ACOUSTIC EXCITATION OF A SUPERRADIATIVE ELECTROMAGNETIC STATE IN MATTER

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The possibility of generating pulsed coherent physical fields at the expense of other coherent fields is investigated when the field frequencies are equal or multiples but their wave vectors are very different. It is shown that if the duration of the excited pulses and the interval between them does not exceed the phase memory time of the active particles of the working medium of the generator, the phase memory effects may "erase" the difference between the wave vectors of the exciting and generated fields. This new principle of transformation of wave vectors in echo signals is applied to the problem of excitation of electromagnetic waves by ultrasound. For this purpose, nuclear and electron spin systems can be transferred into a superradiative electromagnetic state by sound pulses. An experimental realization of these ideas should raise the method of magnetic quantum acoustics to the level of the electromagnetic spin echo technique and, as far as obtaining information on the structure and dynamics of matter is concerned, it should be considerably superior to the usual spin echo method.

THE superradiative electromagnetic state (SES) of a spin system and its creation with the aid of a coherent alternating magnetic field were first theoretically considered by Bloch[1]. In ordinary magnetic resonance[2], which is produced with the aid of stimulated electromagnetic transitions, the intensity of the effect is proportional to the number of active particles N in the sample, and depends strongly on the time of the longitudinal magnetic relaxation T_1. On the other hand, the SES signal is due to coherent spontaneous electromagnetic transitions, and its intensity is proportional to N^2 and depends strongly on the time of the transverse magnetic relaxation T_2. Bloch's ideas were substantially developed by Hahn[3], who showed that following a successive multipulse excitation of SES, it is possible to restore the SES after the cessation of the action of the external generators on the spin system, using only the internal forces of the spin system. This signal has been termed "spin echo," and its appearance is due to the fact that the spin system has "retained the memory" of two or more preceding exciting pulses. By the same token, Hahn introduced the concept of "spin memory" and proved that this phenomonon can be used to restore the coherence in a spin system. The question of which quantum systems and under the influence of which generators is it possible to obtain superradiative signals of the spin-echo type and of the induction type was solved in general form by one of the authors[4]. The answer to this question consisted in the fact that, in principle, any energy absorption process induced by a stimulating disturbance can be set in correspondence with a superradiative analog in the same sense that ordinary magnetic resonance can be set in correspondence with spin induction and echo. This prediction is confirmed at the present by the flow of reports of the discoveries of new superradiative analogs of stationary resonances[5-7] etc.

In the present paper we consider a new effect of "spin or phase memory." In this effect, by choosing the nature and the sequence of the pulsed generators exciting the quantum system, it is possible to create an efficient (in the sense of the radiation power) superradiative state of the system relative to any physical field with arbitrary wave vector. The question of the role of the ratio of the wave vectors of the exciting generators and of the investigated physical field, and also of the dimensions of the radiator and the length of the radiated wave in the determination of the efficiency of the produced superradiative state is not trivial[8]. In cases when the wave vectors taking part in the superradiative process are much larger than the reciprocal dimension of the radiator, several different modes α of superradiance are produced simultaneously under the influence of the coherent excitation, and their powers are proportional to $\beta_\alpha N^2$, whereas among the quantities β_α there is usually only one $\beta_{\alpha_0} \gg N^{-1}$, and greatly exceeds all other β_α. In particular, small values of β_α are always encountered in those cases when a certain coherent physical field $A(\omega, \mathbf{k}_A)$ with frequency ω and wave vector $\mathbf{k}_A$ is used to excite a coherent field $B(\omega, \mathbf{k}_B)$ at $|\mathbf{k}_A| \neq |\mathbf{k}_B|$. This difficulty can be circumvented by the method of multiquantum excitation[9], when $\mathbf{k}_e = \mathbf{k}_B$ for the effective multiquantum field. This method, however, may turn out to be ineffective as a result of the small probability of the multiquantum transitions.

A new situation arises in principle in cases when the coherent field $B(\omega, \mathbf{k}_B)$ is excited by a sequence of short pulses of the field $A(\omega, \mathbf{k}_A)$, which are separated by time intervals $\tau < T_2$ (T_2 - time of phase memory of the active particles). Among the obtained responses $B(\omega, \mathbf{k}_B)$ there exist in this case signals of the "echo" type, which "retain the memory" of the vector $\mathbf{k}_A^{(1)}$ of the first pulse and depend explicitly also on $\mathbf{k}_A^{(2)}$ of the second exciting pulse. We shall show here that the "memory" of $\mathbf{k}_A^{(1)}$ can be used in the "echo" signal to compensate for $\mathbf{k}_A^{(2)}$ or to produce an effective vector $m\mathbf{k}_A^{(1)} + n\mathbf{k}_A^{(2)} = \mathbf{k}_B$, where m and n are real numbers. This principle is applicable to fields of arbitrary nature and frequency. As a concrete example illustrating the foregoing considerations, we consider the excitation of a coherent electromagnetic field by pulses of elastic oscillations of a crystal lattice.

The idea of the possibility of exciting spin induction and echo with the aid of acoustic pulses was predicted by one of the authors (U, K) and was theoretically confirmed in[10]. The advantage of acoustic excitation of spin induction and echo over the electromagnetic excitation method lies in the fact that the elastic wave cannot directly interact with the spins, but only via a complicated chain of internal couplings, including the crystal field and the exchange and spin-orbit interactions. Therefore such a signal carries extensive information on the complicated details of the dynamics of the internal fields. The signal of the stationary acoustic magnetic resonance[11] has analogous properties, but it is presently universally accepted that the pulse procedure makes it possible to distinguish different dynamic processes, and is therefore more promising than stationary resonance.

