Free-polarization decay after pulsed saturation under anticorrelated frequency modulation

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(Submitted 23 November 1994)
Zh. Éksp. Teor. Fiz. **108**, 1907–1916 (December 1995)

This paper studies the kinetics of free-polarization decay after saturation by a pulse of laser radiation of finite length in systems with spectral diffusion. The exact solution for the shape of the free-polarization decay signal that allows for the length of the exciting radiation pulse is obtained in the model of telegraphic noise. The solution is found to be valid for arbitrary values of the spectral exchange rate and the intensity of the saturating field. Finally, it is established that the shape of the free-polarization decay signal is essentially nonexponential and is characterized by modulation related to the spectral exchange in the system.

1. INTRODUCTION

In recent years the literature has seen a number of experimental papers devoted to phenomena forming the basis of coherent nonlinear spectroscopy and saturation spectroscopy of solids: nutations,[1] free-polarization decay,[2–4] photon echo,[5] the burning out of dips,[6,7] etc. In all of this work shows the Bloch equations are found to be inapplicable for analysis of these effects. The reason is that the random modulation of the natural frequencies of the impurity ions in the solids are improperly taken into account by the Bloch equations. Modulation of the frequency of the transition generated by the laser field is caused by random reorientation of the spins of the crystal lattice. This changes the local fields and, accordingly, the frequencies of the impurity ions.

This paper is devoted to the effect of free-polarization decay after saturation in systems with random frequency modulation. To explain the experimental results of this effect,[2] a large number of theoretical papers have used random Markov processes to model the modulation of the frequency of the transition excited by the field. In Refs. 8–14 different versions of the theory of free-polarization decay were based on the assumption of rapid spectral exchange. Often this theory is called Gauss–Markovian. The diffusion model of variation of the transition frequency (correlated Markov modulation), which is based on the numerical solution of the Fokker–Planck equation, was presented in Ref. 15. The model of uncorrelated modulation of the transition frequency was discussed in Refs. 16 and 17: the same model was then used to obtain several exact solutions for the shape of the free-polarization decay signal.[18] Wodkiewicz and Eberly[19] used the "telegraphic noise" model (anticorrelated Markov frequency modulation) to describe free-polarization decay. Later, in Ref. 20, an exact solution was obtained for the free-polarization decay signal in the telegraphic noise model and the limits of applicability were established for the theory developed in Ref. 19. Note that for the free-polarization decay signal the results of the simplified telegraphic noise model[19] coincide with those of the Gauss–Markovian theory,[8–14] which is apparently the reason why the two are sometimes assumed to be the same. The similar coincidence of the results of these two models for the shape of the absorption and emission lines was reflected in Ref. 21, where the theory of rapid spectral exchange (the Gauss–Markovian theory) was referred to as the Born approximation.

The difference between the experimental work of Szabo and Muramoto[3] and that of DeVoe and Brewer[2] is that in the former the length of the exciting radiation pulse ($T=200\ \mu s$) is much shorter than population relaxation time ($T_1=4200\ \mu s$). All the above theories were developed for the free-polarization decay signal after stationary saturation, i.e., they cannot be used to explain the results of Ref. 3.

To analyze the experimental data, Szabo and Muramoto[3] used the theory of rapid spectral exchange and the telegraphic noise model in which the saturating pulse length was taken into account. However, they were unable to describe in a satisfactory manner the experimentally observed dependence of the rate of free-polarization decay on the amplitude of the field of saturating radiation. Kilin and Nizovtsev[22] once more suggested using the telegraphic noise model to describe various transient coherent phenomena: for instance, free-polarization decay after pulsed saturation. They were unable to obtain an exact solution for the shape of the free-polarization decay signal but analyzed the experimental data using approximate calculations, as a result of which they concluded that the spectral exchange in the system under investigation is slow. The existing contradiction can be resolved by comparing the results of approximate methods of calculating the free- polarization decay signal with the results of a rigorous theory free from the restrictions and simplifications adopted in Refs. 3 and 22.

The aim of the present work is to derive general expressions for the shape of the free-polarization decay signal after saturation by a strong-field pulse of arbitrary length and to analyze the experimental data of Ref. 3 and the approximate calculations done in Refs. 3 and 22 with the model of anticorrelated frequency modulation.

