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MICROSCOPIC MODELLING OF COHERENT EFFECTS IN DIPOLE SPIN SYSTEMS

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Микроскопическое моделирование когерентных эффектов в дипольных спиновых системах

Представляются результаты первого, насколько нам известно, исследования когерентных эффектов в поляризованных спиновых системах на основе микроскопической модели, а не феноменологических уравнений Блоха. Проводится компьютерное моделирование для системы ядерных или элементарных спинов, взаимодействующих посредством реалистических диполь-дипольных сил. Анализируются различные начальные и внешние условия спиновой системы, как связанной, так и несвязанной с резонатором, в присутствии внешнего переменного поля, или без накачки. Поскольку феноменологические уравнения предполагают однородность системы, понятно, что только микроскопическая модель позволяет аккуратно показать, когда в действительности появляется когерентность. Для этого вводятся коэффициенты когерентности и тщательно исследуется их временное поведение. Подчеркиваются особенности, связанные с дипольными взаимодействиями спинов и отсутствующие в феноменологической трактовке, основанной на уравнениях Блоха.

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Microscopic Modelling of Coherent Effects in Dipole Spin Systems

Here we present the first, to our knowledge, investigation of coherent effects in polarized spin systems on the basis of a microscopic model but not using the phenomenological Bloch equations. A computer simulation is realized for a system of nuclear or electron spins interacting with realistic dipole-dipole forces. Different initial and external conditions are analyzed for a spin system either coupled with a resonator or not, in the presence of an external oscillating field or without this pumping. As far as phenomenological equations presuppose the uniformity of a system, it is only a microscopic model that is able to accurately show when the coherence does really appear. To this end, we introduce the coherence coefficients and thoroughly consider their time behaviour. The peculiarities due to dipole spin interactions, which are not present in the Bloch equations, are noted.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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I. INTRODUCTION

Close analogy between optical and radiofrequency superradiance is well acknowledged. Optical coherent superradiance is thoroughly studied both experimentally and theoretically. For review one can consult Refs. 1-5. Radiofrequency superradiance is known a bit less, although it has been observed in a number of experiments [6–17] and theoretically considered in several papers [18–22].

All theoretical considerations of radiofrequency superradiance have been done by using the phenomenological Bloch equations. The latter treat the whole system as a uniform object having unique total magnetization. Therefore, the assumption of coherence is already incorporated into the Bloch approximation. In this way, the Bloch equations are not able to describe the onset of coherence and to accurately study the peculiarities of coherent effects occurring in real spin systems. It is just the aim of the present paper to give a thorough analysis of the latter questions by considering a microscopic spin model with realistic dipole interactions.

II. DIPOLE SPIN MODEL

Let us consider a system of N spins whose sites in real space are enumerated with the index $i = 1, 2, ..., N$, and which interact with each other through dipole forces. The Hamiltonian of this system can be written [23] in the form

$$\hat{H} = \frac{1}{2}\sum_{i\neq j} H_{ij} - \mu \sum_i \vec{S}_i \vec{H}_{eff} , \qquad (1)$$

in which the dipole interaction is

$$H_{ij} = \frac{\mu^2}{r_{ij}^3}\vec{S}_i\vec{S}_j - \frac{3\mu^2}{r_{ij}^5}(\vec{S}_i \cdot \vec{r}_{ij})(\vec{S}_j \cdot \vec{r}_{ij}) , \qquad (2)$$

where $\vec{S}_i$ is a spin operator,

$$\mu \equiv \gamma\hbar , \qquad \vec{r}_{ij} \equiv \vec{r}_i - \vec{r}_j , \qquad r_{ij} \equiv | \vec{r}_{ij} | ,$$

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γ is the gyromagnetic ratio whose sign coincides with the sign of a particle charge; $\vec{H}_{eff}$ is an effective magnetic field acting on the system.