Unfortunately, when the frequencies of the sound ν_A and of the radiated electromagnetic field ν_e are equal, the corresponding wave vectors are not equal $|\mathbf{k}_e| \ll |\mathbf{k}_A|$, and if the sample dimensions are $\alpha_0 N^{1/3} \gg |\mathbf{k}_A|^{-1}$ (α_0 is the lattice constant), then the spin system turns out to be practically "locked" after the acoustic excitation relative to the radiation of coherent electromagnetic waves and this, as noted by Kessel'[12], prevents the application of this method. In many papers[13], different variants of simultaneous and successive applications of electromagnetic and sound pulses were proposed, making it possible to avoid the indicated difficulty in part. In all these papers, the superradiance process is regarded semiclassically, thus greatly limiting the possible use of the obtained results in cases of high frequencies and weak excitation intensities. In addition, the case of excitation of quantum systems by traveling waves has not been considered. But it is precisely such waves which excite in identical manner large volumes of radiators, an important factor in the application of the principle of "erasing the difference between the wave vectors by applying the phase memory effect" in order to obtain powerful electromagnetic superradiance as a result of acoustic excitation.

In the present paper we derive formulas that take into account the quantum-mechanical character of the radiation process and are valid for any combinations of exciting pulses of standing and traveling sound waves. The most promising turned out to be the following variants of the pulsed acoustic magnetic spectroscopy: 1) Excitation of a spin system by two successive pulses that follow each other continuously, the first being acoustic and the other electromagnetic. It is convenient for the measurement of the elements of the spin-phonon interaction tensor, since it does not require an analysis of the causes of the resonance-line broadening. 2) Echo after two acoustic pulses under the condition that the form of the amplitude spectrum of the active particles changes in the interval between the pulses. It yields additional information concerning the nature of the local field and the coherent radiative acoustic damping of the induction and echo signals. 3) Experiments at low temperatures on half-wave plates or cylinders, the lengths of which span an odd number of half-waves.

In all our calculations it was assumed that the spin system does not react on the acoustic generator, and that prior to the perturbation the temperature of the spin system is $T \gg k_B^{-1} h\nu_S$, where k_B is the Boltzmann constant and ν_S is the frequency line width of the excited transition.

1. OBSERVED RESPONSES FOR AN EFFECTIVE SPIN R = ½

We consider the following problem. At the instant of time $t = 0$ a certain physical system consisting of N identical particles ($j, l, \ldots = 1, \ldots, N$) with Hamiltonian $\mathscr{H}_0 + \mathscr{H}'$ is in an equilibrium state described by a density matrix ρ_0. At the instant $t = 0$, a strong pulsed perturbation $\mathscr{H}_1$ is turned on for a time interval Δt_1. The system evolves subsequently during a time τ_S under the influence of the internal forces. At the instant $t = \tau_S$, a second strong perturbation is turned on in pulselike fashion for a time interval Δt_2. Finally, the system continues to evolve to a certain new state of equilibrium under the influence of the internal forces. For concreteness we have in mind the excitation of the spin system with the aid of a periodic coherent acoustic pulse. However, the developed formalism and the results are valid for any quantum system with an analogous spectrum and arbitrary pulse perturbations, described by the operator const $\cdot \mathscr{H}_1$ and const $\cdot \mathscr{H}_2$ (systems of electric dipoles, optical excitation, etc.).

We start with consideration of the case when the spins $R = 1/2$ are excited with the aid of an acoustic pulse and the operator of spin-phonon interaction is linear in R. The Hamiltonian of the problem is given by

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{H}' + \mathscr{H}_1 + \mathscr{H}_2;$$
$$\mathscr{H}_0 = \sum_{j=1}^{N} \hbar\omega_0 R_3{}^j, \quad \mathscr{H}' = \sum_{j=1}^{N} \hbar\Delta^j R_3{}^j,$$
$$\mathscr{H}_1 = \delta_1 \sum_j [\alpha_1 R_{k_1+}{}^j(t) + \alpha_1{}^* R_{k_1-}{}^j(t)],$$
$$\mathscr{H}_2 = \delta_2 \sum_j [\alpha_2 R_{k_2+}{}^j(t) + \alpha_2{}^* R_{k_2-}{}^j(t)], \tag{1}$$

Here

$$R_{k_\gamma\pm}^{(y)j}(t) = R_\pm^{(\gamma)j} P_\pm^{(y)(\gamma)}, \quad R_\pm{}^j = R_1{}^j \pm iR_2{}^j,$$
$$\Delta^j \lesssim (\Delta t_1)^{-1}, \quad (\Delta t_2)^{-1}, \quad \gamma = 1, 2, \quad y = \mathrm{I}, \mathrm{II};$$
$$P_\pm^{(\mathrm{I})(\gamma)} = \exp[\pm i(\mathbf{k}_\gamma \mathbf{r}^j - \omega_\gamma t)], \quad P_\pm^{(\mathrm{II})(\gamma)} = 2\cos(\mathbf{k}_\gamma \mathbf{r}^j)\exp(\mp i\omega_\gamma t);$$

ω_γ is the frequency of pulsed perturbation; $\hbar(\omega_0 + \Delta^j)$ is the distance between the levels of the j-th particle, R_1^j, R_2^j, and R_3^j are the Cartesian components of the effective spin R^j of the particle j; $\mathbf{r}^j$ is the radius vector of the particle j; $\mathbf{k}_\gamma$ is the wave vector of the propagation of the pulse excitation over the sample; δ_1 and δ_2 are real constants and α_1 and α_2 are complex constants; $R^{(\gamma)j}$ is a certain combination of the effective-spin components and pertains to j; the indices I and II pertain respectively to traveling and standing waves; α^* denotes the complex conjugate quantity; γ is the index designating the first and second pulses.

