Kinetics of polarized secondary radiation in pulsed resonant excitation of excitons in crystals

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We consider the influence of multiple reradiation-reabsorption and specular reflection from an inner surface on the kinetics of the intensity damping and polarization of secondary radiation in resonant pulsed excitation of excitons in a cubic crystal and in elastic scattering of excitons by impurities. We calculate the mean damping times and the temporal dependences of the radiation intensity and polarization, using successive approximations up to fourth order in the contribution of the scattering. We show that measurements of the polarized-radiation intensity under conditions of stationary and pulsed excitation can determine directly the exciton radiative or nonradiative lifetimes.

Advances in pulsed laser techniques and in methods of recording short pulses has uncovered many possibilities of investigating nonstationary processes of light scattering in semiconducting crystals. Interest has therefore greatly increased lately in the study of the kinetics of exciton-luminescence damping in pulsed excitation, which permits the kinetic parameters of excitons and the fine structure of their energy spectrum to be determined.

Thus, many measurements of the group velocities and of the damping times have been made for certain branches of the polariton spectrum in A_3B_5 (GaAs) and A_2B_6 (CdS, CdSe) crystals excited by picosecond pulses (see the review of Koteles[1]). Similar experiments were performed also on molecular crystals of large optical thickness, i.e., at $\alpha d \gg 1$ (d is the crystal thickness and α is the absorption coefficient at the resonant frequency).[2] Analysis of the results of these experiments shows that multiple reabsorption-reradiation processes influence strongly the time dependence of the exciton-radiation decay, and particularly its average damping time. Reradiation is most substantial in optically thick crystals at not too low values of the quantum yield ($\tilde{\omega}_0 = \tau_{\text{life}}/\tau_{\text{rad}}$ of the elementary light-scattering act ($\tau_{\text{life}}^{-1} = \tau_0^{-1} + \tau_{\text{rad}}^{-1}$, τ_0 and τ_{rad} are the nonradiative and radiative exciton lifetimes).

The principal problems of nonstationary radiation-transport theory were formulated in astrophysics, and independent methods for their solution were developed in the papers by Sobolev,[3] Minin,[4] Stepanov and Samson,[5] and others (a complete bibliography can be found, e.g., in Ref. 5). In particular, Stepanov and Samson developed a successive-approximation method based on inclusion of different orders of the radiation and quite effective at not too large of $\tilde{\omega}_0$. A theory of the temporal kinetics of exciton luminescence under conditions of strong reradiation was developed for crystals in the papers of Agranovich *et al.*, the contents of which are summarized in a monograph by Agranovich and Galanin.[2] They have generalized Ambartsumyan's method of stationary transport theory to include the case of propagation of nonstationary radiation. In none of these papers, however, was the kinetics of the radiation polarization investigated, nor was account taken of reflection and refraction of the light by the boundary of the medium. A method for calculating the secondary-radiation polarization in stationary excitation was developed for astrophysics by Chandrasekhar.[6] This method was generalized in Ref. 7 for solids, where reflection from the surface must be taken into account, and was used to formulate a stationary theory of polarized-radiation transport in the exciton region of the spectrum.

We consider theoretically in this paper the damping kinetics of polarized secondary radiation produced by resonant pulsed excitation of excitons by polarized light in a semi-infinite crystal, with account taken of multiple reflection of the scattered light from an inner surface of the crystal. As in Ref. 7, we consider the case of dipole-active exciton resonance in a crystal of cubic symmetry, and elastic scattering excitons by impurities, at equal frequencies of the incident and emitted light. In analogy with Ref. 7, we neglect the polariton effect and diffusion of the excitons, i.e., their displacement during their lifetime, as well as their spin relaxation. The crystal is assumed to be excited by a δ-function pulse. The solution yields in the usual manner the time dependences at arbitrary waveform of the exciting pulse.

If the foregoing conditions are satisfied, the probability of photon emission in a time interval from t to $t + dt$ after absorbing a primary photon and creation of an exciton at the instant $t = 0$, depends only on the exciton lifetime τ_{life} and is proportional to $\exp(-t/\tau_{\text{life}})\, dt/\tau_{\text{life}}$. The angular distribution of the emission after single scattering is described by the formula

$$\hat{I}(\Omega, t) = \tilde{\omega}_0 \int_0^t \hat{P}(\Omega, \Omega')\, \hat{I}(\Omega', t') \exp\left(-\frac{t-t'}{\tau_{\text{life}}}\right) \frac{dt'}{\tau_{\text{life}}}, \qquad (1)$$

where $\hat{I}(\Omega', t)$ is the Stokes four-component column matrix for primary radiation propagating in the direction of a unit vector $\Omega'(\theta', \varphi')$ specified by the spherical angles θ' and φ', $\hat{I}(\Omega, t)$ is the same for the secondary radiation, and $\hat{P}(\Omega, \Omega')$ is the Rayleigh-scattering angle matrix.

