Optical phase relaxation of impurity ions in a resonant radiation field

A. R. Kessel', R. P. Shakhmuratov, and L. D. Eksin

Kazan Physico-Technical Institute, USSR Academy of Sciences
(Submitted 31 July 1987)
Zh. Eksp. Teor. Fiz. **94**, 202–215 (October 1988)

We describe the kinetics of phase relaxation of the electric polarization of a two-level center in a resonant laser radiation field. The origin of the phase relaxation is interaction with a reservoir, which we model as a shift in the resonant frequency due to a multistage random process of arbitrary strength. We obtain explicit solutions of the integral equation which describes the kinetics for seven different random processes, and find that there are conditions under which the phase relaxation is characterized by two times T_{2u} and T_{2v} which depend in different ways on the intensity of the laser radiation. On the basis of the microscopic theory developed here, we give a rigorous proof of Redfield's hypothesis, which postulates the existence of an effect due to the resonance field which lengthens the time T_{2u} without noticeably lengthening the time T_{2v}; this effect is appreciable when the amplitude of the resonance field (in energy units) is comparable to the local field which causes the relaxation of the impurity center. This type of dependence of the phase relaxation of an impurity on the laser radiation power was observed earlier in experiments on free-induction decay of Pr^{3+}: LaF_3.

1. INTRODUCTION

Recently there has been a new growth of interest in the problem of relaxation of atoms in an intense laser radiation field.[1–11] This growth of interest is driven by an experiment[1] in which anomalous behavior of the optical induction was observed in crystals of LaF_3 doped with Pr^{3+} impurity ions, a material known for its long-duration optical phase memory. The anomaly appears in the dependence of the relaxation time T_2 for the polarization on the intensity of the long laser pulse which prepares the Pr^{3+} ions in a stationary excited state.

The theoretical descriptions of this effect proposed by a number of authors[2–8] are based on the assumption that intense laser radiation averages out the action of the random fields caused by the environment which are responsible for phase relaxation of the impurity ion. These fields are generated by the magnetic moments of ^{19}F nuclei; the resulting Pr–F dipole coupling causes a shift in the resonance frequency ω_0 for the $^3H_4$–$^1D_2$ transition of the Pr^{3+} ion ($\omega = \omega_0 + \Delta\omega$). A random time dependence of $\Delta\omega$ arises due to the mutual reorientations of the magnetic moments of ^{19}F nuclei resulting from their dipole-dipole coupling.

The theories proposed in Refs. 2–8 can be classified as follows, depending on the relations used by their authors between the fundamental characteristics of the random process—i.e., its dispersion $\langle\Delta\omega^2\rangle = \delta^2$ and correlation time τ_c. Those of Refs. 2–5 belong to the first group, in which it is assumed that the random process is weak or rapidly-varying ($\delta\tau_c \ll 1$); on the basis of this, these authors use a differential kinetic equation. In a weak laser field this is the Bloch equation, which describes only exponential relaxation. In the opposite limiting case (strong or slowly-varying random processes, i.e., $\delta\tau_c \gg 1$), nonexponential relaxation can occur and the evolution of the resonant levels is not described by the Bloch equation.[3]

In practice, most crystals exhibit the intermediate case ($\delta\tau_c \sim 1$), and another approach is required in order to describe the relaxation. One method which puts no limits on the magnitude of the parameter $\delta\tau_c$ was developed by Burshtein[12–14] for a particular form of random process. A special form of this process is that of two-step telegraph noise, which was already used in Ref. 7 to describe phase relaxation of Pr^{3+}. The more general case of a multistep random process was studied in Ref. 8. In both Refs. 7, 8 an exponential (or biexponential) law is obtained for the polarization decay independent of the magnitude of the parameter $\delta\tau_c$; this result is questionable, at least in the region $\delta\tau_c \gg 1$. In the first case, exponential decay is a consequence of the statistical "poverty" of the telegraph noise model, which possesses only two states, whereas in the second case it is due to neglect of contributions to the signal from "holes" in the wings which are burned into the inhomogeneously broadened spectrum.

In this paper, we develop a theory of phase relaxation based on Burshtein's method[12] for arbitrary values of the parameter $\delta\tau_c$. We analyze a large set of possible multistep random processes which give rise to the relaxation. We show that for certain ranges of the parameter $\delta\tau_c$ the relaxation acquires a nonexponential character. We find expressions for the boundaries of these ranges, which depend on the form of the random process. For the example of a resonant packet we investigate the effect of laser radiation on the boundaries of these regions and on the relaxation rates.

2. DERIVATION OF THE KINETIC EQUATIONS AND THEIR GENERAL SOLUTION

We will describe the kinetics of dipole dephasing using the method developed in Ref. 12. This method involves a model random process whose origin is the action of a potential $\widehat{V}(t)$, which in turn depends on a certain random parameter $\alpha(t)$. The time variation of α consists of discontinuous and uncorrelated jumps which are separated by intervals of time Δt over which it remains constant. Assume that the length of each such interval is determined by the probability $dW = e^{-t/\tau_c} dt/\tau_c$, where τ_c is the mean time between jumps, and that there are no correlations between values of the parameter α in neighboring intervals. Then the probability $dW(\alpha)$ of finding a given value of α in any cross-section

 0038-5646/88/102071-08$04.00

of the process does not depend on time. The equation for the density matrix $\rho_{ij}(t)$ averaged over all possible realizations of the process has the form[12]

$$\rho_{im}(t)=e^{-t/\tau_c}\,\mathrm{Sp}[\hat{R}^{im}(t,0)\hat{\rho}(0)]+\frac{1}{\tau_c}\int_0^t dt_1 e^{-(t-t_1)/\tau_c}\,\mathrm{Sp}[\hat{R}^{im}(t,t_1)\hat{\rho}(t_1)], \qquad (1)$$

$$R_{lk}{}^{im}=\int_\alpha S_{ik}(\alpha,t,t_1)S_{lm}{}^{-1}(\alpha,t,t_1)\,dW(\alpha),$$

where $\hat{S}$ is the evolution operator which satisfies the equation $i\hbar d\hat{s}/dt=\hat{\mathscr{H}}(\alpha)\hat{s}$, $\hat{\mathscr{H}}(\alpha)$ is the system Hamiltonian.