The total effective field $\vec{H}_{eff}$ can contain a constant external field $H_0\vec{e}_z$, an alternating magnetic field $H_1\vec{e}_x\cos\omega t$ and, when the sample is placed into a resonator, a back acting field $H_{ind}\vec{e}_{res}$ induced by rotating spins of the system:

$$\vec{H}_{eff} = H_0\vec{e}_z + H_1\vec{e}_x\cos\omega t + H_{ind}\vec{e}_{res} \,. \tag{3}$$

Consider a case of a cylindrical resonator with the axis directed along $\vec{e}_{res}$. Let, for definiteness, this be a coil having n turns of cross section A_{res}, a length l, resistance R, inductance L and capacity C. The back acting magnetic field of a high quality resonator, following Bloembergen and Pound [18], can be written as

$$H_{ind} = \frac{4\pi n}{clR}U_{ind} \,, \tag{4}$$

where the voltage induced in the coil by moving spins is

$$U_{ind} = -\frac{4\pi}{c}n\eta\rho A_{res}\frac{\mu}{N}\sum_i \dot{\vec{S}}_i\vec{e}_{res} \,; \tag{5}$$

η being a filling factor; ρ, a density of spins,

$$\eta \approx \frac{V}{V_{res}} \,, \qquad V_{res} = lA_{res} \,, \qquad \rho = \frac{N}{V} \,. \tag{6}$$

For the induced magnetic field (4) we get the expression

$$H_{ind} = -\frac{4\pi\mu}{\omega N}\eta\rho Q\sum_i \dot{\vec{S}}_i\vec{e}_{res} \,; \tag{7}$$

in which Q is the quality factor of the circuit,

$$Q \equiv \frac{\omega L}{R} \,; \qquad \omega = \frac{1}{\sqrt{LC}} \,, \qquad L = \frac{4\pi n^2}{lc^2}A_{res} \,, \tag{8}$$

and the characteristic circuit frequency ω is assumed to be in resonance with the frequency of the alternating field in Eq. (3).

The difference between the classical case considered by Bloembergen and Pound[18] and the quantum case is in the meaning of the notation $\dot{\vec{S}}_i$. In the classical case

this is nothing but a time derivative of the spin $\vec{S}_i$. In the quantum case $\dot{\vec{S}}_i$ means an additional operator commuting with spin operators and, under averaging, having the property $\langle\dot{\vec{S}}_i\rangle \equiv d\langle\vec{S}_i\rangle/dt$. Such a definition of the operator $\dot{\vec{S}}_i$ is given so that the Heisenberg equations for the considered system would yield, when returning to the classical case, the corresponding classical equations of motion. Note that if we use the mean-field method, the Hamiltonian of our system would not contain the terms bilinear in spin operators, the term $\vec{S}_j\sum_i\dot{\vec{S}}_i$ will change to $\vec{S}_j\sum_i\left\langle\dot{\vec{S}}_i\right\rangle$ and the commutator of $\dot{\vec{S}}_i$ with $\vec{S}_j$ does not appear. The equations of motion for the classical quantities $\left\langle\dot{\vec{S}}_i\right\rangle$ are the same for both the cases.

To write the Heisenberg equations for the spin operators S_i^z and $S_i^\pm = S_i^x \pm S_i^y$, we direct the coil axis along the axis x, so that $\vec{e}_{res} = \vec{e}_x$, use the notation

$$\omega_0 \equiv \gamma H_0 \,, \qquad \omega_1 \equiv \gamma H_1 \,, \tag{9}$$

and introduce an important quantity, called the coupling constant,

$$g \equiv \frac{2\pi}{\hbar\omega}\eta\rho\mu^2 Q = g_0 N \,; \qquad g_0 \equiv \frac{2\pi\mu^2 Q}{\hbar\omega V_{res}} \,, \tag{10}$$

which describes the strength of a coupling between the spin system and the resonant coil.

In this way, as equations of motion for the microscopic model with Hamiltonian (1) we obtain the following equation for the z–component of a spin operator

$$\begin{aligned} i\hbar\frac{dS_i^z}{dt} &= \frac{\hbar\omega_1}{2}\left(S_i^- - S_i^+\right)\cos\omega t - \frac{g_0\hbar}{2}\left(S_i^- - S_i^+\right)\frac{d}{dt}\sum_j\left(S_j^- + S_j^+\right) + \\ &+ \sum_{j(\neq i)}\Big[\frac{a_{ij}}{4}\left(S_i^- S_j^+ - S_i^+ S_j^-\right) + \left(c_{ij}S_i^+ - c_{ij}^* S_i^-\right)S_j^z + \\ &+ e_{ij}S_i^+ S_j^+ - e_{ij}^* S_i^- S_j^-\Big] \end{aligned} \tag{11}$$

