New mechanism of photon echo in magnetic systems

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The possibility is considered of observation of a photon echo in an ensemble of localized excitons strongly interacting with magnons. It is shown that even in the absence of nonrenormalized inhomogeneous broadening in the exciton subsystem, the photon echo takes place and is due to fluctuations in the ferromagnetic subsystem at a temperature $T > 0$.

1. INTRODUCTION

It is known that the observation of echo phenomena is possible in an ensemble of inhomogeneusly broadened two-level systems (TLS). In particular, if the transition to the TLS is optical and the considered ensemble is successively subjected to the action of two short pulses of an electromagnetic field with a resonance spectrum of frequencies, then in the time τ after the last pulse, a spontaneous response of the system appears—the photon echo.[1] Here the presence of a spread of energies in the TLS exists in principle and the duration of the echo decreases with increase in the magnitude of this spread.

It was first shown in Ref. 2 that the phenomenon of photon echo can take place even in the absence of nonrenormalized scatter of energies of the TLS if the latter interacts strongly with the phonons.

We demonstrate here the possibility of the excitation of a photon echo in an ensemble of localized excitons (unusual TLS) with the same energy of excitation, under the condition that the interaction with the magnetic subsystem be sufficiently strong.

In the absence of excitons, the considered system represents a lattice ferromagnet with translationally invariant exchange integrals. The excitation of the exciton by the electromagnetic field at some site changes the orbital wave function of the latter (with conservation of spin) and as a result leads to an increase in the corresponding exchange integrals. Since the radius of localization of the excited state of the site exceeds that for the ground state, the exchange integral can grow significantly.

In the case considered in the present work, inhomogeneous broadening in its usual sense (for example, due to disorder) is absent. At the same time, for not very low temperatures, an effect similar to inhomogeneous broadening, connected with fluctuations in the spin subsystem, can be observed in ferromagnets.

Since the characteristic exciton times are significantly shorter than those of magnons, the excitation of an exciton is equivalent to "agitation." Here the eigenstates of the magnetic subsystem excited in such a fashion differ from the nonrenormalized states. As a result, the system turns out to be in a nonstationary state, which is a superposition of "new" eigenstates having a certain energy scatter. In order that this effect lead to the possibility of observation of the echo (with duration $T_2^* < \tau$), the energy scatter that we mentioned should not be very small. The latter can in turn be assured by the large value of the perturbation produced by the exciton in the magnetic subsystem. We note that at temperature contributes in any case to a decrease in the echo duration, since it means an increase in the phase volume of the newly possible eigenstates. Obviously, this leads to a large scatter in the energies of excitation of the excitons. At zero temperature, the time T_2^* increases without limit and the photon echo vanishes. At the same time, if the scatter of energy of the TLS is due to their interaction with phonons, photon echo takes place even at $T = 0$.

The research will be reported in the following fashion: In Sec. 2 we derive the general relations for the nonlinear response of the considered system to electromagnetic excitation. Section 3 is devoted to the problem of the calculation of the response below the Curie temperature. It is shown in this case that photon echo is observed for short times, in the interval between the characteristic exciton and magnon times, in a certain range of temperatures (depending on the parameters of the system). In the opposite case of long times, an exponential decay of the excitation is observed. In Sec. 4, the same effects are studied for the paramagnetic phase. In the Conclusion, the limits of applicability of the studied model are discussed briefly, as is the possibility of experimental observation of the predicted effect.

2. GENERAL RELATIONS FOR THE NONLINEAR RESPONSE

We express the Hamiltonian of interaction of the exciton-magnon system in the form (see, for example, Refs. 3,4):

$$H = H_0 + \sum_i V_i c_i^+ c_i, \qquad (1)$$

$$H_0 = -J \sum_{i,j} \mathbf{S}_i \mathbf{S}_j, \qquad V_i = E - (J' - J)\mathbf{S}_i \sum_j \mathbf{S}_j. \qquad (2)$$

Here H_0 is the usual Heisenberg exchange Hamiltonian, $\mathbf{S}_i$ is the spin operator, V_i describes the contribution to the energy of the system upon excitation of an exciton of energy E at site i, and takes into account the change in the exchange integral, $\Delta J = J' - J$, due to this process; c_i^+ and c_i are the creation and anihilation operators of excitons and obey Pauli statistics.