Let us investigate a two-level atom which undergoes phase relaxation under the influence of $\hat{V}(t)$, where the latter gives rise to a random discontinuous change in the resonant frequency, i.e., $\omega_0\to\omega_0+\Delta\omega(t)$. We will be interested in the behavior of this atom in a coherent radiation field whose frequency Ω coincides with the resonant frequency ω_0. The Hamiltonian of such an atom has the form

$$\hat{\mathscr{H}}=\hat{\mathscr{H}}_0+\hat{\mathscr{H}}_t+\hat{V}(t), \qquad (2)$$

$$\hat{\mathscr{H}}_0+\hat{V}(t)=\frac{\hbar}{2}[\omega_0+\Delta\omega(t)](\hat{P}_{22}-\hat{P}_{11}),$$

$$\hat{\mathscr{H}}_t=-\frac{\hbar\chi}{2}(\hat{P}_{12}e^{i\omega_0 t}+\hat{P}_{21}e^{-i\omega_0 t}),$$

where $\hat{P}_{mn}$ are projection operators with certain prescribed properties: $\hat{P}_{mn}\Psi_k=\delta_{nk}\Psi_k$; Ψ_1,Ψ_2 are unperturbed wave functions for the atomic states with energies $E_1<E_2$ (where $\hbar\omega_0=E_2-E_1$); $\chi=\langle\Psi_1|\mathbf{dE}|\Psi_2\rangle$ is the Rabi frequency; $\mathbf{E}$ is the amplitude of the resonance radiation field; and $\mathbf{d}$ is the dipole moment. For such a system, Eq. (1) takes the form

$$x(\tau)=x(0)k(\tau)+\int_0^\tau d\tau_1 x(\tau_1)k(\tau-\tau_1), \qquad (3a)$$

$$y(\tau)=-y(0)\frac{1}{\varepsilon^2}\ddot{n}(\tau)-z(0)\frac{1}{\varepsilon}\dot{n}(\tau)+\int_0^\tau d\tau_1\left[-y(\tau_1)\frac{1}{\varepsilon^2}\ddot{n}(\tau-\tau_1)-z(\tau_1)\frac{1}{\varepsilon}\dot{n}(\tau-\tau_1)\right], \qquad (3b)$$

$$z(\tau)=z(0)n(\tau)+y(0)\frac{1}{\varepsilon}\dot{n}(\tau)+\int_0^\tau d\tau_1\left[z(\tau_1)n(\tau-\tau_1)+y(\tau_1)\frac{1}{\varepsilon}\dot{n}(\tau-\tau_1)\right], \qquad (3c)$$

where $\tau=t/\tau_c$, $x(\tau)=u(\tau)e^\tau$, $y(\tau)=v(\tau)e^\tau$, $z(\tau)=w(\tau)e^\tau$, $w=\rho_{22}-\rho_{11}$, $\frac{1}{2}(u+iv)e^{i\Omega t}=\rho_{12}$, $\varepsilon=\chi\tau_c$. The functions $k(\tau)$ and $n(\tau)$ are calculated by averaging over all possible realizations of the dimensionless random parameter $\alpha=\Delta\omega\tau_c$:

$$k(\tau)=\int_\alpha\left\{\frac{\alpha^2}{\alpha^2+\varepsilon^2}+\frac{\varepsilon^2}{\alpha^2+\varepsilon^2}\cos[\tau(\varepsilon^2+\alpha^2)^{1/2}]\right\}dW(\alpha),$$
$$(4)$$
$$n(\tau)=\int_\alpha\left\{\frac{\varepsilon^2}{\alpha^2+\varepsilon^2}+\frac{\alpha^2}{\alpha^2+\varepsilon^2}\cos[\tau(\varepsilon^2+\alpha^2)^{1/2}]\right\}dW(\alpha).$$

We can use the Laplace transform

$$F(p)=\int_0^\infty e^{-p\tau}f(\tau)\,d\tau$$

to reduce the system of integral equations (3) to a system of algebraic equations whose solution has the form

$$X(p)=x(0)K(p)[1-K(p)]^{-1},$$
$$Y(p)=[(p-1)y(0)+\varepsilon z(0)]p^2K(p)M^{-1}(p), \qquad (5)$$
$$Z(p)=\{[p^2+\varepsilon^2-p(p+\varepsilon^2)K(p)]z(0)-\varepsilon p^2K(p)y(0)\}M^{-1}(p),$$
$$M(p)=(p^2+\varepsilon^2)(p-1)+p[\varepsilon^2-p(p-1)]K(p),$$

where $X(p)$, $Y(p)$, and $Z(p)$ are transforms of the functions $x(\tau)$, $y(\tau)$, and $z(\tau)$, and

$$K(p)=\int_\alpha\frac{p^2+\varepsilon^2}{p(p^2+\alpha^2+\varepsilon^2)}\,dW(\alpha). \qquad (6)$$

Applying the inversion formula

$$f(\tau)=\frac{1}{2\pi i}\int_{a-i\infty}^{a+i\infty}e^{p\tau}F(p)\,dp \qquad (7)$$

to Eq. (5), we can obtain an exact solution to the integral equations (3) which describe the kinetics of the phase relaxation of the two-level system. In the general case it can take the following form:

$$f(\tau)=\sum_i C_i e^{p_i\tau}+Q(\tau), \qquad (8)$$

where p_i are poles of the transform $F(p)$, $C_i=\mathrm{res}\,F(p_i)$ are the residues at these poles. The function $Q(\tau)$ is made up of contributions from other singularities and has a more complex time dependence. In the experimentally observed relaxation time dependences of the quantities $u(t)$, $v(t)$, and $w(t)$, the first term in Eq. (8) contributes a simple exponential dependence (characteristic of the kinetic limit[15]) with the time constants $T_{u,v,w}=\tau_c/(1-p_i)|_{\chi=0}$. The second term $Q(\tau)$ cannot be obtained from kinetic equations of the Bloch-equation type. It is a consequence of our going beyond the framework of perturbation theory in the interaction $\hat{V}(\alpha)$, and we cannot introduce a relaxation time to represent it in the same way as for an exponential term. However, like the exponential, the function $Q(\tau)$ has a characteristic time scale over which it varies appreciably. In order to specify this scale quantitatively for all functions $f(\tau)$, we define an integrated relaxation time through the maximum of the homogeneous absorption line $g_0(\Delta)$[16–18]:

$$\mathcal{T}=\pi g_0(0)=\int_0^\infty f(t)e^{-t/\tau_c}\,dt=F(1)\tau_c. \qquad (9)$$

In this expression the function $f(\tau)$ is multiplied by an exponential because the solution (7) differs from the desired functions $u(t)$, $v(t)$, and $w(t)$ by the factor e^τ.

In the absence of an AC field ($\chi=0$) the form of the transform (5) considerably simplifies:

$$Z(p)=\frac{z(0)}{p-1},\quad \frac{X(p)}{x(0)}=\frac{Y(p)}{y(0)}=\frac{K(p)}{1-K(p)}=L(p). \qquad (10)$$

From this it follows that $w(t) = \text{const}$ and $u(t)/u(0) = v(t)/v(0)$, i.e., the difference in level populations does not change while the kinetics of the components u and v are identical. Therefore, for $\chi = 0$ the perturbation $\hat{V}(t)$ is adiabatic. Turning on the AC field considerably changes the situation.

