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RADIATIVE ECHOES

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1. A SYSTEM OF SPINS IN A MAGNETIC FIELD

IT is usually assumed that the spontaneous emission from separate and noninteracting atoms or molecules occurs independently, so that the intensity of the radiation from an assembly of particles is obtained by multiplying the transition probability for one particle by the total number of particles. Generally speaking, this is incorrect: if the assembly of particles occupies a volume whose linear dimensions are small in comparison with the wavelength, then even for particles that do not interact directly with each other the emission does not occur independently. In this case there is always an intense indirect interaction through the electromagnetic field of the radiation, and the result is that the process of spontaneous emission occurs coherently for the entire assembly of particles.[1-5] There can then be so-called "superradiative" states, for which the intensity of spontaneous decay is proportional to the square of the total number of particles in the system.

As an example let us consider a system of electrons in a constant magnetic field. Depending on the orientation of the spin of an electron relative to the magnetic field the electron's energy can take two values, $-\epsilon$ and $+\epsilon$ (Fig. 1). The total energy of the system of electrons is

$$\langle \hat{\mathcal{H}}_0 \rangle = (N_2 - N_1)\,\varepsilon, \tag{1}$$

where N_2 (N_1) is the number of electrons with spin orientated along (opposite to) the field.

At the same time $\langle \hat{S}_z \rangle = \frac{1}{2}(N_2 - N_1)$ is the value of the total projection of the spins of the electrons in the direction of the external field. Therefore the energy of the electrons can be written in the form

$$\hat{\mathcal{H}}_0 = \hbar\omega \hat{S}_z. \tag{2}$$

In interaction with the electromagnetic field transitions can occur between the energy levels, with emission or absorption of energy. Transitions between the levels will occur owing to magnetic field components H_x and H_y perpendicular to the constant field. Therefore the interaction operator responsible for the transitions can be written in the form

$$\hat{V} = -\mu \sum_i \{\hat{s}_{xi} H_x(\mathbf{r}_i) + \hat{s}_{yi} H_y(\mathbf{r}_i)\}, \tag{3}$$

where μ is the Bohr magneton: s_{xi} and s_{yi} are components of the spin of an electron; and $H_{x,y}(\mathbf{r}_i)$ is the value of a magnetic field component of the radiation at the location of the i-th electron. The sum is taken over the entire system of electrons.

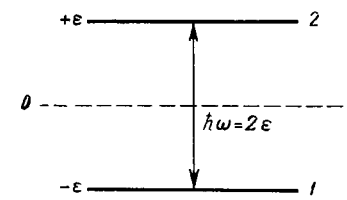

FIG. 1. Diagram for a two-level system.

If the volume occupied by the electrons has linear dimensions much smaller than the wavelength $\lambda = \pi\hbar c/\epsilon$, then in the expression for V we can neglect the dependence of the field on $\mathbf{r}_i$ so that

$$\hat{V} = -\mu(\hat{S}_x H_x + \hat{S}_y H_y), \tag{4}$$

where

$$\hat{S}_x = \sum_i \hat{s}_{xi}; \quad \hat{S}_y = \sum_i \hat{s}_{yi}.$$

It follows from Eqs. (2) and (4) that the complete Hamiltonian describing the energy of our system interacting with the electromagnetic field can be expressed in terms of the components of the total spin of the system

$$\hat{\mathcal{H}} = \hbar\omega \hat{S}_z + \mu(\hat{S}_x H_x + \hat{S}_y H_y). \tag{5}$$

It is not hard to see that the operator for the absolute value of the total spin of the system, $\hat{S}^2 = \hat{S}_x^2 + \hat{S}_y^2 + \hat{S}_z^2$, commutes with the operator $\mathcal{H}$, and accordingly is conserved in electromagnetic transitions accompanied by absorption or emission of electromagnetic energy by the electrons of the system.

For this reason a convenient set of quantities characterizing a pure quantum state of the system is made up of the quantum numbers S, $M = \langle \hat{S}_z \rangle$.