We calculate the mean values of the operators "Q" by means of the formulas

$$\langle\langle Q\rangle\rangle = \mathrm{Sp}\,\rho(t)Q, \quad \rho(t) = \mathscr{L}\rho_0\mathscr{L}^{-1},$$

$$\rho_0 = (\mathrm{Sp}\,Z)^{-1}Z, \; Z = \exp\left(-\frac{\mathscr{H}_0}{k_BT}\right),$$

$$\mathscr{L} = \exp\left[-i\sum_j \Delta^j(t-\tau_s)R_3{}^j\right]\exp\left[-i\delta_2\Delta t_2\sum_j(\alpha_2R_{k_2+}{}^j+\alpha_2{}^*R_{k_2-}{}^j)\right]$$
$$\times\exp\left[-i\sum_j\Delta^j\tau_sR_3{}^j\right]\exp\left[-i\delta_1\Delta t_1\sum_j(\alpha_1R_{k_1+}{}^j+\alpha_1{}^*R_{k_1-}{}^j)\right], \quad (2)$$

where $\mathscr{L}$ is the evolution operator, and $R^j_{k_1\pm}$ and $R^j_{k_2\pm}$ are obtained respectively from $R^j_{k_1\pm}(t)$ and $R^j_{k_2\pm}(t)$ by the substitution $\omega_\gamma = 0$.

We obtain the mean values of the operators Q of the following observables following the two-pulse action:

$$\mu^{(\xi)}_{(y)1} = \frac{1}{2}\langle\langle R_+e^{i\omega_0t}+R_-e^{-i\omega_0t}\rangle\rangle g_\perp\beta_0,$$

$$\mu^{(\xi)}_{(y)2} = \frac{1}{2i}\langle\langle R_+e^{i\omega_0t}-R_-e^{-i\omega_0t}\rangle\rangle g_\perp\beta_0,$$

$$\mu^{(\xi)}_{(y)3} = \langle\langle R_3\rangle\rangle g_\parallel\beta_0, \quad I^{(\xi)}_{(y)} = \int d\Omega I^{(\xi)}_{(y)}(\mathbf{k}), \quad I^{(\xi)}_{(y)}(\mathbf{k}) = I_0(\mathbf{k})\langle\langle R^{(y)}_{k+}R^{(y)}_{k-}\rangle\rangle^{(\xi)},$$
$$I_0(\mathbf{k}) = \frac{\omega^4}{2\pi c^3}g_\perp{}^2\beta_0{}^2\sin^2\theta, \quad R_\pm = R_1\pm iR_2, \quad R_{\xi_0} = \sum_j R_{\xi_0}{}^j,$$
$$R^{(\mathrm{I})}_{k\pm} = \sum_j R_\pm{}^j\exp(\pm i\mathbf{k}\mathbf{r}^j), \quad R^{(\mathrm{II})}_{k\pm} = 2\sum_j R_\pm{}^j\cos\mathbf{k}\mathbf{r}^j, \quad (3)$$

where β_0 is the magneton, ξ the index designating the nature of the physical action, $g_\parallel$ and $g_\perp$ the spectroscopic splitting factors, and θ the angle between $\mathbf{k}$ and the dipole oscillation direction.

Here $\mu^{(\xi)}_{(y)\eta}$ ($\eta = 1, 2, 3$) are the macroscopic components of the magnetization; the transverse oscillating components determine the intensities of the spin induction and echo signals (see[14], formula III.63) $I^{(\xi)}_{(y)}$ is the integral power of the spontaneous electromagnetic radiation (coherent and incoherent) of the spin system, namely $I^{(\xi)}_{(\mathrm{I})}$ if the system radiates a traveling wave, and $I^{(\xi)}_{(\mathrm{II})}$ if the system radiates a standing wave; $I^{(\xi)}_{(y)}(\mathbf{k})$ is the radiation power with wave vector $\mathbf{k}$ in a unit solid angle; $I_0(\mathbf{k})$ is the coefficient for the dipole radiation ([15], p. 209). If the external pulsed interaction has excited only one of two transverse magnetization components, then the coherent part of $I^{(\xi)}_{(y)}$ (i.e., the one proportional to N^2) must be multiplied by $1/2$. This coherent part $\hat{I}^{(\xi)}_{(y)}$ causes the radiative decay of the induction signal in accordance with the law

$$\hat{I}^{(\xi)}_{(y)}(t) = I_r\,\mathrm{sech}(\xi_0t), \quad \xi_0 = \hat{I}^{(\xi)}_{(y)}(N\hbar\omega_0)^{-1}, \quad (4)$$

where I_r is the radiation power directly after the turning off of the pulse $\mathbf{k}_1$, which turns the magnetization through $\pi/2$. In the case of the echo signal, I_r denotes the radiation power at the instant $t = 2\tau_s$ after turning off the pulse $\mathbf{k}_2$ and rotation of the magnetization through $3\pi/2$ (it is then necessary to write $t - 2\tau_s$ in lieu of t). If the substance is placed in an electromagnetic resonator with quality factor Q, then

$$I_0(\mathbf{k}) = 4\pi^2Q\omega_0g_\perp{}^2\beta_0{}^2V^{-1},$$

where V is the volume of the resonator (see[14] p. 248).

