theory of the equations of motion for mean values of functions of spin operators[19] that in real approximations the Bloch equations are valid not only for the spin $R=\frac{1}{2}$, but also for the spin $R=1$. After averaging (4), the parameter T_2 acquires a physical meaning of the time of irreversible relaxation of the mean values of transverse components of the energy spin, while T_1 acquires the physical meaning of the time of irreversible relaxation of the mean value of the longitudinal component of the energy spin.

Gantmakher[20] has shown that the solution of Eqs. (4) can be written in the form

$$R_{j(\gamma)\,\text{phen}}(t)=\sum_{\gamma'=0,\pm1}\Omega^{(j)}_{\gamma,\gamma'\,\text{phen}}(t,0)R_{j(\gamma')\,\text{phen}}(0)+\int_0^t\sum_{\gamma'=0,\pm1}\Omega^{(j)}_{\gamma,\gamma'\,\text{phen}}(t,t')f_{j(\gamma')}(t')\,dt',\quad t\geqslant0,$$

$$f_{j(\gamma')}(t)=\Big[1-\sum_{m=1}^{n}U_0(m)\Big]\frac{1-|\gamma'|}{T_{(\gamma')}}R_{j(\gamma')\,\text{phen}}(0),$$

$$R_{j(\gamma)\,\text{phen}}(0)=R_{j(\gamma)},\qquad \gamma,\gamma'=0,\pm1, \tag{5}$$

where the third-order matrix $\Omega^{(j)}_{\text{phen}}(t,t')$ is by way of an evolution operator and is a matrizant[20] usually written in the form of the universally employed symbolic exponential with the aid of a Dyson chronological operator for the time-dependent perturbation theory series.[21]

Using the properties of step functions,[17] the properties of the matrizant,[20] and the properties of the solution (5), we can write a final expression for $R_{j(\gamma)\text{phen}}(t)$ at the instants of time $t>t_n$ (t_n is the instant when the action of the last n-th exciting pulse on the system (1) terminates) in the following form:

$$R_{j(\gamma_n)\,\text{phen}}(t)=\exp(i\gamma_n\omega_0t)\sum_{\mu=0}^{n-1}\;\sum_{\gamma_\mu,\gamma_{\mu+1},\ldots,\gamma_{n-1}=0,\pm1}i^{\gamma_n-\gamma_\mu}\prod_{m=\mu+1}^{n}\exp\Big(-|\gamma_m|\frac{\tau_m}{T_2}\Big)$$

$$\times\exp\Big[-(1-|\gamma_m|)\frac{\tau_m}{T_1}\Big]\exp[i\gamma_m\varphi_{j(m)}]\alpha_{j(m)}^{\gamma_{m-1}-\gamma_m}A^{(m)}_{\gamma_m,\gamma_{m-1}}Q^{(\mu)}_{j(\gamma_\mu)}+Q^{(n)}_{j(\gamma_n)},$$

$$t\geqslant t_n,\qquad \gamma_n=0,\pm1;\quad m=\mu+1,\mu+2,\ldots,n. \tag{6}$$

In the right-hand side of (6) the operators are only the quantities Q. In addition (6) contains elements of the matrices $A^{(m)}$. The expressions for the elements of the matrices $A^{(m)}$ and for the operators Q can be written in the following symmetrical form:

$$2A^{(m)}_{1,\pm1}=\cos\theta_{j(m)}\pm1,\qquad A^{(m)}_{0,0}=\cos\theta_{j(m)},$$

$$A^{(m)}_{1,0}=-2A^{(m)}_{0,1}=\sin\theta_{j(m)},\qquad A^{(m)}_{-\gamma_m,-\gamma_{m-1}}=A^{(m)}_{\gamma_m,\gamma_{m-1}},$$

$$Q^{(0)}_{j(\gamma_0)}=R_{j(\gamma_0)},\qquad Q^{(\mu)}_{j(\gamma_\mu)}=\Big\{1-\exp\Big[-(1-|\gamma_\mu|)\frac{\tau_\mu}{T_1}\Big]\Big\}R_{j(\gamma_\mu)},$$

$$\gamma_m,\gamma_{m-1},\gamma_0,\gamma_\mu=0,\pm1;\quad m=1,2,\ldots,n;\quad \mu=1,2,\ldots,n. \tag{7}$$