We shall be interested in the response of the system to the action of two sucessive electromagnetic-field pulses with wave vectors k_1 and k_2, respectively, with like frequency ω. Here each pulse duration is so small in comparison with the time interval τ between them, that we can refer to them as δ function and assume that the pulses act successively on the system at the times $t_1 = 0$ and $t_2 = \tau$. This approximation

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turns out to be valid if the width of the frequency spectrum of the pulses exceeds the characteristic magnon frequencies of the problem.

We write down the interaction of the considered system with the electromagnetic field in the form

$$W(t)=-\sum_i (C_i^+ d + c_i d^+)E_i(t), \quad (3)$$

$$E_i(t)=E_{i1}(t)+E_{i2}(t). \quad (4)$$

$$E_{in}(t)=P_{in}(t)\delta(t-t_n)\exp(-i\omega t)+ \text{c.c.}\,,$$
$$P_{in}=P_n \exp(i\mathbf{k}_n\mathbf{R}_i),$$

d is the matrix element of transition upon creation of the exciton and P_n is the amplitude of the field. In (3) and (4), we follow the notation of Ref. 5.

The action of the field (4) polarizes the system described by the Hamiltonian (1). According to classical electrodynamics, the field connected with this polarization, has far from the system the form

$$E_f(r,t) \approx \frac{\exp[i(kr-\omega t)]}{r}\sum_i \langle c_i\rangle \exp(-i\mathbf{k}\mathbf{R}_i), \quad (5)$$

where $k = \omega/c$, $\mathbf{k} = \mathbf{R}/|\mathbf{R}|$ (c = velocity of light). Then the intensity is

$$I \propto \left|\sum_i \langle c_i\rangle \exp(-i\mathbf{k}\mathbf{R}_i)\right|^2. \quad (6)$$

where

$$\langle c_i\rangle = \mathrm{Sp}(c_i\rho(t)), \quad (7)$$

and $\rho(t)$ is the density matrix of the system.

We now consider the response of the system in the direction $2\mathbf{k}_2 - \mathbf{k}_1$ after action on it of the pulse (4). It is known[1,2,5] that such a radiation arises already in third order perturbation theory in the amplitude of the exciting field. In this approximation, the part of the density matrix of interest to us and makes a contribution to the field (5) can be written in the following form in the interaction representation [relative to the operator (3)][6]

$$\delta\rho_i(t)=i\int_0^t dt_1\int_0^{t_1} dt_2\int_0^{t_2} dt_3[\tilde W(t_1)[\tilde W(t_2)[\tilde W(t_3)\rho(0)]]], \quad (8)$$

where $\rho(0)$ is the equilibrium density matrix of the system, taken at zero time and at the temperature $T=\beta^{-1}$, while

$$\tilde W(t_1) = \exp(iHt_i)W(t_i)\exp(-iHt_i).$$

Actually, in optical experiments we always have

$$\beta\omega \gg 1. \quad (9)$$

Therefore, we can write down the density matrix $\rho(0)$ in the approximate form

$$\rho(0)=\prod_i c_i c_i^+\rho_0, \quad (10)$$

$$\rho_0=\exp(-\beta H_0)/\mathrm{Sp}[\exp(-\beta H_0)],$$

where the trace is taken only over spin space.