2. FREE-POLARIZATION DECAY AFTER SATURATION BY A STRONG-FIELD PULSE

Let us assume that the ensemble of the impurity ions of a crystal interacted with a monochromatic radiation wave $\mathcal{E}=E_0 \exp\{i\omega t\}$ and then the radiation was instantaneously switched off. After switch-off there appears the signal of decay of the free polarization that emerged during the time of action of the radiation pulse. Each impurity ion is modeled by a two-level system whose frequency $E_2(t)-E_1(t)=\omega_0+\varepsilon(t)$ is a steady-state random process, with its mean value ω_0 and equilibrium distribution $\varphi(\varepsilon)$ conserved in time (here $E_1(t)$ and $E_2(t)$ are the energy levels in the two-level system).

Since the value ω_0 is distributed across an inhomogeneous contour $\Phi(\omega_0)$ caused by the dispersion of the crystal field, the shape of the signal is determined as follows:

$$R(T+t)=\Phi_0 \mathrm{Im}\int d\Delta\omega \overline{\sigma_{12}(T+t,\Delta\omega)}, \tag{1}$$

where $\Delta\omega=\omega_0-\omega$ is the detuning of the frequency of the two-level system, ω is the frequency of the saturating field, $\Phi_0=\Phi(\omega_0=\omega)=\mathrm{const}$, and $\overline{\sigma 12(T+t,\Delta\omega)}$ is the value of the off-diagonal element of the density matrix determining the polarization at the moment t that elapsed after field switch-off. The general expression for $\overline{\sigma_{12}(T+t,\Delta\omega)}$ has the form

$$\overline{\sigma_{12}(T+t,\Delta\omega)} = \left\langle \sigma_{12}(T,\Delta\omega)\exp\left\{ i\Delta\omega t+i\int_T^{T+t}\varepsilon(t')dt'-\frac{t}{T_2}\right\}\right\rangle, \tag{2}$$

where the angle brackets stand for averaging over random realizations of the $\varepsilon(t)$ process, $\sigma_{12}(T,\Delta\omega)$ is the initial polarization generated by the saturating field, and T_2 allows for the spontaneous decay of the excited level.

Usually when finding $R(t)$ one ignores the correlations of the fluctuations of the frequency of the two-level system before and after field switch-off. This makes it possible to decouple (2). The result is

$$\overline{\sigma_{12}(t,\Delta\omega)}=\overline{\sigma_{12}(T,\Delta\omega)}K(t)\exp\{i\Delta\omega t-t/T_2\}, \tag{3}$$

where

$$K(t)=\left\langle \exp\left\{ i\int_0^t \varepsilon(t')dt'\right\}\right\rangle \tag{4}$$

is the correlation function of the frequency modulation. In Refs. 18 and 20 it was shown that if exact expressions are used for the averages $\overline{\sigma_{12}(T,\Delta\omega)}$ and $K(t)$, the results for the shape of the free-polarization decay signal are valid outside the limits of applicability of the theory of perturbations in the fluctuating frequency. The condition of applicability of a perturbation theory corresponding to the limit of rapid frequency modulation is

$$q^2=\overline{\varepsilon^2}\tau_0^2\ll 1, \tag{5}$$

where $\overline{\varepsilon^2}$ is the dispersion of the distribution in the frequency fluctuations, and τ_0^{-1} is the spectral exchange rate. Most theoretical investigations[8-14] that interpret the results of the experimental work of DeVoe and Brewer[2] have been done in this approximation.

Equations (1) and (2) are exact and determine the shape of the free-polarization decay signal in general form, but usually the averaging specified in Eq. (2) cannot be carried out. For this reason we specify the form of the random process $\varepsilon(t)$ and in Sec. 3 examine the case where the frequency of a two-level system is modulated by an anticorrelated Markov random process. This specification of the type of the random process $\varepsilon(t)$ makes it possible to carry out the averaging in Eq. (2) and derive an exact expression for the shape of the free-polarization decay signal.

3. ANTICORRELATED MARKOV FREQUENCY MODULATION

If the frequency of the two-level system interacting with the laser field is modulated by a steady-state Markov purely disconnected process, then, according to sudden modulation theory,[22] the averaging in Eq. (22) can be written in the following form:

$$\overline{\sigma_{12}(t,\Delta\omega)}=\exp\{i\Delta\omega t-t/T_2\}\int d\varepsilon K(\varepsilon,t)\sigma_{12}(\varepsilon,T), \tag{6}$$

where $K(\varepsilon,t)$ and $\sigma_{12}(\varepsilon,t)$ are the partial, or conditional, mean values whose arguments at the moment of field switch-off are both equal to ε.