In (6) and (7) we used the following notation:

$$\tau_m=t_{0m+1}-t_m\ \text{at}\ m<n,\qquad \tau_n=t-t_n\geqslant0,$$

$$\varphi_{j(m)}=\tau_m\Delta\omega_j,\qquad \theta_{j(m)}={}^1/_2|a_{j(m)}|\Delta t_m,\qquad \alpha_{j(m)}|a_{j(m)}|=a_{j(m)},$$

$$\Delta t_m=t_m-t_{0m},\qquad t_{0m+1}\geqslant t_m>t_{0m}\geqslant0,\qquad m=1,2,\ldots,n, \tag{8}$$

where $a_{j(m)}$ are complex numerical quantities that do not depend on the time and take into account the wave character and the field amplitude of the m-th pulse, and also constants that characterize the force of the interaction of the field with the particles.

By investigating (5) with the aid of the properties of step functions[17] and the properties of the matrizant[20] for instants of time t that satisfy the condition $t_n\geqslant t\geqslant0$, we can show that in the general case relation (6) is valid for instants of time t satisfying the condition $t_{0\nu+1}\geqslant t\geqslant t_\nu$ ($\nu=1,2,\ldots,n-1$), provided n in (6)–(8) is replaced by ν. Consequently, this method can be used to investigate those responses of the system (1) which appear in the course of its multipulse excitation, with allowance for irreversible relaxation, and which occur (in time) between the exciting pulses. It is this method that we shall use in the exposition that follows.

2. ECHO-SIGNAL POWER WITH ALLOWANCE FOR IRREVERSIBLE RELAXATION

We consider the case when the pulse acting on the system (1) constitute traveling plane waves. In this case the quantities $\theta_{j(m)}$ (8) are the same for all particles,[6] $\theta_{j(m)}=\theta_m$ ($m=1,2,\ldots,n$), and the quantities $\alpha_{j(m)}$ in (8) take the form

$$\alpha_{j(m)}=\beta_m\exp(i\mathbf{k}_m\mathbf{r}_j),\qquad |\mathbf{k}_m|=\omega_0/v_m,\qquad m=1,2,\ldots,n, \tag{9}$$

where $\mathbf{k}_m$ is the wave vector of the field of the m-th pulse in the sample, v_m is the absolute value of the phase velocity of the propagation of the m-th pulse in the sample, $\mathbf{r}_j$ is the radius vector of the mass center of the j-th particle, β_m is a complex constant that takes into account the initial phase of the field of the m-th pulse and does not depend on the index j.

We consider the radiation of the system (1), due to the spontaneous transitions of the dipole type between neighboring levels of each particle. The coherent part of the power of this radiation in a unit solid angle in the direction of the wave vector $\mathbf{k}$ at the instant of time $t>t_n$ can be obtained, when account is taken of (3) and (6), from the formula

$$I_{2\,\text{phen}}(\mathbf{k},t)=I_0(\mathbf{k})\sum_j\sum_{l\neq j}\exp[i\mathbf{k}(\mathbf{r}_j-\mathbf{r}_l)]\,\text{Sp}\,\rho_jR_{j(1)\text{phen}}(t)\,\text{Sp}\,\rho_lR_{l(-1)\,\text{phen}}(t),$$

$$t\geqslant t_n,\qquad |\mathbf{k}|=\omega_0/v,\qquad j,l=1,2,\ldots,N. \tag{10}$$

Here $I_{2\,\text{phen}}(\mathbf{k},t)$ is the coherent part of the radiation power (this part is proportional to N^2-N), $I_0(\mathbf{k})$ is the power that an isolated two-level (with spin $R=\frac{1}{2}$) particle in an excited-state radiates into a unit solid angle in the direction of $\mathbf{k}$,[1] and v is the absolute value of the phase velocity of the wave field of the spontaneous radiation of the particle system. With the aid of (6) we can easily investigate relation (10), even without writing it out in detail. It is obvious that in the general case, in order for the coherent part of the radiation power to reach maximal (macroscopic) values, it is necessary that the formula for $I_{2\,\text{phen}}(\mathbf{k},t)$ have some terms independent of the indices j and l in the expression that follows the two symbols of summation over the particles. In addition it is necessary that the exponential factors that contain the relaxation times T_0 and T_1 and describe the damping of these terms with time, $t\geqslant t_n$, be close to unity. All this takes place when, in the general case, the following conditions are simultaneously satisfied (for any one fixed aggregate of the values from $\gamma_1,\gamma_2,\ldots,\gamma_{n-1}=0,\pm1$):