Let the particle j have $2R_0 + 1$ energy levels, among which there is only one pair of levels $E_b \gg E_a$ with interval $E_b - E_a = \hbar\omega_0$. The subsystem of levels E_a and E_b, obviously, contains $N(T)$ particles:

$$N(T) = N\left[\exp\left(-\frac{E_a}{k_BT}\right)+\exp\left(-\frac{E_b}{k_BT}\right)\right]\left[\sum_{\xi=1}^{2R_0+1}\exp\left(-\frac{E_\xi}{k_BT}\right)\right]^{-1}.$$

When $R = 1/2$, we can assume with sufficient degree of generality that $\omega_\gamma = \omega_0$, and

$$R_\pm^{(\gamma)j} = R_\pm{}^j, \quad R^j_{k_\gamma\pm} = R_\pm{}^j\exp(\pm i\mathbf{k}_\gamma\mathbf{r}^j), \quad \gamma = 1, 2.$$

If the perturbations $\mathscr{H}_\gamma$ are of the form

$$\mathscr{H}_\gamma = \frac{1}{4}\sum_j\Big\{R_1{}^j\sum_{\alpha,\beta,\delta}H_\alpha G_{1\alpha\beta\delta}\varepsilon_{\beta\delta}$$
$$+R_2{}^j\sum_{\alpha,\beta,\delta}H_\alpha{}'G'_{2\alpha\beta\delta}\varepsilon_{\beta\delta}{}'\Big\}^{(\gamma)}\cos(\mathbf{k}_\gamma\mathbf{r}^j-\omega_0t), \quad (5)$$

where $\alpha, \beta, \delta = 1, 2, 3$, we have for $\Delta t_\gamma \gg \omega_0^{-1}$

$$\delta_\gamma = \hbar^{-1}, \quad \alpha_\gamma = \frac{\Delta t_\gamma}{4}\sum_{\alpha,\beta,\delta}\{H_\alpha G_{1\alpha\beta\delta}\varepsilon_{\beta\delta}-iH_\alpha{}'G'_{2\alpha\beta\delta}\varepsilon_{\beta\delta}{}'\}^{(\gamma)},$$

where H_α and H'_α are the components of the static external magnetic field, $\epsilon_{\beta\delta}$ are the elements of the relative deformation tensor, $G_{1\alpha\beta\delta}$ are the elements of the spin-phonon interaction tensor[16]; $\{\,\}^{(\gamma)}$ denotes that all the quantities in this bracket pertain to the pulse γ.

According to (3) and (5), we get

$$\mu^{(1)}_{(\mathrm{I})3} = -2a_1b_1[N(T)\cos\theta_1\cos\theta_2-\mathrm{Re}\,c_1]g_\parallel\beta_0,$$

$$\mu^{(1)}_{(\mathrm{I})2} = -a_1b_1g_\perp\beta_0\,\mathrm{Im}\,d_1, \quad \mu^{(1)}_{(\mathrm{I})1} = -a_1b_1g_\perp\beta_0\,\mathrm{Re}\,d_1;$$

$$a_1 = \frac{R(R+1)}{3}, \quad b_1 = \mathrm{th}\frac{\hbar\omega_0}{2k_BT},$$

$$c_1 = \frac{\alpha_1\alpha_2{}^*}{|\alpha_1|\,|\alpha_2|}\sum_j\exp\{i[(\mathbf{k}_1-\mathbf{k}_2)\mathbf{r}^j-\Delta^j\tau_s]\}\sin\theta_1\sin\theta_2;$$

$$d_1 = \sum_{j=1}^{N(T)}(d_{11}{}^j+d_{12}{}^j+d_{13}{}^j)ie^{i\omega_0t},$$

$$d_{11}{}^j = \frac{\alpha_1{}^*}{|\alpha_1|}\exp[-i(\mathbf{k}_1\mathbf{r}^j-\Delta^jt)]\sin\theta_1(1+\cos\theta_2),$$

$$d_{12}{}^j = 2\frac{\alpha_2{}^*}{|\alpha_2|}\exp\{-i[\mathbf{k}_2\mathbf{r}^j-\Delta^j(t-\tau_s)]\}\cos\theta_1\sin\theta_2,$$

$$d_{13}{}^j = -\frac{\alpha_1(\alpha_2{}^*)^2}{|\alpha_1|\,|\alpha_2|^2}$$
$$\times\exp\{-i[(2\mathbf{k}_2-\mathbf{k}_1)\mathbf{r}^j-\Delta^j(t-2\tau_s)]\}\sin\theta_1(1-\cos\theta_2),$$

$$\theta_\xi = 2\delta_\xi\Delta t_\xi|\alpha_\xi| \quad (6)$$

and

$$\langle\langle R^{(1)}_{k+}R^{(1)}_{k-}\rangle\rangle^{(1)} = a_1N(T)\Big\{2-2b_1[\cos\theta_1\cos\theta_2-N^{-1}(T)\,\mathrm{Re}\,c_1]$$
$$+\frac{1}{4}a_1b_1{}^2\sum_{j\neq l}^{N(T)}(d_{11}{}^j+d_{12}{}^j+d_{13}{}^j)(d_{11}{}^{l*}+d_{12}{}^{l*}+d_{13}{}^{l*})\cdot$$
$$\times\exp[i\mathbf{k}(\mathbf{r}^j-\mathbf{r}^l)]N^{-1}(T)\Big\}. \quad (7)$$