It is convenient to proceed using the Laplace representation. Applying the Laplace transformation to Eq. (6) in the variables t and T, we obtain

$$\overline{\sigma_{12}(p,p_1,\Delta\omega)}=\int d\varepsilon K(\varepsilon,p_1)\sigma_{12}(\varepsilon,p), \tag{7}$$

where

$$K(\varepsilon,p_1)=\int_0^\infty dt\, K(\varepsilon,t)\exp\{-p_1 t+i\Delta\omega t-t/T_2\}. \tag{8}$$

The frequency modulation function in the event of anticorrelated spectral exchange has been studied in detail in Ref. 23. Hence for $K(\varepsilon,p_1)$ we immediately write the final expression:

$$K(\varepsilon,p_1)=\frac{p_0+1/\tau_c+i\varepsilon}{p_0^2+p_0/\tau_c+\varepsilon^2}, \tag{9}$$

where $p_0=p_1+1/T_2-i\Delta\omega$.

To obtain the general solution for the free-polarization decay signal we only need to find the value of $\sigma_{12}(\varepsilon,p)$ that determines the polarization induced by the radiation field as a function of ε. To this end we use the kinetic equation of the Markov theory of sudden modulation.[23] Bearing in mind that under anticorrelated modulation the random quantity ε can have only two values, a and $-a$, and the conditional probability density of frequency variation from ε' prior to the jump to ε after the jump is $f(\varepsilon',\varepsilon)=\delta_{\varepsilon',-\varepsilon}$, we can write the equation for the density matrix as follows:

$$\dot{X}(a)=-\left[\hat{L}_0+\frac{1}{2\tau_c}+ia\hat{L}_1\right]X(a)+\frac{1}{2\tau_c}X(-a)+\hat{\Lambda}\varphi(a),$$

$$\dot{X}(-a)=-\left[\hat{L}_0+\frac{1}{2\tau_c}-ia\hat{L}_1\right]X(-a)+\frac{1}{2\tau_c}X(a)+\hat{\Lambda}\varphi(-a), \quad (10)$$

where

$$X=\begin{pmatrix}\sigma_{12}\\ \sigma_{21}\\ n\end{pmatrix},$$

$$\hat{L}_0=\begin{pmatrix}1/T_2-i\Delta\omega & 0 & -i\chi/2\\ 0 & 1/T_2+i\Delta\omega & i\chi/2\\ -i\chi & i\chi & 1/T_1\end{pmatrix},$$

$$\hat{L}_1=\begin{pmatrix}-1&0&0\\0&1&0\\0&0&0\end{pmatrix},\quad \hat{\Lambda}=\begin{pmatrix}0\\0\\n_0/T_1\end{pmatrix},$$

$\sigma_{12}=\sigma_{21}^{*}=\rho_{12}\exp\{i\omega t\}$, $n=\rho_{22}-\rho_{11}$ is the difference in level populations, n_0 is the equilibrium difference in the level populations of the two-level system, $\chi=d_{12}E_0$ is the Rabi frequency, d_{12} is the transition's dipole matrix element, T_1 and T_2 are the longitudinal and transverse relaxation times caused by spontaneous decay, and $\varphi(\varepsilon)=0.5\delta_{\varepsilon,a}+0.5\delta_{\varepsilon,-a}$ is the static equilibrium frequency distribution.

For the discussion that follows it is convenient to use the following representation:

$$\bar{X}=\tfrac{1}{2}[X(a)+X(-a)],\quad X_A=\tfrac{1}{2}[X(a)-X(-a)]. \quad (11)$$

Then Eqs. (10) assume the form

$$\dot{\bar{X}}=-\hat{L}_0\bar{X}-ia\hat{L}_1X_A+\hat{\Lambda},$$

$$\dot{X}=-\left[\hat{L}_0+\frac{1}{\tau_c}\right]X_A-ia\hat{L}_1\bar{X}. \quad (12)$$