and the equation for the ladder operator

$$i\hbar\frac{dS_i^-}{dt} = -\hbar\omega_0 S_i^- + \hbar\omega_1 S_i^z\cos\omega t - g_0 S_i^z\frac{d}{dt}\sum_j\left(S_j^- + S_j^+\right) +$$

$$+ \sum_{j(\neq i)} \left[\frac{a_{ij}}{2} \left(S_i^z S_j^- + 2 S_i^- S_j^z \right) + c_{ij} \left(S_i^- S_j^+ - 2 S_i^z S_j^z \right) + \right.$$
$$\left. + \; c_{ij}^* S_i^- S_j^- - 2 e_{ij} S_i^z S_j^+ \right] , \tag{12}$$

in which

$$a_{ij} \equiv \frac{\mu^2}{r_{ij}^3} \left(1 - 3 \cos^2 \vartheta_{ij} \right) ,$$
$$c_{ij} \equiv -\frac{3\mu^2}{4 r_{ij}^3} \sin \left(2 \vartheta_{ij} \right) \exp \left(-i \varphi_{ij} \right) ,$$
$$e_{ij} \equiv -\frac{3\mu^2}{4 r_{ij}^3} \sin^2 \vartheta_{ij} \exp \left(-2 i \varphi_{ij} \right) ,$$

where ϑ_{ij} and φ_{ij} are the spherical angles corresponding to the vector $\vec{r}_{ij}$.

The radiation processes occurring in the system can be studied by measuring the power of current absorbed by the coil

$$P(t) = \frac{\langle U_{ind}^2 \rangle}{2R} = P_{inc}(t) + P_{coh}(t) , \tag{13}$$

which is presentable as a sum of the incoherent and coherent parts, respectively,

$$P_{inc}(t) = \hbar g_0 \sum_i \left\langle \dot{S}_i^x \right\rangle^2 , \qquad P_{coh}(t) = \hbar g_0 \sum_{i \neq j} \left\langle \dot{S}_i^x \dot{S}_j^x \right\rangle .$$

Another characteristic is the intensity of magnetodipole radiation

$$I(t) = I_{inc}(t) + I_{coh}(t) , \tag{14}$$

also consisting of two terms

$$I_{inc}(t) = \frac{2\mu^2}{3c^3} \sum_i \left\langle \ddot{\vec{S}}_i \right\rangle^2 , \qquad I_{coh}(t) = \frac{2\mu^2}{3c^3} \sum_{i \neq j} \left\langle \ddot{\vec{S}}_i \ddot{\vec{S}}_j \right\rangle ,$$

the intensities of incoherent and coherent radiation.

The radiation intensity (14) can be measured, in principle, by usual detectors of propagating radiofrequency waves, to distinguish the signal of harmonic pumping and complicated $I(t)$ is not difficult. The main difference between (13) and (14) is that the power of current (13) is observed in a resonance circuit surrounding the

sample considered and bound with it by the back action characterized by the coupling constant (10), while the intensity of radiation (14) should be measured by detectors that are not coupled with the spin system, being situated out of the latter. When the system is not placed inside a resonator, the sole measurable radiation characteristic is intensity (14). Although a problem can arise when measuring the intensity of radiation (14) because of its smallness. For example, in the case of proton spins, if we take $\omega_0 \sim 10^8 s^{-1}$ and $N \sim 10^{22}$, we get $I(t) \sim 10^{-8} W$, which is quite small and is rather difficult to measure. However, in the case of electron spins with $\omega_0 \sim 10^{11} s^{-1}$ and the same number of spins $N \sim 10^{22}$, the intensity $I(t)$ can reach tens of watts and is easily detectable. Therefore, the magnetodipole radiation from protons seems to be too small to be measured, but that from electrons can be easily observed, even though in reality the number of coherently radiating particles N is sufficiently smaller because of inhomogeneous broadening. In all the cases the power of current (13) is higher than the intensity of radiation (14) by a factor of $Q\lambda^3/(2\pi)^2 V_{res}$, which is quite large for the radiofrequency wavelength λ and for high quality resonators[18].