Equation (8) can be easily calculated if we take it into account that c_i^+ and c_i actually satisfy the Fermi commutation relations on one site, and use can be made of the identities

$$\exp[-i(H_0+V_ic_i^+c_i)t]c_ic_i^+\rho_0\exp[i(H_0+V_ic_i^+c_i)t]=c_ic_i^+\rho_0,$$

$$\exp[-i(H_0+V_ic_i^+c_i)t]c_i^+c_i\rho_0\exp[i(H_0+V_ic_i^+c_i)t]$$
$$=\exp[-i(H_0+V_i)t]\rho_0\exp[i(H_0+V_i)t]c_i^+c_i,$$

$$\exp[-i(H_0+V_ic_i^+c_i)t]c_i^+\rho_0\exp[i(H_0+V_ic_i^+c_i)t]$$
$$=\exp[-i(H_0+V_i)t]\rho_0\exp(iH_0t)c_i^+,$$

$$\exp[-i(H_0+V_ic_i^+c_i)t]c_i\rho_0\exp[i(H_0+V_ic_i^+c_i)t]$$
$$=\exp(-iH_0t)\rho_0\exp[i(H_0+V_i)t]c_i.$$

Calculating the matrix $\delta\rho(t)$ with the help of these relations and substituting the result in (5), we obtain

$$E_f(r,t)\propto\langle U_0(\tau)U(-\tau)U_0(\tau-t)U(t-\tau)\rangle\delta_{\mathbf{k},2\mathbf{k}_2-\mathbf{k}_1}$$
$$\times\exp(-2i\omega\tau), \quad (11)$$

where the evolution operators are given by

$$U(t)=\exp[-i(H_0+V_i)t],\ U_0(t)=\exp(-iH_0t). \quad (12)$$
$$\langle\ldots\rangle=\mathrm{Sp}(\ldots\rho_0).$$

We introduce the quantity

$$\delta V_i=V_i-\langle V_i\rangle,\ \langle V_i\rangle=E+\mathrm{Sp}(V_i\rho_0) \quad (13)$$

and consider the resonance case $\omega = \langle V_i\rangle$. With the aid of standard field theory methods,[7] the correlator in (11) can be rewritten in the form

$$K(t,\tau)=\langle\sigma(-\tau)\sigma(t-\tau)\rangle, \quad (14)$$

with

$$\sigma(t)=U_0(-t)U(t)=\tilde T_\tau \exp\left[-i\int_0^t \delta V_i(t')dt'\right], \quad (15)$$

where $\tilde T_\tau$ is the operator of chronological (anticrhonological) ordering if $t>0$ ($t<0$). We now proceed to the calculation of the correlator (14).

3. LOW TEMPERATURES, $T<T_c$

At temperatures below the Curie temperature T_c, the excitations in the considered system are weakly interacting magnons. In this case, must change over in the Hamiltonian (1) from spin to Bose operators to with the aid of the Goldstein–Primakov transformation

$$S_i^+=(2S)^{1/2}a_i,\quad S_i^-=(2S)^{1/2}a_i^+,\quad S_i^z=S-a_i^+a_i.$$

where S is the spin of the site. Then the Hamiltonian (1) is rewritten in the form

$$H_0=\sum_{\mathbf{p}}\omega_{\mathbf{p}}a_{\mathbf{p}}^+a_{\mathbf{p}}. \quad (16)$$

$$V_i=E+\Delta JSz\sum_{\mathbf{p},\mathbf{p}'}(1+f_{\mathbf{p}-\mathbf{p}'}-f_{\mathbf{p}}-f_{\mathbf{p}'})a_{\mathbf{p}}^+a_{\mathbf{p}'}\exp[i(\mathbf{p}-\mathbf{p}')\mathbf{R}_i], \quad (17)$$

$$f_{\mathbf{p}}=z^{-1}\sum_{\Delta}\exp(-i\mathbf{p}\Delta),$$

$$\omega_{\mathbf{p}}=2JSz(1-f_{\mathbf{p}}),\qquad a_{\mathbf{p}}^+=\sum_i\exp(i\mathbf{p}\mathbf{R}_i)a_i^+. \quad (18)$$

Here **p** and **p**′ are vectors from the Brillouin zone of the reciprocal lattice. For simplicity, we also assume that in the limits of the interaction radius R_0 the exchange integral is a constant and outside those limits it is equal to zero. Therefore, summation over the vectors Δ in (18) is actually carried out over the sites of the crystal lattice of the ferromagnet in the limits R_0. The number of these sites is $z \propto R_0^3$.