Applying the Laplace transformation to these equations yields

$$\overline{\bar{X}(p)}=\left[p+\hat{L}_0+a^2\hat{L}_1\frac{1}{p+\hat{L}_0+1/\tau_c}\hat{L}_1\right]^{-1}\left[X(0)+\frac{\hat{\Lambda}}{p}\right], \quad (13)$$

$$X_A(p)=a[p+\hat{L}_0+1/\tau_c]-1\hat{L}_1X(p). \quad (14)$$

Using Eqs. (13) and (14), we can now easily determine the values of the elements of the partial density matrix in (11):

$$X(\varepsilon,p)=\left(1-i\varepsilon\frac{1}{p+\hat{L}_0+1/\tau_c}\hat{L}_1\right)\times\left[p+\hat{L}_0+a^2\hat{L}_1\frac{1}{p+\hat{L}_0+1/\tau_c}\hat{L}_1\right]^{-1}\times 2\varphi(\varepsilon)\left[X(0)+\frac{\hat{\Lambda}}{p}\right]. \quad (15)$$

If we now use the explicit form of $\hat{L}_0$, $\hat{L}_1$, and $\hat{\Lambda}$, we arrive at the following expression for $\sigma_{12}(\varepsilon,p)$:

$$\sigma_{12}(\varepsilon,p)=\frac{n_0\chi\varphi(\varepsilon)}{p}\Bigg\{-\Delta\omega^3+i\Delta\omega^2\left(p+\frac{1}{T_2}\right)+\Delta\omega(\varepsilon^2-\kappa^2)+i\kappa^2\left(p+\frac{1}{T_2}\right)+i\varepsilon^2(p+t_2)+\varepsilon\left[\Delta\omega^2-i\Delta\omega\left(2p+t_2+\frac{1}{T_2}\right)-\frac{\kappa^2(p+1/T_2)}{p+t_2}-\varepsilon^2\right]\Bigg\}\frac{1}{\Delta\omega^4+B\Delta\omega^2+C}, \quad (16)$$

where

$$B=\kappa^2-2a^2+\left(p+\frac{1}{T_2}\right)^2+\frac{\chi^2(p+1/T_2)}{p+1/T_1},$$

$$C=\left[\kappa\left(p+\frac{1}{T_2}\right)+\frac{a^2(p+t_2)}{\kappa}\right]\left[\kappa\left(p+\frac{1}{T_2}\right)^2+\frac{a^2\kappa}{p+t_2}+\frac{\chi^2\kappa}{p+1/T_1}\right],\quad \kappa^2=(p+t_2)^2+\frac{\chi^2(p+t_2)}{p+t_1},\ t_{1,2}=\tau_c^{-1}+T_{1,2}^{-1}.$$

Performing the integration with respect to ε specified in (7) and combining the result with Eqs. (9) and (16), we obtain

$$\overline{\bar{\sigma}_{12}(p,p_1,\Delta\omega)}=K(p_1)\overline{\bar{\sigma}_{12}(p,\Delta\omega)}+K'(p_1)\sigma_{12}^{A}(p,\Delta\omega), \quad (17)$$

where

$$K(p_1)=\int d\varepsilon K(\varepsilon,p_1)\varphi(\varepsilon)=\frac{p+1+1/\tau_c}{p_1^2+p_1/\tau_c+a^2},$$

$$K'(p_1)=-\frac{a^2}{p_1^2+p_1/t_c+a^2},$$

$$\overline{\bar{\sigma}_{12}(p,\Delta\omega)}=\frac{n_0\chi}{2p(\Delta\omega^4+B\Delta\omega^2+C)}$$

$$\times\left[-\Delta\omega^3+i(\Delta\omega^2+\kappa^2)\left(p+\frac{1}{T_2}\right)\right.$$

$$\left.-\Delta\omega(\kappa^2-a^2)+ia^2(p+t_2)\right],$$

$$\sigma^A_{12}(p,\Delta\omega)=\frac{n_0\chi}{2p(\Delta\omega^4+B\Delta\omega^2+C)}$$

$$\times\left[-\Delta\omega^2-\Delta\omega\left(2p+t_2+\frac{1}{T_2}\right)\right.$$

$$\left.+\frac{i\kappa^2(p+1/T_2)}{p+t_2}+ia^2\right].$$

After performing the inverse Laplace transformation with respect to p_1 and integrating with respect to $\Delta\omega$, we obtain, according to the general relation (1), the following expression for the shape of the free-polarization decay signal:

$$R(p,t)=\frac{n_0\chi}{2pD}\pi\Phi_0\exp\left\{-\frac{t}{T_2}\right\}\{K(t)[R_1(p)K_1(p,t)$$