From a comparison of the expressions $P_{\pm}^{(I)(\gamma)}$ and $P_{\pm}^{(II)(\gamma)}$ in (1) it follows that in the case of excitation of SES by standing sound waves, the form of relation (6) remains the same. It is also necessary to modify the definitions of the quantities c_1, d_{11}^j, d_{12}^j, d_{13}^j, and θ_ξ:

$$c_2{}^j = \frac{\alpha_1\alpha_2{}^*}{|\alpha_1||\alpha_2|}\exp(-i\Delta^j\tau_s)\sin\theta_1{}^j\sin\theta_2{}^j,$$

$$d_{21}{}^j = \frac{\alpha_1{}^*}{|\alpha_1|}\exp(i\Delta^j t)\sin\theta_1{}^j(1+\cos\theta_2{}^j),$$

$$d_{22}{}^j = 2\frac{\alpha_2{}^*}{|\alpha_2|}\exp[i\Delta^j(t-\tau_s)]\cos\theta_1{}^j\sin\theta_2{}^j,$$

$$d_{23}{}^j = -\frac{(\alpha_1)(\alpha_2{}^*)^2}{|\alpha_1||\alpha_2|^2}\exp[i\Delta^j(t-2\tau_s)]\sin\theta_1{}^j(1-\cos\theta_2{}^j),$$

$$\theta_\xi{}^j = 4\delta_\xi\Delta t_\xi|\alpha_\xi|\cos(\mathbf{k}_\xi\mathbf{r}^j). \qquad (8)$$

In place of (7) we obtain

$$\langle\!\langle R_{k+}^{(II)}R_{k-}^{(II)}\rangle\!\rangle^{(2)} = a_1N(T)\left\{\left[2-2b_1N^{-1}(T)\sum_{j=1}^{N(T)}(\cos\theta_1{}^j\cos\theta_2{}^j - \operatorname{Re}c_2{}^j)\right.\right.$$

$$\left.\times 4\cos^2(\mathbf{k}\mathbf{r}^j)\right] + a_1b_1{}^2\sum_{j\neq l}^{N(T)}(d_{21}{}^j+d_{22}{}^j+d_{23}{}^j)(d_{21}{}^{l*}+d_{22}{}^{l*}+d_{23}{}^{l*})$$

$$\left.\times N^{-1}(T)\cos(\mathbf{k}\mathbf{r}^j)\cos(\mathbf{k}\mathbf{r}^l)\right\}. \qquad (9)$$

If the first pulse is due to standing waves and the second to traveling waves and the radiation power of the traveling waves with wave vector $\mathbf{k}$ is determined then it is necessary to use formula (7) with $d_{1\eta}^j$ and c_1 replaced respectively by

$$d_{31}{}^j = \frac{\alpha_1{}^*}{|\alpha_1|}\exp[i\Delta^j t]\sin\theta_1{}^j(1+\cos\theta_2),$$

$$d_{32}{}^j = 2\frac{\alpha_2{}^*}{|\alpha_2|}\exp\{-i[\mathbf{k}_2\mathbf{r}^j - \Delta^j(t-\tau_s)]\}\cos\theta_1{}^j\sin\theta_2,$$

$$d_{33}{}^j = -\frac{\alpha_1(\alpha_2{}^*)^2}{|\alpha_1||\alpha_2|^2}\exp\{-i[2\mathbf{k}_2\mathbf{r}^j - \Delta^j(t-2\tau_s)]\}\sin\theta_1{}^j(1-\cos\theta_2),$$

$$c_3{}^j = \frac{\alpha_1\alpha_2{}^*}{|\alpha_1||\alpha_2|}\sum_j\exp\{-i[\mathbf{k}_2\mathbf{r}^j + \Delta^j\tau_s]\}\sin\theta_1{}^j\sin\theta_2. \qquad (10)$$

If the first pulse is due to standing waves and the second to traveling waves, but the radiation energy is in the form of standing waves with wave vector $\mathbf{k}$, then it is necessary to use (9) with the corresponding quantities from (10) replacing c_2^j and $d_{2\eta}^j$.

Let the first pulse be due to traveling waves and the second to standing waves. Then the energy radiation power in the form of traveling and standing waves can be calculated, as in the preceding cases, using the parameters

$$d_{41}{}^j = \frac{\alpha_1{}^*}{|\alpha_1|}\exp[-i(\mathbf{k}_1\mathbf{r}^j - \Delta^j t)]\sin\theta_1(1+\cos\theta_2{}^j),$$

$$d_{42}{}^j = 2\frac{\alpha_2{}^*}{|\alpha_2|}\exp[-i\Delta^j(\tau_s - t)]\cos\theta_1\sin\theta_2{}^j,$$

$$d_{43}{}^j = -\frac{\alpha_1(\alpha_2{}^*)^2}{|\alpha_1||\alpha_2|^2}\exp\{i[\mathbf{k}_1\mathbf{r}^j + \Delta^j(t-2\tau_s)]\}\sin\theta_1(1-\cos\theta_2{}^j),$$

$$c_4{}^j = \frac{\alpha_1\alpha_2{}^*}{|\alpha_1||\alpha_2|}\sum_j\exp\{i[\mathbf{k}_1\mathbf{r}^j - \Delta^j\tau_s]\}\sin\theta_1\sin\theta_2{}^j. \qquad (11)$$

It is easy to see how to modify the formulas to calculate $\mu_{(y)\eta}^{(\xi)}$ in the case of excitation with the aid of standing waves, or in mixed cases.