Let us now calculate the probability per unit time w for spontaneous emission of a photon by the system of electrons. According to radiation theory[6]

$$w = \frac{4}{3}\mu^2 \frac{\omega^3}{\hbar c^3} |\langle S, M | \hat{S}_{x,y} | S, M-1 \rangle|^2. \tag{6}$$

Calculating the matrix elements of the operator S_x in the representation S, $S_z = M$, we get

$$w = I_0 (S - M + 1)(S + M), \tag{7}$$

where $I_0 = (4\mu^2/3)(\omega^3/\hbar c^3)$ is the probability of spontaneous emission by one particle.

According to its physical meaning, M can take values from $-S$ to S, and

$$S \leqslant \frac{N}{2}, \tag{8}$$

where N is the total number of electrons in the system. If originally all of the electrons were in the upper level, M = N/2 (or the lower level, M = N/2 (sic)), then S has its maximum value and is equal to N/2.

In the interaction with the electromagnetic field by emission (or absorption) the system can go over into a state with M = 0 (equal populations of the two levels). In this case the intensity of the spontaneous emission is proportional to

$$w = I_0 \frac{N}{2}\left(\frac{N}{2}+1\right) \simeq I_0 \frac{N^2}{4}. \tag{9}$$

Accordingly, the emission from the system as a whole is N/4 times as intense as that from N independent particles.

Using the Hamiltonian (5) and the commutation relations for the operators $\hat{S}_j$, we can describe the change with time of the components of the total spin.

The Heisenberg equation for $\hat{S}_j$ is of the form

$$\frac{d\hat{S}_j}{dt} = \frac{i}{\hbar}[\hat{\mathcal{H}}, \hat{S}_j],$$

and for the average values $\langle \hat{S}_j \rangle$ it goes over into the classical equation of motion of the magnetic moment

$$\frac{d\langle S\rangle}{dt} = \gamma [\mathbf{H} \times \langle S\rangle], \tag{10}$$

where H is the external magnetic field and γ is the gyromagnetic ratio.

For a complete classical description of the process of emission the equation (10) must be solved simultaneously with the Maxwell equations.

The superradiative state has a very intuitive vector interpretation. In the initial state let all of the particles have their spins directed along the external field, so that the total spin of the system is equal to its maximum value $\frac{1}{2}\hbar N$. We then also have

$$\langle S_z\rangle = M = \frac{1}{2}N. \tag{11}$$

Through the action on the system of a resonance field with polarization perpendicular to the external magnetic field the system of spins can be brought into a state with $\langle \hat{S}_Z \rangle = 0$ and the maximum value $\langle \hat{S}_\perp^2 \rangle^{1/2} = \langle S_X^2 + S_Y^2 \rangle^{1/2} = \frac{1}{2}\hbar N$. In this case $\mathbf{S} \perp \mathbf{H}_0$, and the particles are equally distributed between the energy levels. If we now turn off the external resonance field, the system begins to radiate spontaneously, and according to (10) the maximum value of $\langle \hat{S}_\perp \rangle$ corresponds to a maximum intensity of the radiation.

Up to now we have considered the processes of emission from a system of spins without taking into account the relaxation processes that occur in real systems.

Superradiative states are nonequilibrium states, since in thermodynamic equilibrium we always have $\langle \hat{S}^2 \rangle^{1/2} \simeq \langle S_Z \rangle$. It is clear from this that every sort of relaxation process leads to destruction of a superradiating state.

For the system of the spins of the nuclei of a crystal located in a magnetic field (nuclear magnetic resonance) we can designate three different relaxation processes[7]: exchange of energy between different spins leads to a damping of the components $\langle \hat{S}_X \rangle$ and $\langle \hat{S}_Y \rangle$, with the result that $\langle \hat{S} \rangle \to \langle M \rangle$, and the superradiative state is destroyed; exchange of energy between the system of nuclear spins and the lattice—this leads not only to a damping of the components $\langle \hat{S}_X \rangle$ and $\langle \hat{S}_Y \rangle$, but also to an approach of $\langle \hat{S}_Z \rangle$ (i.e., the energy of the system of spins) to its equilibrium value, which depends on the temperature of the crystal lattice. The time for the first of these processes is commonly called T_2, and the time for the second, T_1. Usually $T_2 \ll T_1$, so that a coherent superradiative state of the system is destroyed much more rapidly than the system of spins gets into thermodynamic equilibrium with the lattice (the thermostat).