To study the onset of coherence and all peculiarities of coherent effects in real systems, it is very convenient to introduce the coherence coefficients[24–26] defined here by the equations

$$K_{coh}(t) \equiv \frac{P_{coh}(t)}{P_{inc}(t)} , \qquad C_{coh}(t) \equiv \frac{I_{coh}(t)}{I_{inc}(t)} . \tag{15}$$

Let us stress that the introduction of the coherence coefficients (15) is based on the possibility of separating out in Eqs. (13) and (14) of the corresponding incoherent and coherent terms, which is admissible only for a microscopic model.

III. RESULTS OF NUMERICAL INVESTIGATIONS

We solve the equations of motion (11) and (12) by using a standard method of computer simulation which is applied for treating the dynamics of spin systems [27,28]. In this approach spins are considered as classical vectors, their initial distribution is given by the Monte-Carlo technique, and the differential equations of motion are numerically solved by the Runge-Kutta method.

To check that the qualitative behavior of the system does not depend on the number of spins, we have realized three variants of calculations with $N = 27$, 125 and 343 . For all these cases the time behavior of the system has been found to be qualitatively the same. Therefore,in what follows we present the results of calculations accomplished with $N = 125$.

Everywhere below time is measured in units of $T_2 \equiv \hbar a^3/\mu^2$, where a is a mean interparticle distance, and frequencies in units of T_2^{-1} . We have calculated the coherence coefficients $K_{coh} = K_{coh}(t)$ and $C_{coh} = C_{coh}(t)$ and the radiation characteristics (13) and (14) which, for convenience, are made dimensionless by passing to the quantities

$$P = \frac{T_2^2}{\hbar g_0} P(t) , \qquad I = \frac{3c^2 T_2^4}{2\mu^2} I(t) .$$

We present as well the average polarization

$$P_z(t) \equiv \frac{1}{N} \sum_i \langle S_i^z \rangle = P_z(t) ,$$

in which the spin of a particle is assumed to be 1/2.

It is worth of noting that in our microscopic model we do not take into account the spin-lattice interaction as far as its intensity is much smaller than that of the dipole interaction (2), that is, the spin-lattice relaxation time is much larger than T_2.

The polarized spin system is supposed to be prepared in a strongly nonequilibrium state. This means that if $p_z^{(0)} > 0$, then the external magnetic field is overturned in the case of positively charged particles $(H_0 < 0)$ and is parallel to $\vec{e}_z$ in the case of negatively charged particles $(H_0 > 0)$. In both the cases $\omega_0 = \gamma H_0$ is to be negative if $p_z^{(0)}$ is positive. When the initial polarization $p_z^{(0)}$ is negative, then ω_0 is to be positive in order to make the initial state strongly unstable. Therefore, the general condition showing that the system is initially in a nonequilibrium state is $\omega_0 p_z^{(0)} < 0$.

Figures 1-3 show a transition of the spin system from such a strongly nonequilibrium state to its equilibrium state when the system is coupled with a resonance circuit $(g_0 \neq 0)$ but the alternating pumping field is absent $(\omega_1 \equiv 0)$. Figure 1 demonstrates the influence of the coupling constant g_0 on the delay time and on the duration of a radiation pulse. The dependence of the latter characteristics on the value of ω_0 and on the initial polarization $p_z^{(0)}$ is illustrated in Fig. 2 and Fig. 3, respectively. In Figs. 1-3 the time behavior of the power P is completely analogous to this function measured in the corresponding experiments[13-17]. The first coherent burst is typical of superradiance [11,12] when $P \sim N^2$, after which the incoherent maser generation continues. The Bloch equations, which assume the existence of coherence, can reasonably describe the superradiating pulse itself, but are not able to describe the incoherent maser generation (see Refs.11,12,19-22). This is because the Bloch equations correspond to the classical approximation, while the incoherent radiation is of quantum nature. Our microscopic model allows us to picture the whole process with both its coherent and incoherent parts and to obtain a good agreement with experiment[13-17]. By tracing the time behavior of the coherence coefficient, defined in (15), we can unambiguously decide when the process is really coherent and when is not.