We shall continue the analysis of formulas (6)–(10) for the case $|\mathbf{k}|l \ll 1$, where l describes the linear dimensions of the sample. For example, this inequality is satisfied when electromagnetic waves of frequency 10^9 Hz are excited by hypersound, when $|\mathbf{k}| = 2\pi/30\ \mathrm{cm}^{-1}$ and $l = 1$ cm. The results of the analysis remain qualitatively valid when $l \le \lambda/4$, where λ is the length of the radiated electromagnetic waves.

Formulas (7) and (9) contain single- and two-particle parts, which describe respectively spontaneous incoherent and coherent radiation. The single-particle part of the formula for the radiation power is independent of the summation over the particles in only one case (i.e., it is obtained by multiplication of the single-particle contribution by $N(T)$), namely when the processes of excitation and radiation are realized only with the aid of traveling waves. From among the quantities $\mu_{(y)\eta}^{(\xi)}$, only $\mu_{(I)3}^{(\xi)}$ contains a part that does not depend on the index of the particle j (case of excitation by traveling waves).

The single-particle responses, which depend on the factor c_α ($\alpha = 1, \ldots, 4$), are of interest. It is seen from (7) and (9) that the power of the single-particle spontaneous radiation at $\theta_1 = \pi/2$ or $\theta_1^j = \pi/2$ is determined by the term with the factor c_α. For $\tau_s = 0$, using the corresponding two-pulse excitation technique, we can obtain responses of spontaneous radiation $c_\alpha \sim 0$ and $c_\alpha \sim N(T)$. In the former case the system is "locked" relative to the spontaneous single-particle radiation. The second case can be realized for c_1 by choosing $\mathbf{k}_1 = \mathbf{k}_2$, and for c^j by choosing $4\delta_\xi\Delta t_\xi|\alpha_\xi| < \pi/2$. The cases c_3^j and c_4^j are not suitable for obtaining powerful spontaneous radiation.

The two-particle part in (7) or (9) contains $N(T)[N(T) - 1]$ terms, and its order of magnitude is determined by the factor $\kappa[N(T)]^2$, $\kappa \ll 1$. The quantity κ has a maximum value if the terms in the double sum are not alternating in sign. If at least one of the exciting pulses is applied in the form of a standing wave, $\kappa \ll 1$, and there is no powerful coherent electromagnetic radiation.

In the case when the system is excited with two pulses in the form of traveling waves, powerful coherent radiation may result from the term $d_{13}^j d_{13}^{l*}$. It is described by the formula

$$\langle\!\langle R_{k+}^{(I)}R_{k-}^{(I)}\rangle\!\rangle_{(\mathrm{coh})}^{(1)} = \frac{1}{4}a_1{}^2b_1{}^2\sum_{j\neq l}^{N(T)}d_{13}{}^jd_{13}{}^{l*}\exp[i\mathbf{k}(\mathbf{r}^j-\mathbf{r}^l)]. \qquad (12)$$

According to (6), when $2\mathbf{k}_2 = \mathbf{k}_1$, $t = 2\tau_s$, and $|\mathbf{k}|l \ll 1$, we get from (12)

$$\langle\!\langle R_{k+}^{(I)}R_{k-}^{(I)}\rangle\!\rangle_{(\mathrm{coh})}^{(1)} = {}^1\!/\!_4 a_1{}^2b_1{}^2[\sin\theta_1(1-\cos\theta_2)]^2[N(T)]^2 \qquad (13)$$

It follows from (13) that to excite a powerful electromagnetic radiation with the aid of ultrasound within a time $\tau_s < T_2$ it is necessary to decrease by a factor of 2 the intensity of the static magnetic field and to apply a second sound pulse at half the frequency relative to the first pulse, where T_2 is the time of the irreversible phase relaxation of the transverse magnetization components. Apparently such experiments can be realized in nuclear spin systems[17]. It is seen from (9) that in the case of excitation of a system by two pulses in the form of traveling waves it is possible to obtain a powerful electromagnetic response in the form of standing waves.

Of considerable interest are experiments in which the first pulse is obtained from an acoustic generator and the second from an electromagnetic one (i.e., $|\mathbf{k}_2| l \ll 1$). In such experiments, interest attaches to the signal at the instant $t = \tau_S$, which is described by the formula

$$\langle\langle R_{k+}^{(I)} R_{k-}^{(I)} \rangle\rangle_{e}^{(1)} = a_1^2 b_1^2 (\cos\theta_1 \sin\theta_2)^2 [N(T)]^2. \tag{14}$$

For excitation by standing waves, we obtain from (9) the expression

$$\langle\langle R_{k+}^{(II)} R_{k-}^{(II)} \rangle\rangle_{\gamma}^{(2)} = a_1^2 b_1^2 \sin 2\theta_2 [N(T)]^{-1} \sum_{j\neq l} \cos\theta_1^j \cos\theta_1^l, \tag{15}$$

which for $4\delta_\xi \Delta T_\xi |\alpha_\xi| < \pi/2$ describes a powerful coherent electromagnetic response.