For calculation of the correlator (14), we can construct a diagrammatic expansion. According to the theorem on connected diagrams without free ends,[7] we have

$$K(t,\tau)=\exp(L_c), \tag{19}$$

where L_c is the sum of all connected diagrams.

In second-order perturbation theory in δV_i (terms of first order vanish since $\langle \delta V_i \rangle = 0$), there are three connected diagrams, to which corresponds the expression

$$L_c=-\int_0^{t-\tau} dt' \int_0^{t'} dt'' \Phi(t'-t'') - \int_0^{-\tau} dt' \int_0^{t'} dt'' \Phi(t'-t'') - \int_0^{t-\tau} dt' \int_0^{-\tau} dt'' \Phi(t'-t''), \tag{20}$$

$$\Phi(t'-t'')=\langle \delta V_i(t')\delta V_i(t'')\rangle. \tag{21}$$

Since the intensity (5) is determined by the modulus of the field $E_f(r,t)$, we shall be interested only in Re L_c. Expanding the correlators Re $\Phi(t-t')$ that are even in time in a series in cos $[\omega(t'-t'')]$, we obtain

$$\mathrm{Re}\, L_c=-\int \frac{d\omega}{\pi\omega^2}\Phi(\omega)\{3-2\cos[\omega(t-\tau)]-2\cos\omega\tau+\cos\omega t\}. \tag{22}$$

where

$$\Phi(\omega)=\frac{1}{2\pi}\int e^{i\omega t}\Phi(t)dt. \tag{23}$$

Using (16)–(18) for the correlator (21), we obtain (without limitation on generality, we can set $\mathbf{R}_i=0$)

$$\Phi(t)=(\Delta JSz)^2 \times \sum_{\mathbf{p},\mathbf{p}'} \exp[i(\omega_p-\omega_{p'})t]\,(1+f_{\mathbf{p}-\mathbf{p}'}-f_{\mathbf{p}}-f_{\mathbf{p}'})^2 n_p(1+n_{p'}), \tag{24}$$

where

$$n_p=[\exp(\beta\omega_p)-1]^{-1}.$$

In the region of low temperatures $n_p \ll 1$, only small values of p make a contribution to the sum.

Let $\omega_0 \propto JSz \propto T_c/S$ be the maximal frequency of spin waves corresponding to the Hamiltonian (16). The function (23) is obviously not small only if $\omega \ll \omega_0$. We consider two cases of short $(\omega_0 t \ll 1)$ and long $(\omega_0 t \gg 1)$ times.

In the first case, we can expand in Eq. (22) in powers of the small parameter ωt. As a result we obtain

$$\mathrm{Re}\, L_c=-\Phi(0)\,(t-2\tau)^2/2, \tag{25}$$

where

$$\Phi(0)=(\Delta JSz)^2 \sum_{\mathbf{p},\mathbf{p}'} \left(\mathbf{p}\frac{df_{\mathbf{p}'}}{d\mathbf{p}'}\right)^2 n_p. \tag{26}$$

In the latter expression, we have restricted ourselves to the principal term in the expansion in p. Transforming in (26) from summation to integration over the Brillouin zone and taking it into account that $\omega_p \propto \sim JSz^{5/3}p^2$, we obtain

$$\Phi(0) \propto (T/JS)^{5/2}(\Delta JS)^2. \tag{27}$$

Thus, at the instant of time $t=2\tau$, radiation is generated with duration $T_2^* \propto \Phi(0)^{-1/2}$. In the case $T_2^* \ll \tau$, such a response represents a photon echo. It is easy to show that the compatibility of this inequality with the case considered here of short times leads to a limitation on the temperature:

$$T>\omega_0 z^{-1/5}(J/\Delta J)^{4/5}. \tag{28}$$

The compatibility of the latter condition with the low-temperature approximation considered in the present section imposes a limitation on the change of the exchange integral:

$$\Delta J>J/z^{1/4}. \tag{29}$$

We note also that the duration T_2^* of the response does not depend on the interaction radius R_0.