If the spin system is not coupled with a resonator $(g_0 \equiv 0)$ and there is no alternating pumping $(\omega_1 \equiv 0)$, then the coherence can appear only in the situation typical of free induction, when the initial polarization $p_x^{(0)}$ is more important than $p_z^{(0)}$. This case is illustrated in Fig.4. The description of free induction by the Bloch

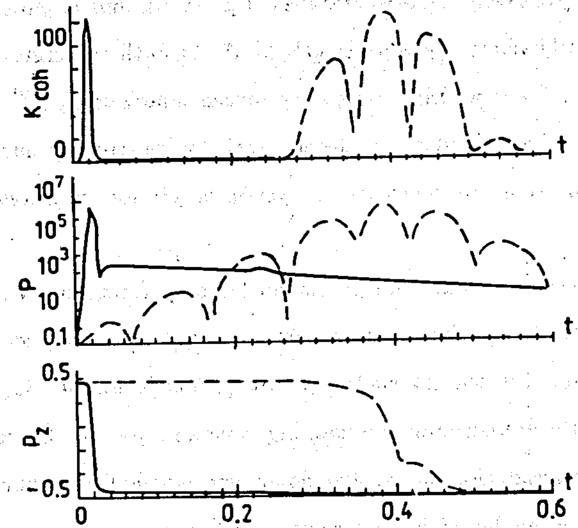

FIG.1. The coherence coefficient K_{coh}, power of current P and polarization p_z as functions of time (measured in units T_2) for the case of the spin system coupled with a resonator but without external pumping $(\omega_1 \equiv 0)$. The solid line is for $g_0 = 0.1$; the dashed line, for $g_0 = 0.01$. The Zeeman frequency is $|\omega_0| = 40$ (in units of T_2^{-1}).

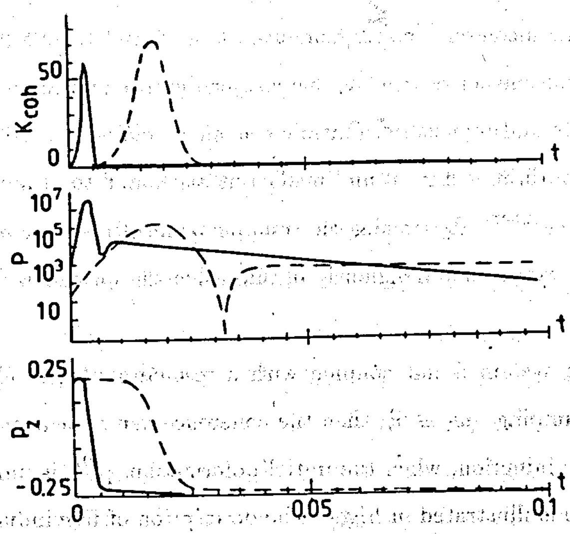

FIG.2. The same functions as in Fig.1 for the parameters $g_0 = 0.1$ and $p_z^{(0)} = 0.25$. The solid line is for $|\omega_0| = 200$; dashed line, for $|\omega_0| = 40$.

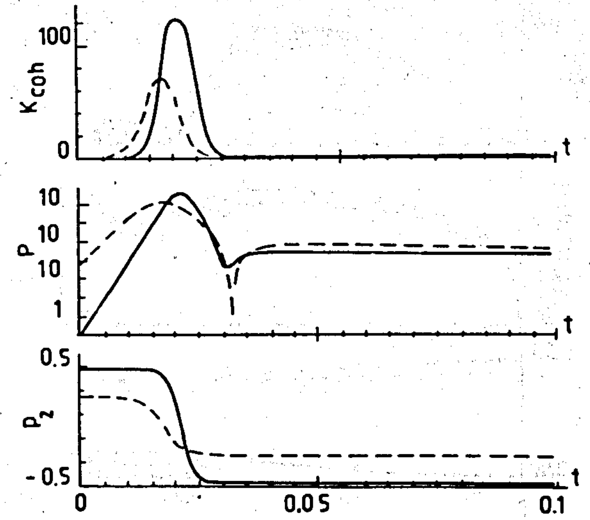

FIG.3. The same functions as in Fig.1 for the parameters $g_0 = 0.1$ and $|\omega_0| = 40$. The solid line is for $p_z^{(0)} = 0.475$; dashed line, for $p_z^{(0)} = 0.250$.