Thus, a two-pulse excitation, in which the first pulse has an acoustic nature and the second an electromagnetic nature, makes it possible to extract the information contained in the factors $\cos\theta_1$ or $\cos\theta_1^j$ concerning the spin-phonon interaction.

Let us consider the case when the sample has the form of a cylinder with axis z, and the ends of the cylinder are plane-parallel and polished with an accuracy much greater than the wavelength of the sound λ_1. If this sample is excited by an acoustic pulse in the form of a traveling wave with $\mathbf{k}_1 \parallel z$ at the magnetic-resonance frequency, then at a cylinder length $l = (2n+1)\lambda_1/2$ ($n = 0, 1, \ldots$) the sample will radiate electromagnetic waves with power $I \sim N_1^2$, where N_1 is the number of particles in the volume $S_0\lambda_1/2$ (S_0 is the area of the end face). This signal is weaker by a factor of approximately 10–100 than the observed nuclear and electron spin induction signals at room temperatures. However, at helium temperatures it can be completely observed in ordinary installations. Observation of this signal is of great interest, since, in accordance with the results of[18], the acoustic pulse transfers the sample to the superradiative state relative to generation of sound waves λ_1, the intensity of sound generation being $I_S \propto N^2$, where N is the number of magnetic particles in the entire volume of the sample. As shown in[18], the damping of the magnetic-induction signal under such conditions is no longer determined by the spin-spin and inhomogeneous mechanisms, but by the coherent spontaneous acoustic radiative damping. Such an effect can be readily observed by comparing the forms of the signals of the ordinary spin induction in acoustic and electromagnetic excitations.

2. EXCITATION OF A SPIN SYSTEM WITH THE AID OF A QUADRUPOLE TYPE INTERACTION

If the spin is $R > 1/2$, then the operators $R_{\pm}^{(\gamma)j}$ may turn out to be quadratic in the spin[16]. In this case $\mathscr{H}_0$ and $\mathscr{H}'$ retain the same form as before, but a scatter of the local field, which is quadratic in the spin, is added. The evolution operator $\mathscr{L}$ from (2) assumes the form

$$\mathscr{L}_1 = \exp[-i(t-\tau_s)(\Delta_1^j R_3^j + \Delta_2^j R_3^{j2})]$$
$$\times \exp\left[-i\frac{\theta_2}{2}(\beta_2^j R_+^{j2} + \beta_2^{j*} R_-^{j2})\right]$$
$$\times \exp[-i\tau_s(\Delta_1^j R_3^j + \Delta_2^j R_3^{j2})]$$
$$\times \exp\left\{-i\frac{\theta_1}{2}[\beta_1^j [R_3^j, R_+^j]_+ + \beta_1^{j*} [R_3^j, R_-^j]_+]\right\},$$

$$\mathscr{L}_2 = \exp\left\{-i\frac{\theta_1}{2}[\beta_1^j [R_3^j, R_+^j]_+ + \beta_1^{j*} [R_3^j, R_-^j]_+]\right\}, \tag{16}$$

where $\mathscr{L}_1$ and $\mathscr{L}_2$ describe respectively the excitation of the signals of the echo and induction type, $[\,,]_+$ is the anticommutator sign, $\theta_\xi = \hbar^{-1}\Delta t_\xi$ ($\xi = 1, 2$), and Δ_2^j is the parameter of the quadrupole scatter of the local field.

The acoustic perturbation acting in the time intervals Δt_1 and Δt_2 can be accordingly written in the form

$$\mathscr{H}_1^* = \sum_j [\beta_1^j [R_3^j, R_+^j]_+ + \beta_1^{j*} [R_3^j, R_-^j]_+],$$
$$\mathscr{H}_2^* = \sum_j (\beta_2^j R_+^{j2} + \beta_2^{j*} R_-^{j2}),$$
$$\beta_1 = \frac{1}{2} \sum_{\alpha,\beta} \varepsilon_{\alpha\beta}(G_{\alpha\beta zx} - iG_{\alpha\beta zy}),$$
$$\beta_2 = \frac{1}{4} \sum_{\alpha,\beta} \varepsilon_{\alpha\beta}(G_{\alpha\beta xx} - G_{\alpha\beta yy} - 2iG_{\alpha\beta xy}). \tag{17}$$

Unlike the case $R = 1/2$, calculations with the aid of formulas (16) and (17) are very cumbersome and lead to complicated formulas, and the calculations must be performed separately for each spin. All the qualitative considerations advanced in the preceding section remain in force. However, there are certain essential differences between the results obtained for integer and half-integer spins. Substitution of (16) and (17) in (3) leads to a result on the basis of which we can draw the following general conclusions.