$$+R_2(p)\dot{K}_1(p,t)]+\dot{K}(t)$$

$$\times[R_3(p)K_1(p,t)R_4(p)\dot{K}_1(p,t)]\},\qquad(18)$$

where

$$K(t)=\frac{p_1\exp\{p_2t\}-p_2\exp\{p_1t\}}{p_1-p_2},$$

$$K_1(p,t)=\frac{\Delta\omega_1\exp\{i\Delta\omega_2t\}-\Delta\omega_2\exp\{i\Delta\omega_1t\}}{\Delta\omega_1-\Delta\omega_2},$$

$$p_{1,2}=-\frac{1\pm\lambda}{2\tau_c},\quad\lambda=\sqrt{1-4a^2\tau_c^2},$$

$$\Delta\omega_{1,2}=\begin{cases}\dfrac{i}{\sqrt{2}}\sqrt{B\pm\sqrt{B^2-4C}},\\ \dfrac{1}{2}[\pm\sqrt{2\sqrt{C}-B}+i\sqrt{2\sqrt{C}+B}],\end{cases}$$

$$R_1=\kappa^2\left(p+\frac{1}{T_2}\right)+a^2(p+t_2)$$

$$+\sqrt{C}\left[p+\frac{1}{T_2}-\sqrt{2\sqrt{C}+B}\right],$$

$$R_2=\kappa^2-a^2-\sqrt{C}-B+\left(p+\frac{1}{T_2}\right)\sqrt{2\sqrt{C}+B},$$

$$R_3=a^2+\frac{\kappa^2(p_1/T_2)}{p+t_2}-\sqrt{C},$$

$$R_4=2p+t_2+\frac{1}{T_2}-\sqrt{2\sqrt{C}+B},$$

$$D=\sqrt{C}\sqrt{2\sqrt{C}+B}.$$

Equation (18) is exact and determines the shape of the signal of free-polarization decay after saturation by a strong-field pulse in the presence of anticorrelated frequency modulation. It is valid for a saturating field of arbitrary intensity and an arbitrary rate of modulation of the transition frequency. Note that by performing the passage to the limit

$$R^s(t)=\lim_{p\to 0}pR(p,t),\qquad(19)$$

we arrive at an expression obtained earlier for the shape of the free-polarization decay signal in the event of stationary saturation.[20] Naturally, this result can be compared with the one obtained in this paper in the limit of $T\gg T_1$, thus making it possible to follow the transition to the stationary saturation regime.

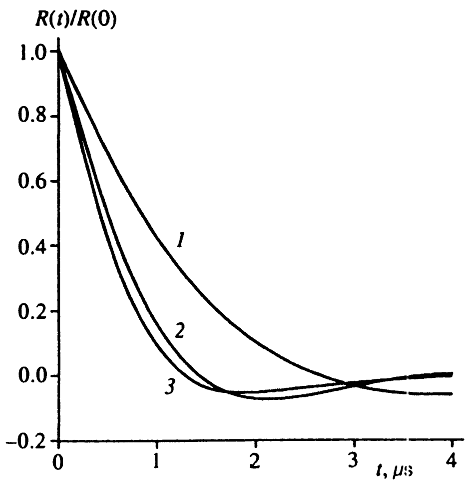

FIG. 1. The shape of the free-polarization decay signal in the case of rapid spectral exchange: $a^2\tau_c^2=0.1$, $\chi=50$ kHz, $T_1=4200\ \mu$s and $\gamma_e=15\ \mu$s. Curve *1* corresponds to $T=200\ \mu$s, curve *2* to $T=2000\ \mu$s, and curve *3* depicts the decay of free polarization after stationary saturation.

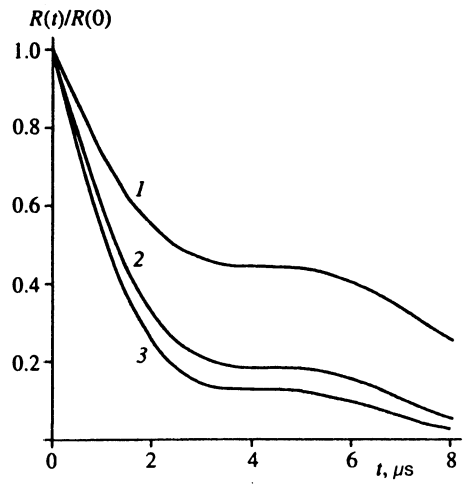

FIG. 2. The shape of the free-polarization decay signal in the case of slow spectral exchange: $a^2\tau_c^2=10$, $\chi=5$ kHz, $T_1=4200\ \mu$s, and $\gamma_e=15\ \mu$s. Curve *1* corresponds to $T=200\ \mu$s, curve *2* to $T=2000\ \mu$s, and curve *3* depicts the decay of free polarization after stationary saturation.