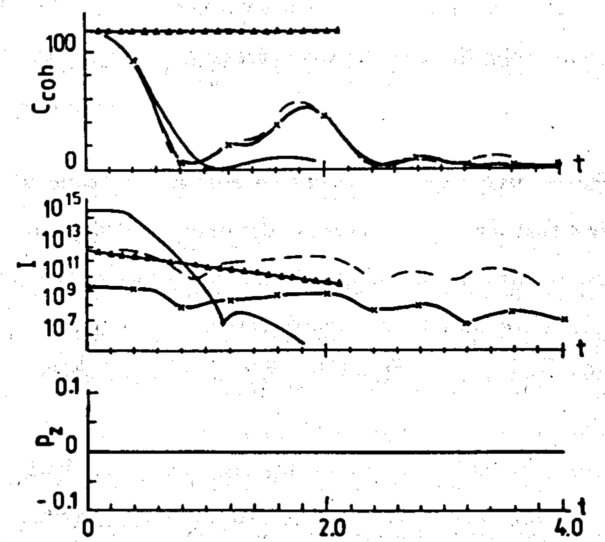

FIG.4. The coherence coefficient C_{coh}, radiation intensity I and polarization p_z as functions of time in the case of the spin system without a resonator $(g_0 \equiv 0)$ and without pumping $(\omega_1 \equiv 0)$ for $p_z^{(0)} = 0.475$. The solid line is for $|\omega_0| = 1000$; dashed line, for $|\omega_0| = 200$; solid line marked with crosses, for $|\omega_0| = 50$; solid line marked with triangles, for $|\omega_0| = 200$ but without dipole interactions. All corresponding curves for the polarization p_z practically coincide.

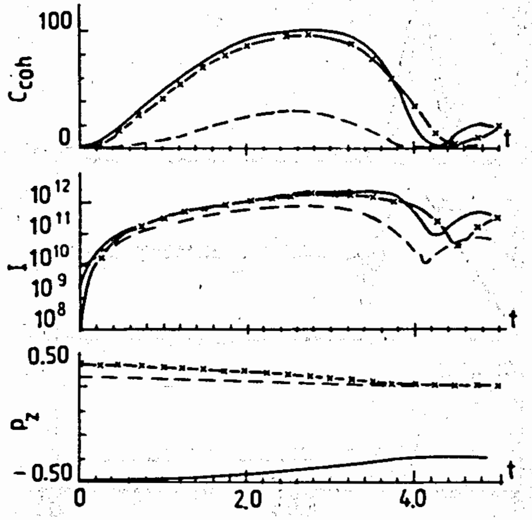

FIG.5. The same functions as in Fig.4 in the case of the spin system without a resonator $(g_0 \equiv 0)$ but in the presence of an external pumping $(\omega = \omega_0)$ for the parameters $|\omega_0| = 200$ and $|\omega_1| = 0.5$. The solid line is for $p_z^{(0)} = -0.475$; dashed line, for $p_z^{(0)} = 0.375$; solid line marked with crosses, for $p_z^{(0)} = 0.475$.

equations displays the disappearance of coherence during the time T_2. Contrary to this, we see in Fig.4 that for $t > T_2$ there are oscillations of coherence with a period close to T_2. These oscillations are due to the nonuniformity of the system, since in our case not all spins have initially the same direction. As is evident, such oscillations of coherence cannot appear in the Bloch description where all spins, by supposition, are unidirected.

Finally, we show that coherence can be obtained in a system without a resonator $(g_0 \equiv 0)$ but in the presence of a resonance external pumping $(\omega_1 \neq 0)$. This is demonstrated in Fig.5. Such a regime cannot be accurately described by the Bloch equations, since, as has been discussed by Redfield [29], with radiofrequency fields, when the energy of spin alignment in these fields is comparable to the energy of typical dipole spin interaction, the simple phenomenological concept of a T_2 relaxation time breaks down.