$$t=t_n+\gamma_1\tau_1+\gamma_2\tau_2+\ldots+\gamma_{n-1}\tau_{n-1},\qquad t\geqslant t_n,$$

$$\mathbf{k}=\mathbf{k}_n+\gamma_1(\mathbf{k}_2-\mathbf{k}_1)+\gamma_2(\mathbf{k}_3-\mathbf{k}_2)+\ldots+\gamma_{n-1}(\mathbf{k}_n-\mathbf{k}_{n-1}),$$

$$\exp\Big[-\frac{2}{T_2}(t-t_n+|\gamma_1|\tau_1+|\gamma_2|\tau_2+\ldots+|\gamma_{n-1}|\tau_{n-1})\Big]\approx1,$$

$$\exp\left\{-\frac{2}{T_1}[(1-|\gamma_1|)\tau_1+(1-|\gamma_2|)\tau_2+\ldots+(1-|\gamma_{n-1}|)\tau_{n-1}]\right\}\approx 1, \qquad (11)$$

where at $n=1$ we should have $t=t_1$, $\mathbf{k}=\mathbf{k}_1$, $\exp[-2(t-t_1)/T_2]\approx 1$, and the exponential containing T_1 should be identically equal to unity. It must be borne in mind that if a set of values for $\gamma_1, \gamma_2, \ldots, \gamma_{n-1}$ is chosen in one of the relations (11), then only the same set of values can be substituted in all the remaining relations (11).

We note that relations (11) were obtained under the assumption that the following conditions are satisfied:

$$\tau_1, \tau_2, \ldots, \tau_{n-1} < T_1. \qquad (12)$$

In this case it is possible to disregard, as we did in (11), the terms containing factors of the type $1-\exp(-\tau_\mu/T_1)$ ($\mu=1, 2, \ldots, n$) and present in (10), since, first, they are not characteristic, second, the behavior of these factors is in a certain sense the opposite of the behavior of the exponential in (11), which contains the time of the irreversible relaxation T_1, and third, these factors can be close to zero when relations (11) and (12) are satisfied.

From relation (11) and from all the foregoing arguments it follows that n-pulse excitation causes the system (1) to emit superradiant coherent signals of the echo and free-induction type, characterized by appearance times and by wave vectors satisfying the relations (11). The damping (fall-off) of these signals in the course of time ($t \geq t_n$) as a result of the irreversible relaxation is described by exponential factors that contain the relaxation times T_1 and T_2 and enter in (11). It follows from (10) that the foregoing signals can be amplified to a definite limit or weakened until they vanish completely, by varying the quantities $\theta_1, \theta_2, \ldots, \theta_n$, that take into account the powers and durations of the exciting pulses.

Relations (11) contain the condition $t \geq t_n$, and consequently relations (11) determine the responses of the system (1) to n-pulse excitation that appears after the last n-th exciting pulse.

If τ_1 is less than any of the remaining time intervals between pulses, then the echo signal that is least influenced by the existence of the process of irreversible transverse relaxation described by the time T_2, corresponds in (11) generally to the condition $\gamma_1=1$, $\gamma_2=\gamma_3=\cdots=\gamma_{n-1}=0$. This response appears at the instant of time $t=t_n+\tau_1$ with a wave vector $\mathbf{k}=\mathbf{k}_n+\mathbf{k}_2-\mathbf{k}_1$ and attenuates (falls off) within a time $t\geq t_n$ in proportion to the factors

$$\exp\left(-2\frac{t-t_n+\tau_1}{T_2}\right), \quad \exp\left(-2\frac{\tau_2+\tau_3+\ldots+\tau_{n-1}}{T_1}\right). \qquad (13)$$

The influence exerted on this response by the irreversible longitudinal relaxation described by the time T_1 is seen here to be relatively strong. At $n\geq 3$ this response has a maximal power, if the conditions $\theta_1=\theta_2=\pi/2$, $\theta_3=\theta_4=\cdots=\theta_{n-1}=\pi$, $\theta_n=\pi/2$ are satisfied. The fact that such a response should really exist (be observable) can be seen from the following simple physical considerations.