The process of relaxation is not the only cause of destruction of a coherent state. Because of imperfections of the crystal and nonuniformities of the external field there are differences between the energies of the spins of different nuclei, and this causes some lack of coincidence of the resonance frequencies. Usually the resonance frequencies have a Gaussian distribution with an effective width $\Delta\omega$. In the emission process this difference between the frequencies of different spins leads to a lack of agreement of phase of the individual emitting centers (spins), which leads to a loss of coherence. The time T_2^* during which this occurs is equal to $(\Delta\omega)^{-1}$. At sufficiently low temperatures T_2^* is smaller than T_2, since the tempo of the relaxation processes is slowed with decreasing temperature. Both T_2^* and T_2 cause broadening of the spectral line.

Relaxation processes cause a so-called "homogeneous" broadening; the difference of the resonance frequencies of the individual radiators leads to an "inhomogeneous" broadening. The essential difference between these two types of broadening is as follows. Relaxation processes are irreversible, and therefore the loss of coherence caused by homogeneous broadening is irreversible. Inhomogeneous broadening is not due to irreversible processes, and the loss of phase of the emitting centers, which leads to loss of coherence, is reversible. This important fact is utilized in the effect of the "spin echo."[8,7]

2. SPIN ECHOES

The main features of the spin-echo phenomenon are as follows: a system of nuclear spins placed in a magnetic field is brought into a superradiative state and one observes the coherent spontaneous emission from this system. In the course of time the coherent state

gets destroyed, and the intensity of the radiation drops sharply.

If the getting out of phase has occurred owing to inhomogeneous broadening ($T_2^* \ll T_2$), the reversibility of the process allows us to restore the coherence by means of time reversal. It can be seen that Eq. (10) remains unchanged if we change the sign of the time and also change the direction of the magnetic field. Therefore in a system of spins in a magnetic field we can "reverse" the time by changing the direction of the magnetic field. We shall measure the time from the instant when the original signal reaches its maximum value (Fig. 2).

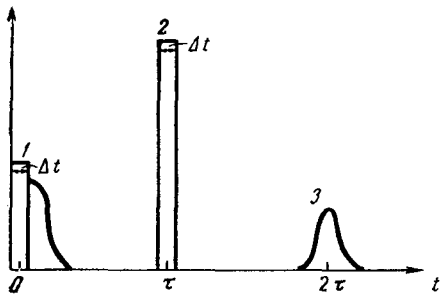

FIG. 2. Time chart of the production of a spin echo. 1 and 2 are the exciting pulses; 3 is the spin echo; Δt is the duration of an exciting pulse.

If the change of the "direction of time" is made at a time τ ($T_2^* < \tau < T_2$), then at the time 2τ a strong radiation signal appears, caused by the recovery of the coherent properties of the system. This phenomenon has received the name "spin echo."

The maximum of this signal is less than the original maximum, since relaxation processes have had time partially to destroy the coherence in an irreversible way.

An estimate of the parameters of the pulse necessary to bring the system into a superradiative state can be given in the following way. According to [9] the probability for a particle to make a transition from one level to the other in the time Δt under the action of the resonance field is given by the following formula:

$$W = \frac{1}{2}\left(1 - \cos\frac{\mu_n H}{\hbar}\Delta t\right), \qquad (12)$$

where H is the amplitude of the magnetic field of the pulse and μ_n is the magnetic moment of the nucleus.

If the particles were originally in the lower level (all spins oriented along the field), then to reach equality of the populations it is necessary that

$$\frac{\mu_n H}{\hbar}\Delta t = \frac{\pi}{2}, \qquad (13)$$

where Δt is the duration of the pulse. The angle through which the total angular momentum of the system is turned is then 90°. Therefore a field pulse satisfying the relation (13) is called a "ninety-degree pulse." It is clear that the duration Δt of the pulse must be less than T_2^*.