3. ANALYSIS OF THE KINETICS

In the formalism developed here the properties of the physical system are derived from those of the density distribution $dW(\alpha)$ for the random variable α. The assignment of $dW(\alpha)$ has a heuristic character, in that our success in describing the kinetics will depend to a considerable degree on how well $dW(\alpha)$ reflects the system's physical properties. In order to clarify the reason for the appearance of nonexponential relaxation, we will analyze several types of distributions. First of all, we will begin with (1) uniform and (2) normal distributions. According to the Central-Limit Theorem, when the number of sources of the local fields, i.e., the spins which participate in shifting an ion's resonant frequency, is large, the fields will be normally distributed. The uniform distribution, which is one possible approximation to a normal distribution, allows us to obtain easily results we can inspect. In order to clarify to what extent the character of the kinetics is affected by the rate at which the wings of the density distribution fall off, we will investigate (3) a Laplace distribution, (4) a Cauchy distribution, and (5) a third distribution which decreases much more slowly in the wings than (4) and (5). Finally, we will investigate two other distributions which provide reasonable approximations to the scatter of local fields in a physical system with a rather small number of particles.

1. Uniform (rectangular) distribution:

$$dW_1(\alpha)=\begin{cases} d\alpha/2\gamma, & |\alpha|<\gamma, \\ 0, & |\alpha|>\gamma. \end{cases} \tag{11}$$

This distribution leads to the following expression for the functions:

$$K_1(p)=\frac{\Psi(p)}{p}\operatorname{arcctg}\Psi(p),\quad \Psi(p)=\frac{1}{\gamma}(\varepsilon^2+p^2)^{1/2},$$

where $\gamma = \beta\tau_c$ and $\beta^2/3 = \langle\Delta\omega^2\rangle$ is the dispersion of the random variable $\Delta\omega$. In the absence of an AC field ($\chi = 0$), the function $L(p)$ has a single pole $p_1 = \gamma \operatorname{ctg} \gamma$ in the region of parameter values $\gamma \leqslant \pi/2$. For $\gamma > \pi/2$ the pole disappears. Thus, the value $\gamma = \pi/2 \equiv \gamma_{cr}$ is critical for the existence of exponential relaxation.

For the case of a uniform distribution we can find an exact solution for the relaxation function $f_{u,v}(t)$:

$$f_{u,v}(t)=\frac{u(t)}{u(0)}=\frac{v(t)}{v(0)}$$
$$=e^{-t/\tau_c}\left\{\theta\left(\frac{\pi}{2}-\gamma\right)\frac{\gamma^2}{\sin^2\gamma}e^{(t/\tau_c)\gamma\operatorname{ctg}\gamma}+Q(t)\right\},$$
$$\theta(x)=\begin{cases}1, & x\geqslant 0\\ 0, & x<0\end{cases},\quad Q(t)=Q_+(t)+Q_-(t). \tag{12}$$

The function $Q(t)$ results from the logarithmic branch point $p = \pm i\gamma$ of the transform $K_1(p)$; its components have the forms:

$$Q_\pm(t)=\frac{\gamma}{\pi}\int_0^1 dx\left\{\left[\frac{\pi}{2}\left(\gamma\pm\frac{\pi}{2}\right)\pm m^2\right]\right.$$
$$\left.\cdot\cos\left(\frac{\gamma x t}{\tau_c}\right)\pm\gamma m\sin\left(\frac{\gamma x t}{\tau_c}\right)\right\}$$
$$\cdot\left[\left(\gamma\pm\frac{\pi}{2}\right)^2+m^2\right]^{-1},\quad m=\frac{1}{2}\ln\left(\frac{1-x}{1+x}\right).$$

For $\gamma\leqslant\pi/2$ the result (12) is well approximated by the expression

$$f_{u,v}(t)=\left(\frac{\gamma}{\sin\gamma}\right)^2e^{-t/T_2}+\left(1-\frac{\gamma^2}{\sin^2\gamma}\right)\frac{\sin\beta t}{\beta t}e^{-t/\tau_c}$$
$$+R(t)e^{-t/\tau_c}. \tag{13}$$

The function $R(t)$ equals zero for $t = 0$ and $t = \infty$. For all remaining values of t its contribution to $f_{u,v}(t)$ is less than a few percent. The purely exponential decay is characterized by a time $T_2 = \tau_c/(1-\gamma \operatorname{ctg}\gamma)$. In the small-$\gamma$ limit this time coincides with the well-known value $T_2 = (\langle\Delta\omega^2\rangle\tau_c)^{-1}$ given by perturbation theory for the case of a weak random process. For $\gamma \gg 1$ Eq. (12) is approximated by the function

$$f_{u,v}(t)=\frac{\sin\beta t}{\beta t}e^{-t/\tau_c}. \tag{14}$$

Turning on even a weak ($\chi\tau_c \ll 1$) AC field leads to the appearance of poles in the supercritical region of parameter values of γ ($\gamma > \pi/2$); this gives rise to exponential relaxation with a time $T_2 \approx (1+\gamma_{cr}\chi/\beta)\tau_c$. However, the contribution from this exponential relaxation (i.e., the weight of amplitude $C_i \approx \gamma_{cr}\chi/\beta$ in Eq. (8)) to the relaxation function is small compared to the contribution from $Q(t)$. Nevertheless, the time T_2 for this process is found to be far longer than the characteristic time scale for nonexponential relaxation ($\sim\beta^{-1}$). Therefore at large times a weak field changes the character of the kinetics from nonexponential to exponential.