After the first 90° pulse, maximum transverse components of the energy spin are produced, and consequently irreversible transverse relaxation should act in the time interval τ_1 between the first and second pulses. In the same time interval, the longitudinal components of the energy spin is equal to zero, so that the irreversible longitudinal relaxation should not act. The expressions agree with this fact, since they contain damping with a relaxation parameter T_2 in the time τ_1 and no damping with the parameter T_1. After the second 90° pulse and the succeeding 180° pulses, a maximum longitudinal component is produced, and consequently irreversible longitudinal relaxation should act in the intervals $\tau_2, \tau_3, \ldots, \tau_{n-1}$ between the second and third, the third and fourth, etc. In the same intervals, the transverse components are equal to zero, and consequently no irreversible transverse relaxation should act. The obtained formulas agree with this, since they contain damping with a parameter T_1 in the time $\tau_2+\tau_3+\cdots+\tau_{n-1}$ and no damping with parameter T_2. Finally, after after the last (n-th) 90° pulse, maximum transverse components appear, and consequently transverse relaxation should act, and the longitudinal component is equal to zero, and consequently the longitudinal relaxation should not act. The obtained formulas agree with this, since they contain damping with the parameter T_2 in a time $t-t_n\geq 0$ and no damping with the parameter T_1. This echo signal has no damping with the relaxation parameter T_2, with time intervals $\tau_2, \tau_3, \ldots, \tau_{n-1}$, but does have damping with the parameter T_2 with a time interval τ_1, and consequently the damping of this signal with parameter T_2 does not depend on the number of exciting pulses. We propose to use this echo signal for observation under multipulse excitation, when T_2 is very small and $T_1 \gg T_2$, and it is necessary to eliminate the influence of the irreversible transverse relaxation.

In the echo signal we encounter so to speak a "lengthening" of T_2 to T_1 at $T_2 \ll T_1$. We note that at $n=3$ this is the ordinary stimulated echo (SE) signal, while at $n>3$ it can be regarded as the multipulse analog of stimulated echo (MASE). Naturally, MASE can also be used to measure T_1. We note that MASE, unlike SE, vanishes if one of the θ_ν ($\nu=3, 4, \ldots, n=1$) is equal to $\pi/2$, i.e., if one of the intermediate ν-th pulses is a 90° pulse. It follows therefore that if an additional fourth 90° pulse is applied in the time interval τ_2, then the signal corresponding to the ordinary SE and appearing at a time τ_1 after the last exciting pulse, vanishes. We note that at $\theta_1=\theta_2=\pi/2$, $\theta_3=\theta_4=\cdots=\pi$, $\theta_n=\pi/2$ there is no MASE in the time interval between the pulses of this n-pulse exciting sequence.

One of the echo signals which is least effected by the existence of the irreversible longitudinal relaxation described by the time T_1 corresponds in (11) in the general case to the condition

$$\gamma_1=\gamma_3=\ldots=\gamma_{n-2}=-1, \quad \gamma_2=\gamma_4=\ldots=\gamma_{n-1}=1$$

when n is odd and to

$$\gamma_1=\gamma_3=\ldots=\gamma_{n-1}=1, \quad \gamma_2=\gamma_4=\ldots=\gamma_{n-2}=-1$$

when n is even. This response appears at the instant of time

$$t=t_n-\sum_{\nu=1}^{n-1}(-1)^{n+\nu}\tau_\nu \quad (14)$$

with a wave vector

$$\mathbf{k}=(-1)^{n+1}\mathbf{k}_1+2\sum_{\nu=2}^{n}(-1)^{n+\nu}\mathbf{k}_\nu. \quad (15)$$

This response attenuates (falls off) with time in proportion to the factor

$$\exp\left(-2\frac{t-t_n+\tau_1+\tau_2+\ldots+\tau_{n-1}}{T_2}\right). \quad (16)$$

The exponential that contains T_1 is equal to unity in this case. The influence of irreversible transverse relaxation described by the time T_2 on this response is seen to be the largest. This response has maximum power if the conditions $\theta_1=\pi/2$, $\theta_2=\theta_3=\cdots=\theta_n=\pi$ are satisfied, but this is none other than the usual sequence of the Carr and Purcell type,[8] and consequently, this echo signal can be used to measure the time T_2. The fact that such a response really exists can also be demonstrated with the aid of approximate simple physical considerations, analogous to those used for MASE. We note that the damping of the echo signal with T_1 does not depend on the number of exciting pulses, since there is no such damping altogether for arbitrary n.

We note also that at no responses is there any damping with a parameter T_1 with changing running time $t-t_n\geqslant 0$ in contrast to the damping with the parameter T_2.