If the crystal is illuminated by a pulse of polarized plane monochromatic light wave $\hat{I}_0$ at an angle $\theta_1 = \arccos(-\mu_1)$ at the instant $t = 0$, we have for the entering radiation after refraction at the angle $\theta_0 = \arccos(-\mu_0)$, at the inner boundary of the crystal ($Z = 0$)

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$$\hat{I}_0(+0,\ \Omega,\ t)=\pi\hat{F}\delta(\Omega-\Omega_0)\,\delta(t),\qquad \pi\hat{F}=(\mu_1/\mu_0)\,\hat{T}\,(\mu_0,\ \mu_1)\,\hat{I}_0. \tag{2}$$

Here $\mu_1,\ \mu_0>0$, $\hat{T}$ is the matrix of the energy transmission coefficients (Ref. 7, Eq. 11). The nonstationary transport equation for the Stokes matrix $\hat{I}(\Lambda,\Omega,t)$ of the scattered radiation can then be represented, taking (1) and (2) into account, in the form

$$\mu\frac{\partial\hat{I}(\Lambda,\Omega,t)}{\partial\Lambda}+\frac{n_0}{\alpha c}\frac{\partial\hat{I}(\Lambda,\Omega,t)}{\partial t}$$

$$=\hat{I}(\Lambda,\Omega,t)-\frac{\tilde{\omega}_0}{4\pi}\int d\Omega'\,\hat{P}(\Omega,\Omega')$$

$$\times\int_0^t\frac{dt'}{\tau_{\text{life}}}\hat{I}(\Lambda,\Omega',t')\exp\left(-\frac{t-t'}{\tau_{\text{life}}}\right)-\frac{\tilde{\omega}_0}{4}\exp\left(-\frac{\Lambda}{\mu_0}\right)$$

$$\times\hat{P}(\Omega,\Omega_0')\,\hat{F}\exp\left(-\frac{t}{\tau_{\text{life}}}\right), \tag{3}$$

where $\Lambda=\alpha z$, z is the distance from the surface, n_0 is the refractive index of the crystal at the frequency of the crossing of the exciton and photon branches, and c is the speed of light in vacuum. The quantity $\tau_{\text{ph}}=n_0/\alpha c$ in (3) is the free-path time of the photon in the crystal, i.e., the average time between the photon emission and its absorption. We consider crystals with weak exciton-photon interaction and with spatial dispersion. For these crystals there is no polariton effect and the following condition is satisfied:

$$\tau_{\text{ph}}\ll\tau_{\text{life}}. \tag{4}$$

Recognizing that

$$\tau_{\text{life}}=\tilde{\omega}_0\tau_{\text{rad}}=\tilde{\omega}_0\,(l_{\text{rad}}/v_{\mathbf{q}_0})\approx\tilde{\omega}_0\,(\alpha v_{\mathbf{q}_0})^{-1},$$

the condition (4) reduces to the inequality $v_{\mathbf{q}_0}\ll v_{\text{ph}}\tilde{\omega}_0$, where $l_{\text{rad}}\approx\alpha^{-1}$ is the mean free path of the exciton with respect to radiative recombination, $v_{\mathbf{q}_0}$ is the group velocity of the exciton at the resonant frequency $\tilde{\omega}_0(\mathbf{q}_0)$, and $v_{\text{ph}}=c/n_0$ is the photon propagation velocity in the crystal. The second term in the left-hand side of (3) can be neglected).

Equation (3) can be solved in general form by means of the Laplace transformation used, e.g., in Ref. 4. For the Laplace-transformed matrix function

$$\hat{I}(\Lambda,\Omega,p)=\int_0^\infty\exp\left(-p\frac{t}{\tau_{\text{life}}}\right)\hat{I}(\Lambda,\Omega,t)\,dt/\tau_{\text{life}}, \tag{5}$$

where p is the Laplace parameter, we obtain from (3) the equation

$$\mu\frac{d\hat{I}(\Lambda,\Omega,p)}{d\Lambda}=\hat{I}(\Lambda,\Omega,p)-\frac{\tilde{\omega}_0}{4\pi}\frac{1}{1+p}\int d\Omega'\,\hat{P}(\Omega,\Omega')$$

$$\times\hat{I}(\Lambda,\Omega',p)-\frac{\tilde{\omega}_0}{4}\frac{1}{1+p}\exp\left(-\frac{\Lambda}{\mu_0}\right)\hat{P}(\Omega,\Omega_0)\,\hat{F}, \tag{6}$$

which differs from the corresponding equation (5) of Ref. 7 only in that $\tilde{\omega}_0$ is replaced by $\omega_0/(1+p)$. To take the inverse Laplace transform and find $\hat{I}(\Lambda,\Omega,t)$ an analytic solution of Eq. (6) is necessary. In Ref. 7 there was developed, besides a numerical method suitable for any $\tilde{\omega}_0$, also an $\hat{S}$-matrix iteration method with which to obtain an analytic solution in the form of an expansion of $\hat{I}(+0,\Omega)$ in powers of $\tilde{\omega}_0$:

$$\hat{I}(+0,\Omega)=\sum_{\nu=1}\frac{\tilde{\omega}_0^{\nu}}{4\mu}\hat{S}_\nu^{\text{R}}(\Omega,\Omega_0)\,\hat{F}. \tag{7}$$