As χ increases, the exponential term in the relaxation function increases. In a strong field ($\chi\tau_c \gg 1,\gamma$) the relaxation becomes predominantly exponential, and the relaxation time grows appreciably; the character of this increase depends on the strength of the random process. For $\gamma \ll 1$ the slowing-down of the relaxation proceeds as long as $\chi\tau_c > 1$; then

$$u(t)=u(0)e^{-t/T_{2u}},\quad v(t)=v(0)e^{-t/T_{2v}}\cos\chi t,$$
$$T_{2u}=\tau_c\frac{1+(\chi\tau_c)^2}{\langle\Delta\omega^2\rangle\tau_c^2},\quad T_{2v}\approx 2T_{2u}. \tag{15}$$

For the case $\gamma \gg 1$, by the time that $\chi \sim (\langle\Delta\omega^2\rangle)^{1/2}$, the time T_{2u} has increased from a value on the order of $(\langle\Delta\omega^2\rangle)^{-1/2}$ to several times τ_c. However, the variation in the relaxation rate of the v-component is not so significant in this case. This difference in the u and v relaxation rates is preserved up to very large values of $\chi \sim \gamma(\langle\Delta\omega^2\rangle)^{1/2}$. The times T_{2u} for the range of values $\chi > (\langle\Delta\omega^2\rangle)^{1/2}$ and T_{2v} for $\chi > \gamma(\langle\Delta\omega^2\rangle)^{1/2}$ depend on the parameters in the following fashion:

$$T_{2u}=\tau_c\chi^2/\langle\Delta\omega^2\rangle,\quad T_{2v}=5\tau_c\chi^2/(\langle\Delta\omega^2\rangle)^2\tau_c^2. \tag{16}$$

For all values of γ and χ the integrated relaxation time (9) of the u-component can be represented in a compact form:

$$T_{2u}=\tau_c\left\{\frac{\gamma}{(\varepsilon^2+1)^{1/2}\operatorname{arcctg}[(\varepsilon^2+1)^{1/2}/\gamma]}-1\right\}^{-1}. \quad (17)$$

Finally, let us note that in a strong field there appear traces of the dynamics in the behavior of the u and v components—oscillations at a frequency $\chi^*=\chi(1+\langle\Delta\omega^2\rangle/2\chi^2)$, which are somewhat shifted from the Rabi frequency by the relaxation processes.

2. Normal (Gaussian) distribution:

$$dW_2(\alpha)=\frac{1}{\gamma(2\pi)^{1/2}}e^{-\alpha^2/2\gamma^2}d\alpha,$$
$$K_2(p)=\left(\frac{\pi}{2}\right)^{1/2}\frac{\Psi(p)}{p}e^{\Psi^2(p)/2}\operatorname{erfc}(\Psi(p)/2^{1/2}), \quad (18)$$

where $\operatorname{erfc}(x)=1-\operatorname{erf}(x)$; $\operatorname{erf}(x)$ is the probability integral[19] and $\gamma=\beta\tau_c=(\langle\Delta\omega^2\rangle)^{1/2}\tau_c$.

Let us investigate the kinetics of phase relaxation in the absence of an external field. For the distribution (18) the dependence of the poles of the function $L(p)$ (10) on the parameter γ is difficult to represent in explicit form. However, when $\gamma\ll 1$ the poles can be found in the form of an expansion in powers of γ: $p_1\approx 1-\gamma^2+\dots$. In this case the kinetics of polarization relaxation has a purely exponential character with $T_2=(\langle\Delta\omega^2\rangle\tau_c)^{-1}$. We also can show that the critical value of the parameter γ is $\gamma_{\rm cr}=(\pi/2)^{1/2}$, so that for $\gamma>\gamma_{\rm cr}$ the exponential part of the solution (8) disappears, and the kinetics of polarization becomes fully nonexponential. As an example of such behavior we will use the simple asymptotic form of the function $Q(t)$, which is valid in the case of a strong random process ($\gamma\gg 1$):

$$f_{u,v}(t)=e^{-t/\tau_c}\left\{e^{-\beta^2t^2/2}+\frac{\pi^{1/2}}{\gamma}\operatorname{erf}\left(\frac{\beta t}{2}\right)e^{-\beta^2t^2/4}+\dots\right\}. \quad (19)$$

Analysis shows that in the subcritical region ($\gamma\leqslant\gamma_{\rm cr}$) the exponential relaxation time decreases from the value $\tau_c/\gamma^2\gg\tau_c$ for $\gamma\ll 1$ to τ_c as $\gamma\to\gamma_{\rm cr}$ as the strength of the random process increases.

The integrated relaxation time for all values of the parameter γ equals

$$T_2^{-1}=\tau_c^{-1}\left\{\left[\left(\frac{\pi}{2}\right)^{1/2}\frac{1}{\gamma}e^{1/2\gamma^2}\operatorname{erfc}(1/2^{1/2}\gamma)\right]^{-1}-1\right\}. \quad (20)$$

Turning on the AC field changes the relaxation function in the same way as for the case of a uniform distribution (see Subsec. 1). A small quantitative difference is observed in the region of parameter values $\gamma\gg 1, \chi>\gamma(\langle\Delta\omega^2\rangle)^{1/2}$, where the coefficient 5 in Eq. (16) for T_{2v} is changed to a 2. Another difference is observed in the total relaxation time for small values of the AC field amplitude ($\chi\tau_c<1$):

$$T_{2u}=\tau_c\left\{\frac{\gamma\exp[-(\varepsilon^2+1)/2\gamma^2]}{(\pi/2)^{1/2}(\varepsilon^2+1)^{1/2}\operatorname{erfc}((\varepsilon^2+1)^{1/2}/2^{1/2}\gamma)}-1\right\}^{-1}. \quad (21)$$

In Ref. 8, in which the distribution (18) was used, it was shown that when β is the largest parameter ($\beta\gg\chi,\tau_c^{-1}$) the relaxation kinetics are exponential with the characteristic time τ_c. A more detailed analysis shows that in this case the relaxation function (8) is essentially nonexponential and is determined by the function $Q(t)$, which falls off rapidly over short times on the order of β^{-1}. Therefore at large times the exponential part of the relaxation function takes over, with $C_i\approx\gamma_{\rm cr}\chi/\beta$ and $T_2\approx(1+\gamma_{\rm cr}\chi/\beta)\tau_c$. These kinetics are characteristic of a homogeneous line with a narrow Lorentz center [with a width $(2\pi T_2)^{-1}\approx(2\pi\tau_c)^{-1}$] and broad Gaussian wings. In Ref. 8 only the Lorentzian part of the spectrum was taken into account, which is caused by processes which mediate the flow of the statistical ensemble of atoms out of a state with a given shift $\Delta\omega_i$. The terms of the kinetic equation which describe flow into this state (i) from all remaining states are excluded, and it is just these terms which give rise to the Gaussian line broadening. Therefore the result of Ref. 8 is valid only for large times.

3. Laplace distribution:

$$dW_3(\alpha)=\frac{1}{2\gamma}\exp\left(-\frac{|\alpha|}{\gamma}\right)d\alpha, \quad (22)$$
$$K_3(p)=\frac{\Psi(p)}{p}[\operatorname{ci}\Psi(p)\sin\Psi(p)-\operatorname{si}\Psi(p)\cos\Psi(p)],$$

where $\operatorname{ci}(\Psi(p))$ and $\operatorname{si}(\Psi(p))$ are the cosine and sine integrals,[19] and $\gamma=\beta\tau_c$, $2\beta^2=\langle\Delta\omega^2\rangle$.

