Transient processes and optical Bloch equations

R. N. Shakhmuratov
Physicotechnical Institute, Kazan' Science Center of the Academy of Sciences of the USSR

(Submitted 22 March 1990)
Pis'ma Zh. Eksp. Teor. Fiz. **51**, No. 9, 454–456 (10 May 1990)

The anomalous behavior of transient optical processes in ruby is not associated with a change in phase relaxation.

Several recent experimental studies have cast doubt on the validity of the optical Bloch equations and the reliability of the assumption that the relaxation times which appear in them remain constant. The observed widths $\Delta\nu_D$ of the holes burned by laser beams in inhomogeneously broadened lines of the Pr^{3+}:LaF_3 crystal[1] and the Cr^{3+}:Al_2O_3 crystal[2,3] do not agree with the theoretical predictions based on the optical Bloch equations. It has been suggested in these studies that phase relaxation is slowed during the application of an intense laser beam, and the observed behavior has been explained successfully on the basis of the assumption that the phase relaxation time T_2 lengthens to T_1 (the energy relaxation time). However, a direct test of the possibility of a slowing of the phase relaxation in a nutation-echo experiment yielded a negative result.[4] Over a broad range of intensities (the Rabi frequency χ, proportional to the field amplitude, was varied from 215 kHz to 1800 kHz) no change was observed in the phase relaxation time. On the other hand, the width of the burned hole, $\Delta\nu_D$, was observed to deviate from that predicted by the optical Bloch equations in the same sample, but at substantially lower intensities (the Rabi frequency was varied from 10 to 85 kHz).[3] In the present letter we propose an explanation for the observed behavior on the basis of the following assumption: No change in T_2 occurred in the experiments of Refs. 2 and 3, and all effects stemmed from an ambiguity in the determination of the energy relaxation time.

To illustrate the suggestion, we examine the phase relaxation of a Cr^{3+} dopant ion in Al_2O_3 on the transition $(-1/2)^4A_2-(-1/2)E(^2E)$. According to the present understanding, this relaxation stems from fluctuations of the magnetic moments of nuclei surrounding the paramagnetic dopant. The nuclei act through the superhyperfine interaction to cause a random shift of the resonant frequency of the ion, which in turn causes an optical dephasing. It is assumed here that remote nuclei play a leading role, since the magnetic moments of the nuclei nearest the dopant are frozen by virtue of the strong superhyperfine interaction. In the present letter we suggest a fundamentally new model for the hole-burning process, on the basis of a modification of the dephasing mechanism. This new model can be summarized as follows: The Al^{27} nuclei surrounding the paramagnetic Cr^{3+} dopant have an electric quadrupole moment. The interaction of this moment with the gradient of the crystal field leads to a nonequidistant arrangement of the energy levels of the nuclear spins in the magnetic field. This nonuniformity lifts the prohibition against mutual flips of the spins nearest the dopant with remote nuclear spins, since there is a partial coincidence of resonant frequencies (between 3 and 32 kHz) of the remote nuclei and the N nucleus, of N and L nuclei, of

0021-3640/90/090513-04$01.00

L and K nuclei, etc. Here we are using the notation adopted in Ref. 5 for the nuclei in nonequivalent positions. As a result, the dipole–dipole interaction runs into no substantial barrier in the transfer of magnetization from nearest neighbors to nuclei remote from the dopant, so a fluctuation of the local field at the dopant due to nearest neighbors is possible. This fluctuation leads to a random shift of the resonant frequency of the dopant, ω_0, in the interval $(\omega_0 - \bar{\Delta}/2, \omega_0 + \bar{\Delta}/2)$, $\bar{\Delta} = 1.2$ MHz. Another source of fluctuations might be mutual flips of spins in nonequivalent positions in which the total energy of the nearest neighborhood is conserved. The local field changes because of a conversion of the quadrupole-interaction energy into energy of the superhyperfine interaction. The rate of the fluctuations in the superhyperfine interaction might be higher than the rate of the nuclear flip-flop process in the interior, since the nearest nuclei are coupled more strongly with each other because of the indirect interaction through the paramagnetic dopant. Each process leads to a random shift of the resonant frequency of the ion, with a long correlation time $\tau_c \gg \bar{\Delta}^{-1}$, which is characteristic of slow spectral diffusion. Following Ref. 6, we approximate the frequency change by an uncorrelated discontinuous Markov process. All the dopant ions can then be allotted to magnetic packets, each with its own deviation Δ_i from ω_0 due to the superhyperfine interaction. In this model, flip-flop processes lead to a transition of the ion from state i to state j with a deviation Δ_j. The probability density for a transition per unit time is given by $F(\Delta_j)/\tau_c$, where $F(\Delta_j)$ is the equilibrium distribution function of the packets. The equation describing the behavior of an individual packet in the resonant field is

$$\frac{\partial u(\delta,\Delta)}{\partial t} = -(\delta+\Delta)v(\delta,\Delta) - \frac{1}{\tau_c}u(\delta,\Delta) + \frac{F(\Delta)}{\tau_c}\int u(\delta,\Delta')d\Delta';$$

$$\frac{\partial v(\delta,\Delta)}{\partial t} = (\delta+\Delta)u(\delta,\Delta) + \chi w(\delta,\Delta) - \frac{1}{\tau_c}v(\delta,\Delta) + \frac{F(\Delta)}{\tau_c}\int v(\delta,\Delta')d\Delta';$$