In the second case of long times we make use of the fact that at large t

$$\omega^{-2}(1-2\cos\omega t) \propto \delta(\omega).$$

We then obtain

$$\mathrm{Re}\, L_c=-t/T_2, \tag{30}$$

where

$$T_2^{-1}=(\Delta JSz)^2 z^{4/3} \sum_{\mathbf{p},\mathbf{p}'} \delta(\omega_p-\omega_{p'})\,(2\mathbf{p}\mathbf{p}')^2 n_p. \tag{31}$$

where we limit ourselves in (31), to the principal terms of the expansion in the momenta. Calculating this expression by analogy with (25), we obtain

$$T_2 \sim (\Delta JS)^{-2}(T_c/S)^5/T^4. \tag{32}$$

The last expression shows that the nonlinear response of the system at long times decays exponentially, and the decay rate has a strong power-law dependence on the temperature. To conclude this section, we note that its results have meaning, of course, only for the condition $T_2^* < T_2$. However, it is easy to verify that the limitation imposed by this inequality on the permissible change of the exchange integral is much weaker than the condition (29).

4. HIGH TEMPERATURES, $T \gg T_c$

At high temperatures, when the system is in the paramagnetic phase, the magnon description is no longer applicable and the results of the previous section are not valid. However, photon echo can again be observed if the interaction of the exciton with the magnetic medium, which is characterized by the parameter J', significantly exceeds the nonrenormalized exchange interaction J. In this case the spin (of the exciton) and its surroundings, in the limits of the interaction radius, can be regarded as a quasi-isolated subsystem. The echo arises because of the scatter of the eigenenergies of the considered subsystem which effectively plays the role of inhomogeneous broadening.

In the paramagnetic phase, the nonlinear response can in turn be calculated only under the conditions

$$J \ll T/Sz,\ J';\quad z \gg 1. \tag{33}$$

For simplicity, we limit ourselves to a spin $S = 1/2$. In the paramagnetic phase, by virtue of conditions (33), we can neglect the time dependence in the operator $\delta V_i(t)$ in (15), and also omit the Gibbs factor in (14). Then

$$K(t,\tau) = \mathrm{Sp}\{\exp[-i\delta V_i(t-2\tau)]\}/2^{z+1}, \tag{34}$$

Here the trace is taken over all states of the chosen site i and of the z sites located within the interaction radius.

Calculation of this expression is most simply carried out on the basis of the eigenfunctions of the operator δV_i. It is obvious that each eigenvalue of this operator is completely determined by the parameter σ—the total spin of the z surrounding sites and the total spin of all $z + 1$ sites, which is thus equal to $\sigma \mp 1/2$. Then we have for the eigenvalues of the operator δV_1

$$E_{1\sigma} = J'\sigma/2, \quad E_{2\sigma} = -J'(\sigma+1)/2. \tag{35}$$

The multiplicities $g_{i,\sigma}$ of the degeneracy of these eigenvalues are respectively equal to

$$g_{1\sigma} = 2(\sigma+1)n(\sigma), \quad g_{2\sigma} = 2\sigma n(\sigma), \tag{36}$$

where[8] the quantity

$$n(\sigma) = C_z^{z/2-\sigma} - (1-\delta_{\sigma,z/2})C_z^{z/2+\sigma+1} \tag{37}$$