In conclusion, we would like to emphasize that the model considered in the present paper makes it possible to correctly portray different coherent effects in spin systems not only because this model is microscopic but also owing to a direct numerical solution of nonlinear equations of motion. If we would start from a microscopic description but invoking perturbation theory, as was done in Ref. 30 at very high polarizations p_z such as discussed in our work, then we would be able to depict solely an incoherent behavior of the system.

References

1. L. Allen and J. Eberly, Optical Resonance and Two-Level Atoms (Wiley, New-York, 1975).

2. A.V. Andreev, V.I. Emelyanov and Y.A. Ilyinsky, Cooperative Phenomena in Optics (Nauka, Moscow, 1988).

3. Problems of Quantum Optics, edited by V.I. Yukalov (Joint Inst. Nucl. Res., P17-88-689, Dubna, 1988).

4. Interaction of Electromagnetic Field with Condensed Matter, edited by N.N. Bogolubov, A.S. Shumovsky and V.I. Yukalov (World Scientific, Singapore, 1990).

5. P.V. Zinoviev, V.V. Samartsev and N.B. Silaeva, Laser Phys. **1**, 1 (1991).

6. A. Abragam, J. Combrisson and I. Solomon, Compt. Rend. **245**, 157 (1957).

7. G. Feher, J. Gordon, E. Buchler, E. Gere and C. Thurmond, Phys. Rev. **109**, 221 (1958).

8. P.F. Chester, P.E. Wagner and J.G. Castle, Phys. Rev. **110**, 281 (1958).

9. V.M. Fain and Y.I. Khanin, Quantum Radiophysics (Sov. Radio, Moscow, 1965).

10. J.P. Wolfe and A.R. King, Chem. Phys. Lett. **40**, 451 (1976).

11. P. Bösiger, E. Brun and D. Meier, Phys. Rev. Lett. **38**, 602 (1977).

12. P. Bösiger, E. Brun and D. Meier, Phys. Rev. A **18**, 671 (1978).

13. J.F. Kiselev, A.F. Prudkoglyad, A.S. Shumovsky and V.I. Yukalov, Mod. Phys. Lett. B **1**, 409 (1988).

14. Y.F. Kiselev, A.F. Prudkoglyad, A.S. Shumovsky and V.I. Yukalov, Sov. Phys. JETP **67**, 413 (1988).

15. Y.F. Kiselev, A.F. Prudkoglyad, A.S. Shumovsky and V.I. Yukalov, in: Problems of Quantum Optics, edited by V.I. Yukalov JINR, P17-88-689, Dubna, 1988. p.68.

16. N.A. Bazhanov, D.S. Bulyanitsa, A.I. Kovalev, V.V. Polyakov, V.Y. Trautman, E.D. Trifonov and A.V. Shvedchikov, Fiz. Tverd. Tela **31**, 206 (1989).

17. Y.F. Kiselev, A.S. Shumovsky and V.I. Yukalov, Mod. Phys. Lett. B**3**, 1149 (1989).

18. N. Bloembergen and R.V. Pound, Phys. Rev. **95**, 8 (1954)

19. S. Bloom, J. Appl. Phys. **28**, 800 (1957).

20. A. Yariv, J. Appl. Phys. **31**, 740 (1960).

21. W.H. Krause, Z. Phys. **219**, 434 (1969).

22. A.V. Andreev, Usp. Fiz. Nauk **160**, 1 (1990).

23. C.P. Slichter, Principles of Magnetic Resonance (Springer, Berlin, 1980).

24. V.I. Yukalov, J. Mod. Opt. **35**, 35 (1988).

25. V.I. Yukalov, in: Abstracts of Symposium on Photon Echo and its Application (Samara, 1989), p.163.

26. V.I. Yukalov, J. Mod. Opt. **37**, 1361 (1990).

27. S. Jensen and O. Platz, Phys. Rev. B **7**, 31 (1973).

28. T.S. Belozerova and E.K. Henner, Fiz. Tverd. Tela **26**, 83 (1984).

29. A. Redfield, Phys. Rev. **98**, 1787 (1955).

30. E.K. Henner and V.K. Henner, Physica A **172**, 431 (1991).

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