1. In the case of integer and half-integer spins R at $\Delta_1^j \neq 0$ and $\Delta_2^j \neq 0$ (i.e., the values of both the local magnetic field and electric field are subject to scatter over the volume of the crystal), it is possible to obtain SES of the spin system with the aid of two successive acoustic pulses for $t = \tau_S$ (i.e., the electromagnetic signal appears immediately after the termination of the action of the second pulse). For half-integer R we should have $\tau_S = 0$ (i.e., the pulses must follow one another continuously), and for integer R the distance τ_S between pulses should be smaller than the phase relaxation time T_2. For half-integer R, the intensity of the signal depends on the factor $\exp[i(\mathbf{k}_1 - \mathbf{k}_2)(\mathbf{r}^j - \mathbf{r}^l)]$, which will not depend on $\mathbf{r}^j - \mathbf{r}^l$ at $\mathbf{k}_1 = \mathbf{k}_2$ (i.e., particles in the entire volume take part in the radiation process). On the other hand, in the case of integer spin, this factor has the form $\exp[i\mathbf{k}_2(\mathbf{r}^j - \mathbf{r}^l)]$, and only the particles in a layer with half-wave thickness make a contribution to the signal.

2. If the two types of local signals are not present simultaneously, then additional signals appear. At $\Delta_j^j \neq 0$ and $\Delta_2^j = 0$, a signal appears for half-integer spin $R = 5/2$ at the instant $t = 2\tau_S$, with a factor $\exp[i(\mathbf{k}_2 - \mathbf{k}_1)(\mathbf{r}^j - \mathbf{r}^l)]$, and for an integer spin $R = 2$ at the instant $t = \tau_S$ there appear additional terms with new combinations of the wave vectors in the exponential. These new combinations are of the form: 1) $3\mathbf{k}_1 - 2\mathbf{k}_2$, 2) $3\mathbf{k}_1 - 4\mathbf{k}_2$, 3) $\mathbf{k}_1$, 4) $\mathbf{k}_1 - 2\mathbf{k}_2$, 5) $2\mathbf{k}_1 - \mathbf{k}_2$, and 6) $3\mathbf{k}_2 - 2\mathbf{k}_1$. For $\Delta_1^j = 0$ and $\Delta_2^j \neq 0$,

we obtain the following new terms and wave-vector combinations.

Integer spin R = 2 (we give the instant of appearance of the signal and the combinations of the wave vectors for this signal):

a) $t = 0$; $3\mathbf{k}_1 - 2\mathbf{k}_2$, $3\mathbf{k}_1 - 4\mathbf{k}_2$, $\mathbf{k}_1$, $2\mathbf{k}_2 - \mathbf{k}_1$;
b) $t = {}^4/_3\tau_s$; $\mathbf{k}_1$, $2\mathbf{k}_2 - \mathbf{k}_1$;
c) $t = 2\tau_s$; $3\mathbf{k}_1 - 2\mathbf{k}_2$, $4\mathbf{k}_2 - 3\mathbf{k}_1$, $\mathbf{k}_1$, $2\mathbf{k}_2 - \mathbf{k}_1$;
d) $t = {}^7/_3\tau_s$; $2\mathbf{k}_1 - \mathbf{k}_2$, $3\mathbf{k}_2 - 2\mathbf{k}_1$;
e) $t = 4\tau_s$; $3\mathbf{k}_1 - 2\mathbf{k}_2$, $4\mathbf{k}_2 - 3\mathbf{k}_1$, $\mathbf{k}_1$, $2\mathbf{k}_2 - \mathbf{k}_1$;
f) $t = 5\tau_s$; $2\mathbf{k}_1 - \mathbf{k}_2$, $3\mathbf{k}_2 - 2\mathbf{k}_1$.

Half-integer spin $R = {}^5/_2$:

a) $t = {}^3/_2\tau_s$; $\mathbf{k}_2 - \mathbf{k}_1$;
b) $t = 2\tau_s$; $\mathbf{k}_1$.

The experiments described in this paper can be performed with standard nuclear quadrupole echo apparatus, supplemented with a generator of short acoustic pulses. The pulse duration should be of the order of $\Delta t \sim 10^{-3} - 10^{-6}$ sec. The amplitude of the relative deformation ϵ, due to the sound wave, should satisfy the condition $\epsilon \geq \pi\hbar/G\Delta t$, where $G \sim 10^{-18}$ erg per deformation unit is the nuclear spin-phonon interaction constant[16], i.e., $\epsilon \sim 10^{-3} - 10^{-6}$. To excite electron spin echo with hypersound, it is necessary to have the same values of ϵ, since the pulses required are shorter by a factor 10^3, but the values of G for the ions are 10^3 times larger than for nuclei. Such short acoustic pulses can be obtained with the aid of laser light pulses[19].

We have considered also cases of arbitrary spin R for an interaction Hamiltonian linear in the spin[20], and spins equal to 1, $^3/_2$, and 2 in the case of an interaction bilinear in the spin[21]. On the basis of these data we can draw the following conclusions.

1. Experiments on the generation of coherent electromagnetic waves with the aid of coherent acoustic pulses are possible on existing spin-echo installations, using either half-wave plates or the method of compensation of the wave vectors in the echo signal.

2. The induction and echo signals give detailed information concerning the exponent n in the local-field operator $\Delta^j (R_3^j)^n$.

3. With increasing spin R and with increasing rank of the irreducible spin tensor operators, the number of instants of time when echo signals are produced increases sharply. It is very important that the time interval prior to the occurrence of the last echo increases simultaneously. This makes it possible to lengthen the time during which it is possible to modify the spectrum of the unperturbed system.

4. Integer spins R give many more different types of responses than half-integer R.

Detailed formulas for spins $^5/_2$ and 2 with allowance for coherent and incoherent terms will be published by the depository method.

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