"Time reversal" by switching the direction of the magnetic field is inconvenient. The change of direction of the field is, however, equivalent to a change of orientation of the spins relative to the field by 180°. This latter change is equivalent to an inversion of the populations of the energy levels under consideration, and can be accomplished by the action on the system of an 180-degree pulse, whose amplitude is twice that of the 90-degree pulse, while their durations are the same. A time chart of the production of the echo is shown in Fig. 2.

3. THE GENERAL CASE OF AN ASSEMBLY OF TWO-LEVEL SYSTEMS

The results we have obtained are not valid merely for electrons or nuclei placed in an external magnetic field; they can be extended to any assembly of systems whose spectra consist of two energy levels.* The wave function of such a particle has only two components ψ_1 and ψ_2, and all operators acting on this function can be represented as two-rowed matrices. As is well known, [9] any two-rowed matrix can be put in the form of a linear combination of the Pauli matrices

$$r_1 = \frac{1}{2}\begin{Vmatrix} 0 & 1 \\ 1 & 0 \end{Vmatrix}, \quad r_2 = \frac{1}{2}\begin{Vmatrix} 0 & i \\ -i & 0 \end{Vmatrix},$$
$$r_3 = \frac{1}{2}\begin{Vmatrix} 1 & 0 \\ 0 & -1 \end{Vmatrix}, \quad I_0 = \frac{1}{2}\begin{Vmatrix} 1 & 0 \\ 0 & 1 \end{Vmatrix}. \qquad (14)$$

For example, the matrix for the dipole moment

$$\|\mu\| = \begin{Vmatrix} \mu_{11} & \mu_{12} \\ \mu_{21} & \mu_{22} \end{Vmatrix}$$

can be written

$$\|\mu\| = \mathbf{e}_1 r_1 + \mathbf{e}_2 r_2 + \mathbf{e}_3 r_3 + \mathbf{e}_0 I_0, \qquad (15)$$

where

$$\mathbf{e}_1 = \mu_{12} + \mu_{21};\ \mathbf{e}_2 = i(\mu_{21} - \mu_{12});$$
$$\mathbf{e}_3 = \mu_{11} - \mu_{22};\ \mathbf{e}_0 = \mu_{11} + \mu_{22}.$$

For dipole interaction between molecules and the radiation field the operator for the interaction of a molecule with the radiation field can be put in the form

$$\hat{V} = -(\mathbf{e}_1 r_1 + \mathbf{e}_2 r_2)\,\mathbf{F}(\mathbf{q}, t). \qquad (16)$$

*Actually, there exist in nature practically no systems having only two energy levels. If, however, the interaction with a field is of a clearly marked resonance character, the effect of other levels can as a rule be neglected. In this case a two-level system is a sufficiently good approximation to the real system, and is widely used in the theory of the interaction of radiation with matter. It is important to take into account the "many-level" nature of the system in the description of the interaction with very intense fluxes of radiation, when there can be such nonlinear effects as the generation of harmonics, and so on.

We here assume that the diagonal matrix elements μ_{11} and μ_{22} are equal to zero. In the first place, this is always true for the electric dipole moment in a nondegenerate state. Secondly, this assumption is of no importance in principle, and only makes the further treatment less cumbersome.

The operator for the internal energy of a noninteracting particle can be written in the form

$$\hat{\mathcal{H}}_0 = \hbar\omega\hat{r}_3, \tag{17}$$

where ω is the frequency of the transition between the energy levels. The vector $\hat{\mathbf{r}}$ with the three components $\hat{r}_1$, $\hat{r}_2$, $\hat{r}_3$ can be regarded as the operators of a sort of spin acting in the energy space, since the commutation relations for the operators $\hat{r}_j$ ($j = 1, 2, 3$) are completely analogous to those for a spin.

When an assembly of identical particles is in interaction with the field the Hamiltonian of the system is of the form

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{V},\ \hat{V} = -\sum_i (\mathbf{e}_1\hat{r}_{1i} + \mathbf{e}_2\hat{r}_{2i})\,\mathbf{F}(\mathbf{q}_i, t), \tag{18}$$

where the sum is taken over all the particles.