If in the adopted model we ignore the correlations of the frequency fluctuations in the two-level system before and after field switch-off, as was done by Szabo and Muramoto[3] and Wodkiewicz and Eberly,[19] then, according to Eqs. (3) and (4), we get

$$R(p,t)=\frac{n_0\chi}{2pD}\pi\Phi_0\exp\left\{-\frac{t}{T_2}\right\}K(t)\times[R_1(p)K_1(p,t)+R_2(p)\dot{K}_1(p,t)]. \quad (20)$$

Comparing Eqs. (20) and (18), we see that the second term, proportional to $\dot{K}(t)$, occurs in the exact solution because of the correlations of the frequency fluctuations in the two-level system before and after field switch-off. Below we demonstrate the difference between the exact solution (18) and the approximate solution (20).

Note that the expressions for the elements of the density matrix averaged over the realization of the random process in the telegraphic noise model[19,20] are identical to the results of the non-Markovian theory of perturbations in a random frequency detuning $\varepsilon(t)$ (see Refs. 11 and 14). In view of this the expression for the shape of the free-polarization decay signal in the rapid spectral exchange limit (5) coincides with Eq. (20), where we must put $K(t)=\exp\{-\overline{\varepsilon^2}\tau_c t\}$ (see Ref. 23).

4. THE SHAPE OF THE SIGNAL OF FREE-POLARIZATION DECAY AFTER SATURATION

There are two independent parameters in the final expression for the signal shape, a and τ_c. The usual approach in selecting these quantities unambiguously when interpreting the results of experiments in the field dependence of the decay rate is to employ the experimental data on the rate of photon-echo decay. In the anticorrelated frequency modulation model, the shape of the echo signal is determined by the following expression:[24]

$$V(t)=\exp\left\{-\left(\frac{1}{T_2}+\frac{1}{2\tau_c}\right)t\right\}\left[\frac{1-\lambda}{2}\times\exp\left\{-\frac{t\lambda}{2\tau_c}\right\}+\frac{1+\lambda}{2}\exp\left\{-\frac{t\lambda}{2\tau_c}\right\}-4a^2\tau_c^2\right]\frac{1}{\lambda^2}, \quad (21)$$

where $\lambda=\sqrt{1-4a^2\tau_c^2}.^{1/2}$ We see that the echo signal is exponential for rapid spectral exchange (5) and for slow spectral exchange ($a^2\tau_c^2\gg 1$). The velocity of the echo signal for these cases is

$$\gamma_e=\begin{cases}1/T_2+a^2\tau_c & \text{if } a^2\tau_c^2\ll 1,\\ 1/T_2+1/2\tau_c & \text{if } a^2\tau_c^2\gg 1.\end{cases} \quad (21)$$

Thus, if the echo signal is exponential, knowing γ_e we can determine either a^2t_c, assuming the presence of rapid spectral exchange, or $1/2\tau_c$, assuming the presence of slow spectral exchange.

To analyze the shape of the signal of free-polarization decay after saturation by a finite-length pulse, we used the algorithm for numerical inversion of the Laplace transformation suggested by Stehfest.[25] The algorithm has been tested in reconstructing Laplace images of various types and yields good results. Moreover, we checked the reliability of numerical inversion by comparing the results for the case of $T\gg T_1$, where the stationary saturation regime is realized and the exact analytical solution for the shape of the free-polarization decay signal is known.[20]

Figures 1 and 2 depict the signal shapes calculated for different lengths of the saturating pulse. For comparison the shape of the signal of free-polarization decay after stationary saturation is also given. We see that as the saturation pulse becomes longer, in the limit $T\rightarrow T_1$, the free-polarization decay signal becomes similar in shape to the signal of decay after stationary saturation; we use this fact to check the calculations. Here the picture is similar for both rapid exchange

