Entanglement in electron-nuclear spin system as immediate cause of the dependence of dephasing rate $1/T_2$ on intensity in the optical Bloch equations

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A B S T R A C T

Dephasing rate $1/T_2$ in the optical Bloch equations doesn’t depend on Rabi frequency $\chi = Ed/h$. It seemed true for various mechanisms responsible for optical dephasing processes. In this paper we show that entanglement of nuclear spin with electronic transition in an atom results in dependence of $1/T_2$ on $\chi$. Rate $1/T_2(\chi)$ decreases if $\chi$ increases. This effect is able to explain experimental data on free-induction decay measured in Pr$_3^+$:LaF$_3$ crystal.

1. Introduction

The Bloch equations derived for magnetic resonance [1] play important role in optical resonance as well [2]. Physical idea enabling us to use Bloch equations for optical resonance was based on formal analogy between two-state spin system and two-level atom (molecule).

Nevertheless, some doubts emerged that the optical Bloch equations are able to describe all details of the optical resonance. Therefore, DeVoe & Brewer in their paper [3] entitled “Experimental Test of Optical Bloch Equations for Solids” decided to verify applicability of the optical Bloch equations for crystals with paramagnetic ions. Result of their measurement of free-induction decay (FID) in Pr$^{3+}$:LaF$_3$ crystal was in strong disagreement with the decay predicted by the optical Bloch equations.

Crystal Pr$^{3+}$:LaF$_3$ was chosen for FID experiments because values of $T_1 = 0.5\mu$s and $T_2 = 21.7\mu$s for this crystal were found earlier in Refs. [4,5]. Excitation of ions Pr$^{3+}$ was realized via $^1D_2 \rightarrow ^3H_4$ transition at wave length 5925 Å during 400 μs. After that laser was switched off and light emitted by induced electronic polarization was detected. In accordance with the optical Bloch equations dependence of emission on time is described by the following function \[ \Delta \omega(\chi) = \sqrt{(1/T_1)^2 + \chi^2(T_2/T_1)}, \] where $\Delta \omega(\chi)$ is rate of the decay depending on Rabi frequency $\chi = Ed/h$, and $\omega_0$ is a resonant frequency. Eq. (1) doesn’t include unknown parameters because values $T_1 = 0.5\mu$s and $T_2 = 21.7\mu$s have been measured in independent experiments earlier [4,5]. Dependence of $\Delta \omega(\chi)$ on Rabi frequency described by Eq. (1) is shown in Fig. 1 by solid line whereas the dependence measured is shown by dots.

We see strong disagreement with prediction derived from the optical Bloch equations. At small values of $\chi$ the measured function $\Delta \omega(\chi)$ is in agreement in accordance with the prediction of the optical Bloch equations. However, at large values of $\chi$, the measured function $\Delta \omega(\chi)$ is increased approximately as $\chi$ instead of the dependence $\chi(T_1/T_2)^{1/2}$ predicted by the optical Bloch equations.

Later Szabo and Muramoto [6] have found that FID in ruby demonstrates disagreement with Eq. (1) similar that shown in Fig. 1. It became clear that the optical Bloch equations cannot describe transient phenomena in paramagnetic crystals without serious modification in these equations.

Therefore, immediately many theories have been advanced for modification of the optical Bloch equations aimed to explain disagreement found in the experiment. Influence of Markovian [7–17] and non-Markovian [18,19] processes on the optical Bloch equations have been analyzed.

Analysis of these theories has been carried out by Berman [14,15], Javanainen [10], Szabo and Muramoto [6]. As it was pointed out by Szabo and Muramoto in accordance with analysis made by Berman and Javanainen “…none of the numerical calculations presented so far consistently describe the data for Pr$^{3+}$:LaF$_3$” Summing up their own analysis Szabo and Muramoto [6] pointed out «We have found that the Gaussian-Markov or random-telegraph modifications of the optical Bloch equations do not consistently describe free-induction and echo-decay measurements with a single correlation time for the assumed stochastic frequency fluctuations. It is suggested that a nonstochastic model …may be useful to study”
2. Hamiltonian of electron-spin system without entanglement

In accordance with Ref. [3], optical dephasing in Pr$^{3+}$:LaF$_3$ crystal results from the dipolar interaction between spin in nucleus $^{19}$F and electronic transition in Pr$^{3+}$ ion. Such electron-spin system mathematically looks similar to electron-two level (TLS) system considered in many works for a guest molecules embedded to polymer or glass. Non-perturbative theory for electron-TLS system has been developed earlier by Osad’ko [20–22]. This very theory for electron-TLS system we take as basis for our consideration of electron-spin system in paramagnetic crystals with guest ions.

The Hamiltonian of electron-spin system interacting with light we take in the following form:

$$H = H_0 + H_{\text{phus}} + H_{\text{el--phus}}.$$  \hspace{1cm} (2)

$$H_0 = \hbar \omega_0 B^+ B + \epsilon c^+ c + \hbar \omega_0 B^+ B.$$  \hspace{1cm} (3)

Here $B^+$ and $B$ are operators of creation and annihilation of electronic excitation, $c^+$ and $c$ are operators of creation and annihilation of excitation in spin system.