For small γ the polarization kinetics exhibit many of the features investigated previously, qualitatively and in many respects quantitatively: there exists a $\gamma_{\rm cr}=\pi/2$, for $\gamma\ll 1$ the exponential relaxation time $T_2=(\langle\Delta\omega^2\rangle\tau_c)^{-1}$, etc.

Differences are observed for large γ. For example, for $\gamma\gg 1$ a different asymptotic form is valid for the relaxation function:

$$f_{u,v}(t)=Q(t)\approx e^{-t/\tau_c}\left(1+\frac{1}{2}\langle\Delta\omega^2\rangle t^2\right)^{-1}. \quad (23)$$

The effect of an AC field on the kinetics is fully analogous (qualitatively and quantitatively) to that investigated in Subsecs. 1 and 2.

The integrated relaxation time for this distribution has the form

$$T_{2u}^{-1}=\tau_c^{-1}[K_3^{-1}(1)-1]. \quad (24)$$

4. Cauchy distribution:

$$dW_4(\alpha)=\frac{1}{1+(\alpha/\gamma)^2}\frac{d\alpha}{\gamma},\quad \gamma=\beta\tau_c,$$
$$K_4(p)=\frac{1}{p}\frac{\Psi(p)}{1+\Psi(p)}. \quad (25)$$

In the absence of an external field we can find an exact solution to this problem, which turns out to be exponential relaxation with a time $T_2=\beta^{-1}$ for all values of the parameter γ.

Turning on the field complicates the problem. The image $K_4(p)$ acquires a square-root branch point $p=\pm i\varepsilon$, thanks to which the term $Q(t)$ in the relaxation function (8) becomes different from zero. In the limiting cases of weak ($\chi\tau_c\ll 1$) and strong ($\chi\tau_c\gg 1,\gamma$) AC fields, $Q(t)$ gives rise to small corrections to (8) of order $(\chi\tau_c)^2$ and (β/χ), respectively; however, in intermediate fields this correction can turn out to be substantial. For example, for $\chi\sim\beta$ (for $\gamma\gg1$) $Q(t)$ amounts to 50% of the signal, appearing as an oscillating function which decays with time.

The AC field also changes the exponential relaxation. A weak field produces a small correction:

$$T_2=\frac{1}{\beta}\left[1+\frac{(\chi\tau_c)^2}{2(1-\gamma)^2}\right]\quad(\gamma\neq1),\tag{26}$$

while a strong one considerably slows it down:

$$\begin{aligned}&T_{2u}=\tau_c(\chi/\beta)\quad(\chi\tau_c>1,\ \gamma),\\&T_{2v}=2\tau_c(\chi\tau_c)^{1/2}/\gamma\quad(\chi\tau_c>1,\ \gamma^2).\end{aligned}\tag{27}$$

For strong random processes ($\gamma\gg1$) we observe a slowing-down of the exponential relaxation of the u-component even for $\chi\sim\beta=T_2^{-1}$; it is found that the time T_{2u} is lengthened by a factor of 2γ, becoming equal to $2\tau_c$. A considerable lengthening of T_{2v} also takes place for stronger fields ($\chi\tau_c>\gamma^2$).

The integrated relaxation time reinforces the conclusions about the kinetics of the u-component:

$$\tilde{T}_{2u}=\frac{1}{\beta}(1+(\chi\tau_c)^2)^{1/2}.\tag{28}$$

5. We now investigate the distribution

$$dW_5(\alpha)=\frac{2^{1/2}}{\pi}\frac{1}{1+(\alpha/\gamma)^4}\frac{d\alpha}{\gamma},\quad\gamma=\beta\tau_c,\quad\beta^2=\langle\Delta\omega^2\rangle,$$

which is similar to a Cauchy distribution but which falls off more rapidly in its wings. In this case we have

$$K_5(p)=\frac{\Psi(p)}{p}\frac{\Psi(p)+2^{1/2}}{1+2^{1/2}\Psi(p)+\Psi^2(p)}.\tag{29}$$

In the absence of an AC field the problem has an exact solution whose form depends critically on the strength γ of the random process. For $\gamma<\gamma_{\rm cr}=1+2^{-1/2}$ the relaxation function evolves according to a biexponential law:

$$\begin{aligned}f_{u,v}(t)=\frac{1}{2D^{1/2}}\{&(A+D^{1/2})\exp[-(A-D^{1/2})t/2\tau_c]\\&-(A-D^{1/2})\exp[-(A+D^{1/2})t/2\tau_c]\},\end{aligned}\tag{30}$$

where $D=|1+2^{3/2}\gamma-2\gamma^2|$, $A=1+2^{1/2}\gamma$. At the critical point ($\gamma=\gamma_{\rm cr}$) there arises a power-law deviation from the exponential:

$$f_{u,v}(t)=(1+\gamma_{\rm cr}t/\tau_c)\exp(-\gamma_{\rm cr}t/\tau_c),\tag{31}$$

while in the supercritical region ($\gamma>\gamma_{\rm cr}$) there is an exponential decay accompanied by oscillations:

$$f_{u,v}(t)=\left[\cos\frac{D^{1/2}t}{2\tau_c}+\frac{A}{D^{1/2}}\sin\frac{D^{1/2}t}{2\tau_c}\right]e^{-At/2\tau_c}.\tag{32}$$

As in Secs. 1–3, turning on a strong AC field leads to a purely exponential relaxation with a time $T_{2u}=\tau_c\chi^2/\langle\Delta\omega^2\rangle$ for any γ so long as $\chi\tau_c\gg1,\gamma$. The relation $T_{2v}=2T_{2u}$ obtains only for a weak random process ($\gamma\ll1$) and a limited range of field amplitudes ($1\ll\chi\tau_c\ll2/\gamma^2$). If $\chi\tau_c>2/\gamma^2$, then the dependence of the time T_{2v} on the parameters changes:

$$T_{2v}=2^{3/2}\tau_c\left(\frac{\chi}{\langle\Delta\omega^2\rangle\tau_c}\right)^{1/2}.\tag{33}$$

In the case of a strong stepwise process ($\gamma\gg1$) the exponential relaxation of the v-component is also realized in a stronger field ($\chi>\gamma(\langle\Delta\omega^2\rangle)^{1/2}$ with the time T_{2v} given in (33).