If the linear dimensions of the system are much smaller than the wavelength, we have

$$\hat{\mathcal{H}} = \hbar\omega\hat{R}_3 - (\mathbf{e}_1\hat{R}_1 + \mathbf{e}_2R_2)\,\mathbf{F}(\mathbf{q}_0, t), \tag{19}$$

where $R_j = \sum_i r_{ij}$, $j = 1, 2, 3$, and $\mathbf{q}_0$ is a value of $\mathbf{q}$ inside the system of particles. Along with the operators $\hat{R}_j$ we can introduce the operator for the absolute value of the vector $\hat{\mathbf{R}}$: $\hat{R}^2 = \hat{R}_1^2 + \hat{R}_2^2 + \hat{R}_3^2$. The physical meaning of the operator $\hat{R}_3$ is clear: its eigenvalues are equal to the half difference of the polulations of the energy levels in question, and we have $\langle\hat{R}_3\rangle = M \leq |R|$, $R \leq N/2$, where N is the total number of particles in the system. The operators $\hat{R}_1$ and $\hat{R}_2$ are associated with transitions between the energy levels. Their role in the emission or radiation is analogous to that of the total spin components $\hat{S}_x$ and $\hat{S}_y$. In dipole transitions the mean values of $\hat{R}_1$ and $\hat{R}_2$ determine the mean value of the dipole moment of the system[3]

$$\langle\mathbf{P}\rangle = \mathbf{e}_1\langle\hat{R}_1\rangle + \mathbf{e}_2\langle\hat{R}_2\rangle. \tag{20}$$

The commutation relations between the operators $\hat{R}_j$ are the same as those between the components S_j of the spin operator:

$$\hat{R}_1\hat{R}_2 - \hat{R}_2\hat{R}_1 = i\hat{R}_3;\ \hat{R}_2\hat{R}_3 - \hat{R}_3\hat{R}_2 = i\hat{R}_1;\ \hat{R}_3\hat{R}_1 - \hat{R}_1\hat{R}_3 = i\hat{R}_2.$$

In the general case of a two-level system the intensity of the spontaneous emission is given by the expression

$$I = I_0(R+M)(R-M+1), \tag{21}$$

where I_0 is the intensity of the emission from an individual molecule.

The change of $\hat{R}_j$ with time is described by the Heisenberg equations

$$\frac{d\hat{R}_j}{dt} = \frac{i}{\hbar}[\hat{\mathcal{H}}, \hat{R}_j], \tag{22}$$

and by the introduction of the effective field $\mathbf{F}_{\text{eff}}$ these can be written in the form[15]

$$\frac{d\langle\hat{R}_j\rangle}{dt} = \gamma_{\text{eff}}[\mathbf{F}_{\text{eff}} \times \langle\hat{R}_j\rangle], \tag{23}$$

where

$$F_{1\,\text{eff}} = \frac{\mathbf{e}_1\mathbf{F}}{|\mu_{12}|};\quad F_{2\,\text{eff}} = \frac{\mathbf{e}_2\mathbf{F}}{|\mu_{12}|};$$

$$F_{3\,\text{eff}} = \frac{\hbar\omega_{21}}{|\mu_{12}|};\quad \gamma_{\text{eff}} = \frac{|\mu_{21}|}{\hbar}. \tag{24}$$

$F_{3\,\text{eff}}$ plays a role analogous to that of the constant magnetic field in spin-echo experiments.

The coherent spontaneous emission of molecules has been observed in quite a number of systems. The most interesting of these is the molecular generator (maser) with tandem resonators.[3,10-14] In a molecular generator a beam of molecules which are in an excited state goes into a resonator. For such molecules, as follows from what has been said, R is a maximum. The generation process, which occurs in the resonator when the threshold condition for generation is oversatisfied to a sufficient degree, tends to equalize the populations of the levels, so that at the exit from the resonator there is a superradiative state. If a second resonator is placed just beyond the first one, intense coherent spontaneous radiation is observed in the second resonator. The two-resonator molecular generator is remarkable for the great variety of types of operation that are possible.[3] Some of them have found practical application in the improvement of the stability of frequency standards,[19,20] the increase of resolving power of spectroscopes,[21] and so on.