Our 4-state system consists of two-level atom with two electronic states described by populations $n_{0,1}$ and two nuclear spin states with populations $\rho_0$ and $\rho_1$. This 4-state system with Hamiltonian (3) is described by the diagram presented in Fig. 2.

Operator $V^{(d)}_{c\gamma}$ describes electron-spin interaction that is responsible for optical dephasing. Probabilities $n_0\rho_0$, $n_1\rho_\gamma$, $n_0\rho_\gamma$, $n_1\rho_0$ of finding four quantum states are products of electronic probabilities $n_{0,1}$ and spin probabilities $\rho_{0,\gamma}$:

$$P_0 = n_0\rho_0, \quad P_1 = n_1\rho_\gamma, \quad P_2 = n_0\rho_\gamma, \quad P_3 = n_1\rho_0.$$  \hspace{1cm} (4)

Here $\rho_\gamma = f = [1 + \exp(i/kT)]^{-1}$. $\rho_\gamma = 1 - f$ describes spin probability in thermal equilibrium. They are independent on state of Pr$^{3+}$ ion.

Operator $V^{(d)}_{c\gamma} = \hbar \omega_0 c^+ c B^+ B$ of the interaction is diagonal with respect to electron and spin variables. This operator is responsible for appearance of optical dephasing in the system. Calculation of optical dephasing in the system which is described by Hamiltonian $H_0$ is presented in Appendix A. Final Eq. (A19) for optical dephasing rate in the electron-spin system depends solely on temperature via function $f = [1 + \exp(i/kT)]^{-1}$.

3. Entanglement in electron-spin system excited by light

Let us include in Hamiltonian (3) operator

$$V^{(d)}_{c\gamma} = V^0(c + c^+) + V^1 B^+ B(c + c^+).$$  \hspace{1cm} (5)

that is off-diagonal with respect to spin operators. In presence of this operator electronic and spin variables are entangled and we arrive at the energy diagram presented in Fig. 3.

Here rate constants are described by the following equations:

$$a = 2\pi \sum_{a,b} w_{a,b}(\beta)(\omega_{a,b}^2)\delta(\Omega_\alpha - \Omega_\beta - \epsilon/h)$$  \hspace{1cm} (6a)

$$A = 2\pi \sum_{a,b} w_{a,b}(\beta)(\omega_{a,b}^2)\delta(\Omega_\alpha - \Omega_\beta - \epsilon/h)$$  \hspace{1cm} (6b)

$$B = 2\pi \sum_{a,b} w_{a,b}(\alpha)(\omega_{a,b}^2)\delta(\Omega_\alpha - \Omega_\beta - \epsilon/h + \Delta)$$  \hspace{1cm} (6c)

$$b = 2\pi \sum_{a,b} w_{a,b}(\alpha)(\omega_{a,b}^2)\delta(\Omega_\alpha - \Omega_\beta - \epsilon/h + \Delta)$$  \hspace{1cm} (6d)

Here $\Omega_\alpha$ and $\Omega_\beta$ are frequencies of phonon excitations. Two left levels in Fig. 3 describe two electronic states with nucleus spin in $\alpha$ state. Two right levels describe the same electronic levels when nucleus is in $\beta$ spin state.

If $V^{(d)}_{c\gamma} \neq 0$, constants $a,A,b$ and $B$ are not equal zero and we arrive at the following four rate equations for the populations $P_0 = P_1 = P_2 = P_3$:

$$\dot{P}_0 = -k(\epsilon)P_0 + k(\gamma)P_1 + k(\gamma)P_2 + k(\beta)P_3,$$

$$\dot{P}_1 = k(\epsilon)P_0 - (\gamma + k)P_1 + bP_3,$$

$$\dot{P}_2 = aP_0 + 0 - AP_2 + P_3,$$

$$\dot{P}_3 = 0 + bP_1 - (b + 1/kT)P_1.$$  \hspace{1cm} (7)

Here $k(\epsilon) = k(\gamma) + 1/kT$. Rate $k$ of electronic excitation is given by:

$$k(\epsilon) = \frac{\chi^2}{2} \frac{1/kT}{2} \frac{1}{\Delta T^2 + (1/kT)^2},$$

where $\Delta = \omega - \omega_0$. Rates $A$, $a$, $b$, $B$ describe jumps from spin state $\alpha$ to spin state $\beta$ and back. Probabilities $P_j$ found from Eq. (7) cannot be written as product of electronic and spin probabilities similar to Eq. (4).

Hence Eq. (7) describes evolution in time of entangled electron-nuclear spin states.

Consider the following two probabilities for spin:

$$1 - f = P_0 + P_1, \quad f = P_2 + P_3.$$  \hspace{1cm} (9)

They describe probabilities of finding spin in state $\alpha$ and $\beta$, respectively. Eq. (9) determines probabilities which should be inserted to Eq. (A4) for the spin density matrix in Appendix A. In such case Eq. (A4) will look as follows:

$$\hat{\rho}^{(s)} = (P_0 + P_1)c^+ c + (P_2 + P_3)c^+ c.$$  \hspace{1cm} (10)
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