For all parameters of the system the integrated relaxation time in this model has the form

$$\tilde{T}_{2u}=\tau_c\left\{\frac{(\chi\tau_c)^2+1}{\gamma^2}+\left[2\frac{(\chi\tau_c)^2+1}{\gamma^2}\right]^{1/2}\right\}.\tag{34}$$

6. Telegraph noise

$$\begin{aligned}dW_6(\alpha)&=\frac{d\alpha}{2}[\delta(\alpha+\gamma)+\delta(\alpha-\gamma)],\quad\gamma^2=\langle\Delta\omega^2\rangle\tau_c^2,\\K_6(p)&=\frac{\Psi^2}{p(\Psi^2+1)}.\end{aligned}\tag{35}$$

In the absence of an AC field the system relaxation proceeds according to the same laws (30)–(32) which were obtained in the previous case. For the telegraph noise model, the parameters which enter into Eqs. (30)–(32) are: $\gamma_{\rm cr}=\frac{1}{2}$, $A=1$, and $D=|1-4\gamma^2|$.

In the case of a strong stepwise process and in the region where the field is comparable to the local fields ($\chi\sim(\langle\Delta\omega^2\rangle)^{1/2}$), the relaxation function has the form

$$\begin{aligned}f_u(t)&\approx\frac{1}{2}e^{-t/2\tau_c}+\frac{1}{2}\cos[(2\langle\Delta\omega^2\rangle)^{1/2}t]e^{-3t/4\tau_c},\\f_v(t)&\approx\cos[(2\langle\Delta\omega^2\rangle)^{1/2}t]e^{-t/4\tau_c},\end{aligned}\tag{36}$$

where we assume that $\chi=(\langle\Delta\omega^2\rangle)^{1/2}$. Hence, the AC field under these conditions, while changing the character of the relaxation, does not cause much change in its slowing-down. In a strong field the essential contribution to the relaxation function is given by one exponential; the corresponding times $T_{2u}=\tau_c\chi^2/\langle\Delta\omega^2\rangle$ and $T_{2v}=2T_{2u}$ are considerably longer than the relaxation times in weak fields.

In this model the integrated relaxation time coincides in form with that predicted by perturbation theory:

$$T_{2u}=\tau_c\frac{1+(\chi\tau_c)^2}{\langle\Delta\omega^2\rangle\tau_c^2}.\tag{37}$$

7. The distribution

$$dW_7(\alpha)=\frac{\sin^2(\alpha/\gamma)}{(\alpha/\gamma)^2}\frac{d\alpha}{\pi\gamma},\quad\gamma=\beta\tau_c\tag{38}$$

leads to the following expression for the function (6):

$$K_7(p)=\frac{1}{p}\left[1-\frac{1}{2\Psi(p)}(1-e^{-2\Psi(p)})\right].\tag{39}$$

Here, in contrast to all the previous cases, in the absence of a field not only the magnitude but also the very form of the relaxation function changes as time passes; this is reflected in the solution:

$$f(t)=\sum_{n=0}^{\infty} f_n(t-t_n)\theta(t-t_n), \qquad (40)$$

where $\theta(t)$ is the Heaviside function, $t_n = 2n\beta^{-1}$, $n = 0,1,2,...,$ and $f_n(t)$ are the original functions $F_n(p) = [(1+a/b)b^{-n} - \delta_{n,0}]$, $a = p - \gamma/2$, $b = p^2 - 2p + \gamma/2$. While staying well-defined on the time interval $\Delta t = t_n - t_{n-1}$, the form of the relaxation function changes as we go to the next interval Δt_{n+1}.

The function $f_0(t)$ repeats the behavior of the relaxation functions in Subsecs. 5 and 6 with $\gamma_{cr} = \frac{1}{2}$, $A = 1 - \gamma$, and $D = |1 - 2\gamma|$; however, there are some small changes. They include replacement of the exponent A of Eqs. (30), (32) by 1, and replacement of the preexponential factor γ_{cr} in Eq. (31) by $\gamma_{cr}/2$. The subsequent functions $f_n(t - t_n)$ differ from $f_0(t)$ by preexponential factors which are polynomials in t, whose degree increases with increasing n.

For $\gamma \gg 1$, the series (40) is replaced by the function

$$f(t)=(1-\beta t/2)\theta(2/\beta-t),$$

which coincides with the original transform $K_7(p)$.

The AC field leads to approximately the same variation in the relaxation as predicted by Subsecs. 1–6. In particular, a strong field causes the relaxation to become purely exponential in character, with times $T_{2u} = \tau_c(\chi/\beta)$ (for $\chi\tau_c > 1,\gamma$) and $T_{2v} = [4(\chi\tau_c)^{1/2}/\gamma]\tau_c$ (for $\chi\tau_c > 1,\gamma^2$) which are longer than in the absence of a field.

4. DISCUSSION OF RESULTS

The basic results of the theory developed here, which are independent of the strength of the random process, can be formulated briefly as follows:

1. For all distributions except the Cauchy distribution there exists a critical value of the strength of the noise $\gamma_{cr} = (\langle\Delta\omega^2\rangle_{cr})^{1/2}\tau_c$ such that for $\gamma > \gamma_{cr}$ an exponential evolution of the system is impossible. For $\gamma \leqslant \gamma_{cr}$ the relaxation of the polarization is described by a function which includes exponential and nonexponential decay terms. For $\gamma \ll 1$, in all cases we can neglect the nonexponential contribution with high accuracy; therefore the Bloch equations can be used to describe the kinetics. In the intermediate region $\gamma \sim 1$ (but $\gamma \leqslant \gamma_{cr}$) the contribution from nonexponential relaxation grows, and can reach 50% at $t = 0$. It should be noted that the Cauchy distribution is the only one for which the relaxation is purely exponential for all values of the parameter γ.

2. Turning on a weak AC field for $\gamma > \gamma_{cr}$ leads to the appearance of long exponential "tails" for long times, although the polarization during the initial instants of time relaxes nonexponentially. Turning on a strong field ($\chi > \tau_c^{-1}$, $(\langle\Delta\omega^2\rangle)^{1/2}$ transforms the kinetics so that the behavior of the system becomes exponential for all times. In addition, a strong field slows down the relaxation.

3. For $\gamma \leqslant 1$ all the distributions except for the Cauchy and $dW_7(\alpha)$ give the same expressions for the exponential relaxation times: $T_2 = (\langle\Delta\omega^2\rangle\tau_c)^{-1}$ for $\chi = 0$, $2T_{2u} = T_{2v} = 2\tau_c(\chi^2/\langle\Delta\omega^2\rangle)$ for $\chi\tau_c \gg 1$. The Cauchy distribution and $dW_7(\alpha)$ have other dependences on the parameters

$$T_2=\begin{cases}1/\beta & \text{Cauchy}\\ 2/\beta & (dW_7(\alpha))\end{cases} \quad \text{for} \quad \chi=0,$$

$$T_{2u}=\tau_c\begin{cases}\chi/\beta\\ 2\chi/\beta\end{cases},$$

$$T_{2v}=\tau_c\begin{cases}2(\chi\tau_c)^{1/2}/\gamma & \text{Cauchy}\\ 4(\chi\tau_c)^{1/2}/\gamma & (dW_7(\alpha))\end{cases} \quad \text{for} \quad \chi\tau_c \gg 1.$$

The qualitative differences in the distributions appear in the supercritical region, where the form of the relaxation function depends significantly on the choice of distribution function for the random field $dW(\alpha)$. In the large-γ limit the relaxation function in the absence of a field is found to be the Fourier transform of the distribution function for $dW(\alpha)$.