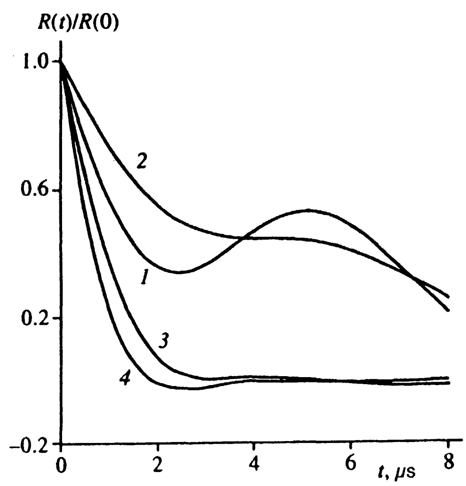

FIG. 3. The shape of the free-polarization decay signal after pulsed saturation ($T=200$ μs): $a^2\tau_c^2=10$, $T_1=4200$ μs, and $\gamma_e=15$ μs. Curve *1* corresponds to $\chi=0.5$ kHz, curve *2* to 5 kHz, curve *3* to 50 kHz, and curve *4* to 100 kHz.

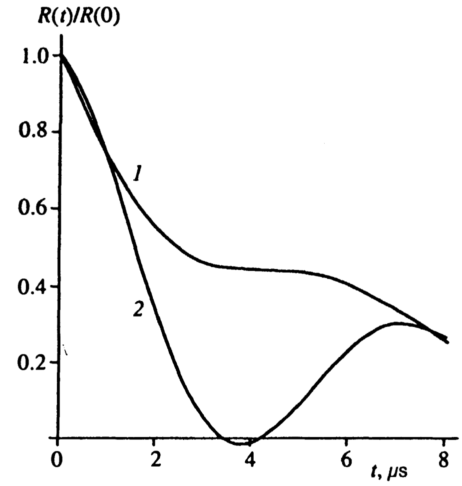

FIG. 4. The shape of the free-polarization decay signal after pulsed saturation ($T=200$ μs): $a^2\tau_c^2=10$, $T_1=4200$ μs, $\gamma_e=15$ μs, and $\chi=5$ kHz. Curve *1* corresponds to exact calculations by (18), and curve *2* to approximate calculations by (20).

(Fig. 1) and slow exchange (Fig. 2). Note that the condition $T \gg \gamma_e$ for $T_1 > T$ is not sufficient to justify the use of the expressions for the shape of the signal of free-polarization decay after stationary saturation, as Boscaino and La Bella did in Ref. 4.

Figure 3 clearly shows the effect of the field strength of the saturating radiation on the kinetics of the free-polarization decay. As the Rabi frequency increases, the decay of the induced-polarization signal increases and the oscillations related to the presence of spectral exchange in the system are suppressed.

To illustrate the differences between the exact solution (18) and the approximate solution (20), Fig. 4 depicts the free-polarization decay signals calculated by the appropriate formulas. We see that in calculating the free-polarization decay in the case of slow spectral exchange one must allow for the correlations of the frequency fluctuations in the two-level system before and after field switch-off. Note that as the Rabi frequency grows, the difference between the approximate and exact calculations diminishes because of the effect of field broadening, which suppresses the oscillations of the free-polarization decay signal.

The shapes of the free-polarization decay signals have been calculated for the conditions specified in Ref. 3. This is done in order to show that the exact solution for the free-polarization decay signal in the telegraphic noise model yields a nonexponential decay signal, in contrast to the experimental data[3] and the approximate calculations done in Ref. 22. Thus, Kilin and Nizovtsev's approximate solution,[22] which yields an exponential decay for the induced polarization, cannot serve to explain the experimental results of Ref. 3. Apparently, the simplifying assumptions used in Ref. 22 led to qualitative changes of the kinetics of the free-polarization decay signal.

5. CONCLUSION

Thus, we have arrived at an exact solution for the shape of the signal of free-polarization decay after saturation by a radiation pulse of finite length under anticorrelated spectral migration. We have found that outside the region of fast spectral exchange the free-polarization decay signal has a characteristic nonexponential shape related to spectral diffusion. We have also shown that it is important, when calculating the free-polarization decay signal, to allow for the correlations of frequency fluctuations before and after the saturating field had been switched off. Finally, our analysis makes it possible to conclude that the telegraphic noise model cannot be employed in describing spectral diffusion if we wish to explain the experimental data of Ref. 3.

The present work was done with the partial financial support of the International Center of Fundamental Physics in Moscow.

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Translated by Eugene Yankovsky