In molecular beams it is possible to observe effects similar to spin echoes. For this it is necessary that the time of flight of the molecules through the resonator (the analog of T_1) be much larger than the inverse of the Doppler width of the line (the analog of T_2^*). For the mode E_{0lm} in a cylindrical resonator the Doppler width of the line is of the order of $\pi(v/L)m$, where L is the length of the resonator, v is the mean speed of the molecules, and m is the number of half-waves along the resonator in the characteristic mode of vibration. In the same approximation the time of flight of a molecule through the resonator is L/v. From this we get a requirement on the index of the vibration mode: $\pi m \gg 1$. It is also necessary that m be odd, since for even m the resonator is poorly excited by the molecular beam.[22]

4. PECULIARITIES OF THE COHERENT EMISSION IN THE OPTICAL REGION

The analysis we have made applies to the case of wavelengths λ so long that there are many radiators in the volume λ^3. This is as a rule the case in the radio region, and all of the experiments mentioned above (spin echoes, two-resonator molecular generators) have been made in the radiofrequency range.

Quite recently experiments have been made to observe effects analogous to spin echoes in the optical region[16,17]; the authors of these papers call these effects "photon echoes."* Coherent spontaneous emission in the optical region has a number of special features, with which we shall deal in this section.

In the optical region the linear dimensions of the radiating object are as a rule much larger than the wavelength, because the wavelength is so small. Therefore our previous arguments cannot be applied without modification in the optical region.

If, however, we treat the radiation field as a superposition of plane waves with wave vectors **k** and polarizations **e**, we can introduce a conserved vector $\mathbf{R_k}$ which describes the coherent spontaneous emission of field components with prescribed direction of propagation and polarization.

When we represent the radiation field as an expansion in plane waves

$$F(q, t) = \sum_k (F_k e^{ikq} + F_k^* e^{-ikq}),$$

the interaction Hamiltonian (18) can be written in the form[1]

$$\hat{V} = -\sum_k (e_1 \hat{R}_{1k} + e_2 \hat{R}_{2k}) F_k, \tag{25}$$

where

$$\left.\begin{aligned} \hat{R}_{1k} &= \sum_i (\hat{r}_{1i} \cos kq_i - \hat{r}_{2i} \sin kq_i), \\ \hat{R}_{2k} &= \sum_i (\hat{r}_{1i} \sin kq_i - \hat{r}_{2i} \cos kq_i). \end{aligned}\right\} \tag{26}$$

The summation in (26) is taken over the entire volume occupied by the system of particles.

A coherent state with given values of R_k^2 and R_{3k} can be produced by applying to the system a pulse of radiation propagated in the direction **k**.

Introducing R and M, the eigenvalues of the operators $\hat{R}_k^2 = \hat{R}_{1k}^2 + \hat{R}_{2k}^2 + \hat{R}_{3k}^2$ and $\hat{R}_{3k}$, we can show that the selection rules for transitions with $\Delta k = 0$ are the same as before: $\Delta R = 0$; $\Delta M = \pm 1$.

In this case the direction of the spontaneous emission is that of the exciting pulse which produces the coherent state described by R_k; the quantity R is conserved ($\Delta R = 0$), and the emission does not destroy the coherent state.

If, on the other hand, $\Delta k \neq 0$, then the selection rules are[1]

$$\Delta R = 0, \pm 1; \quad \Delta M = \pm 1. \tag{27}$$

The possibility of emission with change of the wave vector ($\Delta k \neq 0$) leads to nonconservation of $\langle \hat{R}_k^2 \rangle$, i.e., in the final analysis to a loss of coherence.

It can be shown[1] that the intensity of the spontaneous emission from a sufficiently large number of particles is given by the relation

$$I(\mathbf{k}') = I_0(\mathbf{k}') \frac{1}{2} N \left\{ 1 - \cos\theta + \frac{1}{2} \sin^2\theta [N |\langle e^{i(\mathbf{k}-\mathbf{k}')\mathbf{q}} \rangle|^2 - 1] \right\}. \tag{28}$$

Here $I_0(\mathbf{k}')$ is the intensity of the spontaneous emission of one molecule in the direction $\mathbf{k}'$; θ is the mean angle, determined by the relation $\cos^2\theta = \langle M^2 \rangle / \langle \hat{R}_k^2 \rangle$—in other words, it is the angle through which the vector **R** is rotated owing to the applied pulse of light; for a 90° pulse $\theta = \pi/2$. The symbol $\langle\ \rangle$ denotes averaging over the entire volume occupied by the particles. It is assumed that all of the particles were originally known with certainty to be in one of the energy levels considered.