In addition, after the n-th pulse there exists a free-induction signal that is altogether independent of T_2 at the initial instant of time, i.e., this signal can be observed even under the condition $T_1>\tau_1+\tau_2+\cdots+\tau_{n-1}\gg T_2$. This does not take place for echo signals, since the maximum intensities of the echo signals are always decreased as a result of the T_2 relaxation.

We call attention to the fact that the n-th exciting pulse is followed by responses that do not agree with the simple physical considerations analogous to those advanced for MASE.

We note that the irreversible relaxation processes have little effect at $t>2t_n$, for in the case of our series no responses appear at all at these times.

If the sequence of exciting pulses is such that all the intervals between these pulses are equal to zero ($\tau_1=\tau_2=\cdots=\tau_{n-1}=0$), and all these pulses are different in all other respects, then the maximum number $M_0^{(n)}$ of responses is determined by the relation

$$M_0^{(n)}=3^{n-1}. \quad (17)$$

These responses do not differ from one another in their appearance time, since they all appear at $t=t_n$ and are free-induction signals. However, if there are many different spontaneous-emission modes in the sample, then all these signals will differ from one another in the propagation direction. We note that in this case all the terms of (10) are realized, since the number of all the essentially different terms in (10) is also equal to 3^{n-1}. There are no "ghost" echo signals[22] in this case.

For one common sequence of exciting pulses, the maximum total number $M^{(n)}$ of responses, with allowance for the echo signals that appear in the interval between all pairs of neighboring exciting pulses, under the condition $\tau_\nu\geqslant\tau_1+\tau_2+\cdots+\tau_{\nu-1}\neq 0$ ($\nu=1,2,\ldots,n-1$), is determined by the following relation:

$$M^{(n)}=(3^n+2n+1)/4. \quad (18)$$

It follows from relation (11), (17), and (18) that by choosing a sequence of n pulses it is possible to obtain echo signals in arbitrary prescribed instants of time and to obtain emission of physical fields with arbitrary wave vector and with the required damping on account of the parameters T_1 and T_2.

We note that the total (integrated) coherent part of the power of the coherent spontaneous radiation of the system (1) can be obtained by integrating expression (1) for $I_{2\,\text{phen}}(\mathbf{k},t)$ over all the angles that determine the direction of the wave vector $\mathbf{k}$, for example, using the procedure employed in[4].

Obviously, the theory considered by us can describe real pulse excitation of a system of particles, if the following conditions are satisfied

$$\sum_{m=1}^{n}\Delta t_m<T_{2\,\text{rev}},\ \overline{T_2},\ \overline{T_1}, \quad (19)$$

where T_{rev} is the time of the reversible transverse relaxation due to the spread $\Delta\omega_j$ of the resonant frequencies. The restrictions (19) follow from the fact that in our theory (in particular, in Eqs. (4)) no account was taken of reversible and irreversible transverse and of irreversible longitudinal relaxation during the time of action of the exciting pulses on the system (1). They were taken into account only in the intervals between the pulses.

The relations obtained in this paper can help predict which of the sequences of the excited pulses will exert the strongest influence on the character of the spin-spin interaction (on the "spin-spin" Hamiltonian).[9] These relations can help select for each concrete substance and for each concrete experimental setup the exciting-pulse sequences that are most convenient for the experiment, and in particular for the measurement of the relaxation times T_1 and T_2, as was done in[8,11,23].

We note that the theory of multipulse excitation permits a better understanding of the echo phenomenon even in the two-pulse case. In addition, this theory provides a general approach to the echo phenomenon, since it not only makes it possible to obtain the solution for very large n, but also a general solution for $n=1$ and 2, which demonstrates the general regularities of the echo in general and of two-pulse echo in particular.

We point out that in the case of energy transitions between neighboring levels there should be no difference in principle between multilevel particles and two-level particles. The effect of many levels manifests itself only in that the right-hand side of (10) contains the value of the spin R. The calculation scheme and the matrix method used here to take into account the symmetry of the indices γ are applicable, however, also to the following: a) investigations of arbitrary transitions (in an equidistant spectrum), particularly those described by bilinear combinations of spin operators; b) in the case when the relaxation matrix is used[24] instead of phenomenological spin operators; c) in the investigation of the solution of the modified Bloch equations[7,23] with allowance for the diffusion term by the method of Das and Saha.[13,25]

In conclusion, I am grateful to U. Kh. Kopvillem and V. V. Samartsev for useful discussions, valuable remarks, and help with the work.

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Translated by J. G. Adashko