$$\frac{\partial w(\delta,\Delta)}{\partial t} = -\chi v(\delta,\Delta) - \frac{1}{\tau_c}w(\delta,\Delta) + \frac{F(\Delta)}{\tau_c}\int w(\delta,\Delta')d\Delta' - \frac{1}{T_1}[w(\delta,\Delta) + F(\Delta)g(\delta)], \qquad (1)$$

where u, v, w are the following combinations of elements of the dopant density matrix $\hat{\rho}$: $w = \rho_{22} - \rho_{11}$; $u + iv = \rho_{12}e^{i\Omega t}$. The indices 1 and 2 correspond to resonant energy levels E_1 and E_2; Ω is the frequency of the laser light; $\Delta + \delta = (E_2 - E_1)/\hbar - \Omega$; Δ is the given realization of the frequency shift due to the superhyperfine interaction; Δ' are all other realizations; and δ is the frequency shift due to the static inhomogeneous broadening, which is determined by the distribution function $g(\delta)$. The experiments of Ref. 3 used a laser light source with a narrow line $\Delta\nu < 2$ kHz. Since the frequency spread $\bar{\Delta}$ due to the superhyperfine interaction is substantially greater than $\Delta\nu$, τ_c^{-1}, and the Rabi frequency ($\chi < 100$ kHz; Ref. 3), a small number of particles, in the interval $|\Delta + \delta| \lesssim \chi$ are excited in the sample. Accordingly, the integrals on the right sides of the equations for the components u and v (the "incoming" terms) make a small contribution, on the order of $\chi/\overline{\Delta} \ll 1$, in comparison with that of the terms which describe the transition of the dopant out of the state with a frequency difference Δ. In the equation for the population difference w, the situation is the opposite: This integral is equal within $\chi/\overline{\Delta}$ to its equilibrium value $-g(\delta)F(\Delta)/_{\tau_c}$. Accordingly, Eq. (1) can be reduced to an ordinary Bloch differential equation with relaxation times $\widetilde{T}_2 = \tau_c$ and $\widetilde{T}_1 = (1/\tau_c + 1/T_1)^{-1}$. Solving this equation, we find the following dependence on the width parameters of the burnt hole: $\Delta\nu_D = [(1/\widetilde{T}_2^2) + \chi^2(\widetilde{T}_1/\widetilde{T}_2)]^{1/2}$. If $\tau_c \ll T_1$, the relaxation times are equal: $\widetilde{T}_1 = \widetilde{T}_2 = \tau_c$. In the experiments of Refs. 2 and 3 ($\tau_c < \chi \ll \overline{\Delta}$), the value of $\Delta\nu_D$ was thus nearly equal to the Rabi frequency. We recall that the assumptions $\widetilde{T}_2 = T_2$ and $\widetilde{T}_1 = T_1(T_2 \ll T_1)$ were made in Ref. 1, so the theory predicted $\Delta\nu_D \gg \chi$. The observed agreement of $\Delta\nu_D$ with χ was interpreted as an intensity dependence of T_2: $T_2 \to T_1$ with increasing χ. In strong fields, in which the Rabi frequency becomes comparable to the interval $\overline{\Delta}$, we cannot ignore the integral terms in Eq. (1). In this case $\widetilde{T}_1$ increases to T_1, and the relaxation time of the u component, which appears in the expression[1] for $\Delta\nu_D$, depends on the Rabi frequency in accordance with $T_{2u} = \tau_c\chi^2/\overline{\Delta}^2$ (Ref. 7). It follows that a change in phase relaxation in hole-burning experiments can be observed only at $\chi > \overline{\Delta}$.

We conclude with an estimate of the correlation time τ_c. We know[8,9] that if the phase relaxation stems from a discontinuous Markov process, then the primary echo decays exponentially with a time τ_c as a function of the interval between exciting pulses I and II in the case of a slow spectral diffusion. The echo experiment of Ref. 3 yielded a value $\tau_c = 15$ μs. The stimulated echo, under conditions of a slow spectral diffusion, at a fixed interval between exciting pulses I and II, also decays exponentially with a time τ_c as a function of the interval between exciting pulses II and III (Refs. 8 and 9). A stimulated-echo experiment[10] has yielded the value $\tau_c = 20$ μs, close to the first value.[3] One might thus expect that in ruby in a strong magnetic field, at a low temperature, and with a low dopant concentration, there would indeed be a slow spectral diffusion. In turn, the fact that the nutation echo did not reveal a slowing of the phase relaxation, although the Rabi frequency reached large values, $\chi = 1.8$ mHz ($\chi > \overline{\Delta}$), can be explained on the basis that the phase relaxation time of the v component (T_{2v}) was playing a leading role here. As was shown in Ref. 7, substantially stronger fields, $\chi > \overline{\Delta}^2\tau_c$, are required for controlling the v relaxation. The reason is that the spectral diffusion disrupts the regular dephasing in the w–v plane, and only an intense field which satisfies the condition $(\sqrt{\chi^2 + \overline{\Delta}^2} - \chi)\tau_c \approx \overline{\Delta}^2\tau_c/2\chi \ll 1$ ($\chi > 70$ MHz) can prevent such a disruption.

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Translated by Dave Parsons