For a system of sufficiently large extent

$$\langle e^{i(\mathbf{k}-\mathbf{k}')\mathbf{q}} \rangle = \delta_{\mathbf{k}\mathbf{k}'}.$$

Therefore for a 90° pulse the intensity in directions $\mathbf{k}' \neq \mathbf{k}$ is close to zero.

If, on the other hand, $\mathbf{k}' = \mathbf{k}$, then under the same conditions

$$I(\mathbf{k}') = I_0(\mathbf{k}') \frac{1}{2} N \left(\frac{1}{2} N - 1 \right). \tag{29}$$

It follows from this that a system of great extent radiates coherently in the direction **k** which is that of the exciting pulse.

In the observation of a photon echo two pulses act on the system, a 90° pulse and a 180° pulse. In this case the intensity of the echo is[17]

$$I(\mathbf{k}) = I_0(\mathbf{k}) \frac{1}{2} N \left\{ 1 + \frac{1}{2} N |\langle e^{i(\mathbf{k}+\mathbf{k}_1-2\mathbf{k}_2)\mathbf{q}} \rangle|^2 - 1 \right\}, \tag{30}$$

where **k** is the direction of observation of the echo, $\mathbf{k}_1$ is the direction of the 90° pulse, and $\mathbf{k}_2$ is the direction of the 180° pulse. For a system of large extent an echo is observed only in the direction for which

$$\mathbf{k} + \mathbf{k}_1 - 2\mathbf{k}_2 = 0. \tag{31}$$

If the polarizations of the first and second pulses are different this does not affect the magnitude of the echo signal, but changes the polarization of the signal.

5. EXPERIMENTAL RESULTS ON THE OBSERVATION OF PHOTON ECHOES

Recent papers[16,17] give descriptions of experiments made to observe photon echoes by means of a ruby crystal irradiated with the light of a ruby laser. The laser which was the source of radiation was operated with modulation of the figure of merit (Q), generating light pulses of duration 10 nsec.

The width of the R_1 line of the ruby* used in the experiment was due both to homogeneous broadening caused by the interaction of the chromium ions with the lattice, and to inhomogeneous broadening caused by nonuniformity of the crystalline field. At room temperature the homogeneous broadening is much

*Since spin echoes and photon echoes are of the same physical nature, it is convenient to introduce a single term for them — radiative echoes.

*The designation of the ruby lines must not be confused with the components of the vector R.

larger than the inhomogeneous, and it is not possible to observe an echo. At liquid-helium temperature, however, the inhomogeneous broadening of the line is larger than the homogeneous, and this makes it possible to observe an echo. One of the essential difficulties of making the photon-echo experiment is due to the rather large temperature shift of the peak of the spectral line. When the temperature is changed from room temperature to helium temperature, to which the ruby specimen must be cooled, the shift of the peak of the spectral line goes beyond the limits of its width. Therefore it would seem that it is necessary to have a laser with figure-of-merit modulation and sufficiently high power operating at helium temperature. This difficulty has been overcome to a large extent in the following way.

Figure 3 shows the structure of the R lines of the ruby. Because of the splitting of the lower level into two sublevels in the crystalline field, the R_1 line (and also the R_2 line) consists of two components:

$$^4A_2\left(M=\pm{}^3/_2\right)\rightarrow {}^2E\,(\bar{E})\,(R_{13/2});$$
$$^4A_2\left(M=\pm{}^1/_2\right)\rightarrow {}^2E\,(\bar{E})\,(R_{11/2}).$$

It turns out that the peak of the component $R_{1_{1/2}}$ at temperature 4.2°K coincides remarkably closely with the peak of the component $R_{1_{3/2}}$ at 77°K. This makes it possible to use a laser with figure-of-merit modulation operating at liquid-nitrogen temperature, which simplifies the technique of the experiment.