4. The value of the AC field amplitude at which a significant lengthening of the relaxation time occurs depends on the strength of the random process: for $\gamma \gg 1$ it occurs when $\chi T_2 \gg \chi\tau_c \gg 1$; for $\gamma \gtrsim 1$ it is sufficient to fulfill the less stringent condition $\chi\widetilde{T}_2 \sim 1$. In the latter case, when $\gamma \gg 1$ the relaxation time lengthens appreciably (by a factor $\sim\gamma$). In the telegraph-noise model the character of the relaxation slowing-down does not depend on the strength of the noise.

5. For the case of a strong random process the relaxation time of the u and v polarization components depend in different ways on the amplitude of the AC field. Thus, as described in the previous subsection, T_{2u} increases when the value of χ is on the order of the average value of the local field $(\langle\Delta\omega^2\rangle)^{1/2}$, while in order to substantially lengthen T_{2v} it is necessary to fulfill the more stringent condition $\chi > \gamma(\langle\Delta\omega^2\rangle)^{1/2}$. Such behavior of the relaxation time corresponds to the Redfield scenario, in which the kinetics of the spin system changes in a strong resonance field. This change in kinetics was predicted on the basis of a hypothesis concerning the existence of a spin temperature in a rotating coordinate system.[20] Hence, we have shown that the change in relaxation can be explained within the spirit of Redfield's scenario, but starting from fundamentally different considerations, the basic factors being strong coupling of the dynamic system with a heat bath and the existence of a continuous band of values of the random variable $\Delta\omega$ reflecting the presence of large numbers of states in the heat bath.

These conclusions were arrived at based on our investigation of seven distributions for scatter in the values of the resonance frequency $\Delta\omega$. These are model distributions; each can serve more or less as an approximation to the real distribution.

Let us note that two of these model distributions, and one other (which was not discussed here), are given by the Fourier transforms of $\exp(-\Delta|t|^n)$ where $n = \frac{1}{2}, 1,2$; these functions were also used in Refs. 12, 21 for analyzing phase relaxation in the absence of a resonance field. The Gaussian distribution ($n = 2$) is the only one for which an analysis was given of the effect of a radiation field on the kinetics with the goal of describing the contribution to the local field $\Delta\omega$ from the distant environment of an impurity center. In order to describe the contribution to $\Delta\omega$ from the small number of spins in the immediate vicinity of the ion which are ordered in the crystal lattice, it may be necessary to use a different distribution.

As a limiting simple case which typifies these short-range effects, we can use the situation where a single spin which has only two states is located near the center under study. In the first state this spin causes a shift in the resonance frequency of the center by the value $+\alpha$, in the second by $-\alpha$; these states are occupied with equal probabilities (the high-temperature approximation). This case corresponds to telegraph noise, i.e., a two-step random function $\alpha(t)$. In this model the Redfield scenario is not realized, and the dependence of the relaxation time on the amplitude of the AC field is roughly the same as in the case of a weak random process in the other models. This is a consequence of the statistical poverty of telegraph noise and does not contradict Redfield's idea, because the latter was formulated for a physical system which interacts with a subsystem possessing a large number of degrees of freedom.

It is more realistic to consider a situation where the deviation of the frequency $\Delta\omega$ of the center under study is caused by a small number of spins. Then $\Delta\omega$ should take on a finite set of discrete values. In particular, when all the spins belong to the first coordination sphere and are positioned in the same way relative to the center under study, these discrete values of $\Delta\omega$ are equidistant, while the probability of achieving a given value of $\Delta\omega$ decreases monotonically as $\Delta\omega$ increases. This is the situation we have modeled with the distribution $dW_7(\alpha)$, which has a sharp maximum of $\pi(n+\frac{1}{2})$ at $\alpha = 0$. Analysis of the kinetics shows that the Redfield scenario is realized in the present case only because of broadening of the spectrum of discrete values of $\Delta\omega$, i.e., the participation of a large number of spins.

5. AN ESTIMATE OF THE STRENGTH OF THE RANDOM PROCESS FOR Pr^{3+} IONS in LaF_3

In conclusion, let us estimate the strength of the random process which corresponds to irreversible phase relaxation of Pr^{3+} impurity ions in LaF_3, and compare it with the critical values of the theory. It is well-known that the resonance transition ${}^3H_4$–${}^1D_2$ of the Pr^{3+} ion has a strong static inhomogeneous broadening of 2.5 GHz.[2] At low temperatures ($T = 1.6$ K) the phonon relaxation mechanism is "frozen out" and spectral packets of the static lineshape are broadened due to magnetic interactions with fluorine nuclei, which cause fluctuations in the packet frequency $\Delta\omega(t)$. This process can be treated either as division of the static packet into a collection of "magnetic" subpackets among which the ion migrates, or as a random time variation of the frequency of each static packet. The well-known mean-square displacement of the resonant frequency of a packet $(\langle\Delta\omega^2\rangle)^{1/2}$ (Refs. 2,3,22) almost coincides with the measured "homogeneous" line broadening $(2\pi T_2)^{-1} = 7.3$ kHz.[1,23] Therefore, to first order we can ignore the variations in the static packets by this mechanism, i.e., neglect the migration of the resonant frequency over the static spectrum. Then we can apply the theory we have developed for an isolated static packet to estimate the characteristics of the random process which causes irreversible phase relaxation of Pr^{3+}.