is the number of levels formed by the z spins $S = 1/2$ and having a total spin σ. The remaining components in (35) take into account the degeneracy of the eigenvalues of the operator δV_i from the projection of the total spin of the $z + 1$ sites. Using (35) and (36), we rewrite the correlator (34) in the form

$$K(t,\tau) = \sum_{\sigma=0}^{z/2} (C_z^{z/2-\sigma} - C_z^{z/2+\sigma+1}) \cdot \times\{2(\sigma+1)\exp[-i\sigma J'(t-2\tau)/2] + 2\sigma\exp[-i(\sigma+1)J'(t-2\tau)/2]\}. \tag{38}$$

At large numbers z we can use for the binomial coefficients the asymptotic representation

$$C_z^{z/2-\sigma} = 2^z \exp(-2\sigma^2/z)(2/\pi z)^{1/2}.$$

Transforming in (36) from summation to integration and keeping the leading terms in the parameter z, we obtain

$$K(t,\tau) \propto \exp[-(t-2\tau)^2/(T_2^*)^2], \quad (T_2^*)^{-1} \sim z^{1/2}J'. \tag{39}$$

The time T_2, which characterizes the decay of the quasi-isolated state due to interaction with the surroundings, can be estimated from the formula $T_2 \sim R_0^2/D$, where D is the coefficient of spin diffusion in the paramagnetic phase. We can estimate the latter quantity with the help of the relation $D \sim z^{1/2}JR_0^2$ (see, for example, Ref. 9). Then

$$T_2 \sim z^{1/2}J. \tag{40}$$

For observation of the photon echo, it is necessary that $T_2 > T_2^*$. This obviously occurs at $J' > J$.

5. CONCLUSION

It was thus shown that the photon echo can be observed even in the absence of inhomogeneous broadening, at not very low temperatures, if the system of excitons is sufficiently tightly coupled with the magnetic system. The latter condition is apparently well satisfied if there are rare-earth elements in the system. Then the exchange integral of interest to us can vary over wide limits (by two orders of magnitude), which is quite sufficient for observation of the echo. As shown above, the duration of the exciting pulse should be significantly shorter than the magnon times and can reach 10^2 fs.

In the present work we neglected the possibility of hopping of the excitons between sites. Therefore, from the point of view of the experiment, the materials that might serve as candidates for the discovery of the photon echo in them are those in which the exciton is created on impurity atoms. Such excitons can be regarded with much more justification as localized.

It is known that if a strong local excitation is present in the system, this can lead to a radical rearrangement of the nonrenormalized spectrum, which in turn could lead to new effects. Such a consideration, however, goes beyond the limits of the present work.

In conclusion, we emphasize again photon echo can take place in the considered ferromagnetic system only at a temperature $T > 0$. This is due to the evident fact that at the temperature $T = 0$, upon excitation of the exciton, the magnetic subsystem transforms from its proper ground state into a new, again proper, ground state. Therefore, the required spread of energies can take place only at a temperature different from zero. In this connection, we note that if the exchange interaction in the system were to bear an antiferromagnetic character, then even at the temperature $T = 0$ the transition from the ground state after excitation of the exciton would take place in a nonground state. The latter would contain in itself a superposition of the excited proper states of the locally perturbed antiferromagnet with some energy spread. In this sense, the situation of the antiferromagnet would be close to that of the phonon considered in Ref. 2, where, as was pointed out in the Introduction, the photon echo takes place even at zero temperature.

The study of the photon echo in antiferromagnets, moreover, can turn out to be interesting because of certain singularities of its interaction with light. The fact is that there is a broad class of antiferromagnet in which this interaction cannot conserve the spin of the site[3] and lead upon creation of an exciton, to a change in the projection of the spin on a neighboring site in a sublattice with opposite magnetization. Such materials include, in particular, high-temperature superconductors.[10] However, photon echo in antiferromagnets will be considered in a seperate paper.

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