Another feature of the experiment is perhaps more a matter of principle. It was found that to observe the echo signal it is necessary to place the specimen being studied in a constant external magnetic field. The largest echo signal is obtained in the case in which the external magnetic field is oriented along the optic axis of the crystal. The authors of [17] ascribe this unexpected result to peculiarities of the relaxation mechanism in the ruby crystal. To produce the two successive pulses they used a single pulse of the figure-of-merit modulated quantum generator, and this was divided into two signals by means of a semitransparent mirror placed at an angle with the laser beam. The signal reflected from the mirror was sent into the ruby specimen after a time $\tau = 50$ nsec, at a small angle φ relative to the direction of propagation ($\mathbf{k}_1$) of the first signal. In this case, as follows from (31), the direction of propagation of the echo signal is at the angle 2φ, which made it possible to use a diaphragm to exclude the effects of the direct signal from the receiving device.

Figure 4 shows oscillograms of the experiment. It can be seen from Fig. 4 that there is no echo if there is no magnetic field (Fig. 4,a), or if the first exciting pulse is absent (Fig. 4,c), but the echo appears distinctly in Fig. 4,b in the presence of an external magnetic field.

It was predicted in [18] that there can be multiple echoes, and these have been successfully observed experimentally in the optical region by the use of a ruby crystal.[17] For a series of echo signals to be observed in the same direction it is necessary that the directions of the first and second exciting pulses be the same ($\mathbf{k}_1 = \mathbf{k}_2$). In this case, to avoid direct illumination of the photomultiplier by the pumping light, a supplementary shutter based on a Kerr cell was placed in front of the photomultiplier.

Figure 5 shows oscillograms of the observation of multiple echoes. Oscillogram a shows two exciting pulses, an echo pulse, and a small additional pulse of the second echo. The small amplitude of the signal is due to the fact that the Kerr-cell shutter was open only for a period of 100 nsec, and with the interval between

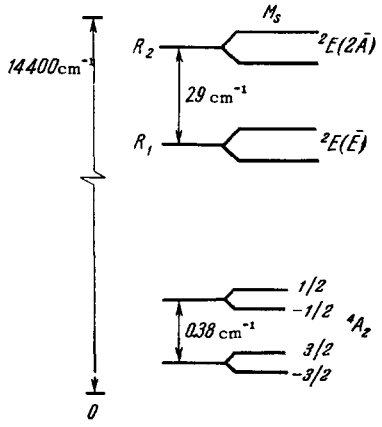

FIG. 3. Scheme of ruby energy levels used in the experiment.

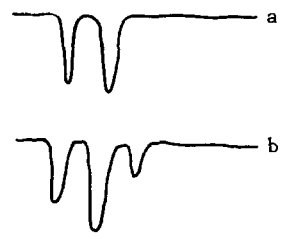

FIG. 4. Photon echo (experimental results of [17]).

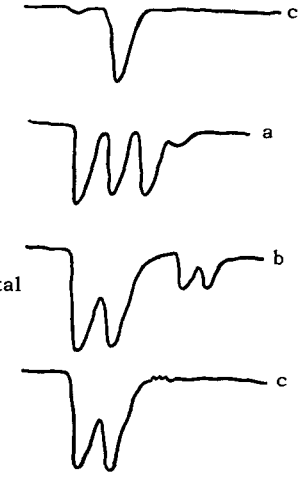

FIG. 5. Multiple echoes (experimental results of [17]).

the pulses $\tau = 50$ nsec this led to cutting off of the pulse of the second echo. In Fig. 5,b we can see the signals of the exciting beams and of the second and third echoes. In this case the signal of the first echo was "cut off" by the shutter, since it was opened with a time delay relative to the first pulse. Figure 5,a and b, is for the case of optimal orientation of the magnetic field. By a change of the orientation of the magnetic field the echo signals can be made to vanish completely (Fig. 5,c).

Multiple echoes can be explained if we regard the echo signal as a reorienting pulse. The second echo is then formed by the pair of pulses consisting of the second exciting pulse and the first echo signal; the third echo can be formed by two pairs of pulses: the first and second echoes, or the first exciting pulse and the first echo.

The successful observation of radiative echoes in the optical region opens up new possibilities for using this interesting effect in spectroscopic researches.

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