Because we have shown that all the distributions (except for the Cauchy and telegraph noise distributions) lead to qualitatively the same results, we choose to analyze the experimental data with the theory based on the uniform distribution. The problem reduces to establishing the relation between the process parameters τ_c and $(\langle\Delta\omega^2\rangle)^{1/2}$ and the known values of T_2 as $\chi\to 0$. In Ref. 2, two possible estimates were given for the correlation time τ_c of the reservoir, which is made up of the system of ${}^{19}F$ nuclear spins. According to the first estimate, τ_c is the spin-spin relaxation time caused by magnetic dipole-dipole interactions among the nuclei throughout the whole crystal volume. The experimental value of the latter equals 16.4 μsec.[24] This estimate gives too large a value for the rate τ_c^{-1} of the process, which actually should be smaller because the large magnetic moment of a Pr^{3+} ion shifts the resonance frequencies of neighboring ${}^{19}F$ nuclei, and thereby hinders the flip-flop processes by which they interact with ${}^{19}F$ nuclei throughout the crystal volume.[22] Therefore, the spin-spin relaxation time for nuclei near a Pr^{3+} ion may be longer than the one observed in experiment for bulk nuclei.[24] A second estimate for τ_c was made based on direct calculation of the correlation function $\langle\Delta\omega(t)\Delta\omega\rangle$ using the Monte Carlo method; this function is found to decay exponentially with a time $\tau_c = 70\,\mu$sec.[2] In this calculation there were no free parameters and the only assumption used was that spin reversals were uncorrelated.[25]

In comparing theory and experiment the value $\tau_c = 20$ μsec given in Ref. 2 was used, which is close to the first estimate. This is probably connected with the fact that the kinetics were described using the Bloch type of differential equations, which are valid when the condition $\tau_c < T_2 = 21.7\,\mu$sec holds. The second method estimates the times for precisely those correlations which take part in the relaxation process; however, as shown by the authors of Ref. 2, its use requires a new theory which is not limited by the condition $\langle\Delta\omega^2\rangle\tau_c^{\,2} \ll 1$. In this paper we have proposed just such a theory.

So as to understand the extent to which this new theory is necessary, let us estimate the strength of the random process $\delta\tau_c$ (where $\delta = (\langle\Delta\omega^2\rangle)^{1/2}$), using known values of T_2 and τ_c, and compare them with the critical value $(\delta\tau_c)_{cr}$. Perturbation theory gives the following expression for the strength of the process: $\delta\tau_c = (\tau_c/T_2)^{1/2}$. This expression is valid when $\tau_c \ll T_2$. For arbitrary ratios of the times τ_c and T_2 it is necessary to use a different theoretical estimate which is not limited to certain ranges of the strength of the process:

$$3^{1/2}\delta\tau_c \,\mathrm{ctg}(3^{1/2}\delta\tau_c) = 1 - \tau_c/(T_2)_{\exp} \quad (\delta\tau_c \leqslant (\delta\tau_c)_{cr} = 0.9),$$

$$3^{1/2}\delta\tau_c/\mathrm{arctg}(3^{1/2}\delta\tau_c) = 1 + \tau_c/T_2 \quad (\delta\tau_c - \text{arbitrary}),$$

$(T_2)_{\exp}$ is the exponential relaxation time which applies to the subcritical regime; T_2 is the general (integrated) relaxation time. In the case $\tau_c = 70$ μsec we obtain a value of $\delta\tau_c = 3.42$ for the strength, which is larger than critical by almost a factor of 4, implying that the random process which causes phase relaxation of Pr^{3+} is a strong one. This assertion remains in force even when the correlation time τ_c coincides with the relaxation time, because in this case $\delta\tau_c$ is 50% larger than the critical value $(\delta\tau_c)_{cr} = 0.9$.

[1] R. G. DeVoe and R. G. Brewer, Phys. Rev. Lett. **50**, 1269 (1983).
[2] A. Schentzle, M. Mitsunaga, R. G. DeVoe, and R. G. Brewer, Phys. Rev. **A30**, 325 (1984).
[3] E. Hanamura, J. Phys. Soc. Jpn. **52**, 2258 (1983).
[4] E. Hanamura, J. Phys. Soc. Jpn. **52**, 2267 (1983); J. Phys. Soc. Jpn. **52**, 3265 (1983); J. Phys. Soc. Jpn. **52**, 3678 (1983).

[5]P. A. Apanasevich, S. Ya. Kilin, A. P. Nizovtsev, and N. S. Onishchenko, Opt. Commun. **52**, 279 (1984); J. Opt. Soc. Am. **B3**, 587 (1986).
[6]M. Yamanoi and J. H. Eberly, Phys. Rev. Lett. **52**, 1353 (1984).
[7]K. Vódkiewicz and J. H. Eberly, Phys. Rev. **A31**, 2314 (1985); Phys. Rev. **A32**, 992 (1985).
[8]P. R. Berman and R. G. Brewer, Phys. Rev. **A32**, 2784 (1985).
[9]K. Wódkiewicz, B. W. Shore, and J. H. Eberly, Phys. Rev. **A30**, 2390 (1984).
[10]G. Hazak, M. Strauss, and J. Oreg, Phys. Rev. **A32**, 3475 (1985).
[11]A. G. Jodth, J. Golub, W. W. Carlson, and T. W. Mössberg, Phys. Rev. Lett. **53**, 659 (1984).
[12]A. I. Burshtein, Zh. Eksp. Teor. Fiz. **48**, 850 (1965) [Sov. Phys. JETP **21**, 567 (1965)]; Zh. Eksp. Teor. Fiz. **49**, 1362 (1965) [Sov. Phys. JETP **22**, 939 (1965)].
[13]A. I. Burshtein and Yu. S. Oselebchik, Zh. Eskp. Teor. Fiz. **51**, 1071 (1966) [Sov. Phys. JETP **24**, 716 (1966)].
[14]A. I. Burshtein, Zh. Eksp. Teor. Fiz. **54**, 1120 (1968) [Sov. Phys. JETP **27**, 600 (1968)].
[15]V. M. Fain, *Kvantovaya Radiofizika (Quantum Radiophysics). V. 1: Fotony i Nelineinye Sredy (Photons and Nonlinear Media).* Moscow: Sov. Radio, p. 126 (1972).
[16]N. Bloembergen, E. M. Purcell, and R. V. Pound, Phys. Rev. **73**, 679 (1948).
[17]P. Noack and G. Held, Z. Phys. **210**, 60 (1968).
[18]V. D. Korepanov, *Elektromagnitnoye Sverkhizlucheniye* (*Electromagnetic Superradiance*). Kazan', 1975, p. 383.
[19]M. A. Abramowitz and I. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1975).
[20]A. G. Redfield, Phys. Rev. **98**, 1787 (1955).
[21]A. B. Doktorov and A. I. Burshtein, Zh. Eksp. Teor. Fiz. **63**, 784 (1972) [Sov. Phys. JETP **36**, 411 (1972)].
[22]R. M. MacFarlane, C. S. Yannoni, and R. M. Shelby, Opt. Commun. **32**, 101 (1980).
[23]R. M. MacFarlane, R. M. Shelby, and R. L. Shoemaker, Phys. Rev. Lett. **43**, 1726 (1979).
[24]L. Shen, Phys. Rev. **172**, 259 (1968).
[25]R. G. DeVoe, A. Wokaun, S. C. Rand, and R. G. Brewer, Phys. Rev. **B23**, 3125 (1981).
[26]M. J. Weber, J. Chem. Phys. **48**, 4774 (1968).

Translated by Frank J. Crowne