The matrices $\hat{S}$ are defined by a matrix integral equation [Ref. 17, Eq. (26)]. Solution of this equation, in accordance with Eq. (33) of Ref. 7, reduces to iteration of relatively simple equations [Eqs. (36), (42), and (46) of Ref. 7 for the matrices $\hat{A}$, $\hat{b}$, and $\hat{a}$].

Replacing $\tilde{\omega}_0$ by $\tilde{\omega}_0/(1+p)$ in (7) and taking the inverse Laplace transform, we get

$$\hat{I}(+0,\Omega,t)$$

$$=\sum_{\nu=1}\frac{\tilde{\omega}_0^{\nu}}{4\mu}\hat{S}_\nu^{\text{R}}(\Omega,\Omega_0)\,\hat{F}\frac{1}{(\nu+1)!}\left(\frac{t}{\tau_{\text{life}}}\right)^{\nu-1}\exp\left(-\frac{t}{\tau_{\text{life}}}\right). \tag{8}$$

In accordance with Ref. 7, for $\nu=1,2$ the matrices $\hat{S}_\nu^R$ are determined by the following expressions: at $n_0=1$, when $\hat{S}^{\text{R}}\equiv\hat{S}$, we have

$$\hat{S}_1(\Omega,\Omega_0)=\frac{\mu\mu_0}{\mu+\mu_0}\hat{P}(\mu,\varphi;-\mu_0,\varphi),$$

$$\hat{S}_2(\Omega,\Omega_0)=\frac{\mu\mu_0}{\mu+\mu_0}\frac{1}{4\pi}\int_0^1 d\mu'\int_0^{2\pi}d\varphi'\left[\frac{\mu_0}{\mu+\mu_0}\hat{P}(\mu,\varphi;\mu',\varphi')\right.$$

$$\times\hat{P}(\mu',\varphi';-\mu_0,\varphi_0)$$

$$\left.+\frac{\mu}{\mu+\mu_0}\hat{P}(\mu,\varphi;-\mu',\varphi')\,\hat{P}(-\mu',\varphi';-\mu_0,\varphi_0)\right],$$

and at $n_0^2\neq1$

$$\hat{S}_1^{\text{R}}(\Omega,\ \Omega_0)=\hat{S}_1(\Omega,\ \Omega_0),$$

$$\hat{S}_2^{\text{R}}(\Omega,\Omega_0)=\hat{S}_2(\Omega,\Omega_0)+\frac{\mu\mu_0}{4\pi}\int_0^1\frac{\mu'd\mu'}{(\mu_0+\mu')\,(\mu+\mu')}\int_0^{2\pi}d\varphi'$$

$$\times\hat{P}(\mu,\varphi;-\mu',\varphi')\,\hat{R}(\mu')\,\hat{P}(\mu',\varphi';-\mu_0,\varphi_0),$$

where the matrix $\hat{R}(\mu)$ of the specular-reflection coefficients is determined by Eq. (8) of Ref. 7.

In accordance with (8), the Stokes matrix $\hat{I}(-0,\Omega_2,t)$ for radiation emitted from a crystal in the direction $\Omega_2=\Omega_2(\theta_2,\varphi)$, takes the form

$$\hat{I}(-0,\Omega_2,t)=\exp\left(-\frac{t}{\tau_{\text{ж}}}\right)\sum_{\nu=1}\frac{\tilde{\omega}_0^{\nu}}{(\nu-1)!}\left(\frac{t}{\tau_{\text{life}}}\right)^{\nu-1}\hat{I}^{(\nu)}(-0,\Omega_2)$$

$$=\exp\left(-\frac{t}{\tau_{\text{ж}}}\right)\sum_{\nu=1}\frac{\tilde{\omega}_0^{\nu}}{(\nu-1)!}\left(\frac{t}{\tau_{\text{life}}}\right)^{\nu-1}\frac{1}{4\mu_2}\hat{S}_\nu^{\text{H}}(\Omega_2,\Omega_1)\,\hat{F}. \tag{9}$$

Here

$$\hat{S}_\nu^{\text{H}}(\Omega_2,\Omega_1)=\frac{1}{n_0^2}\frac{\mu_2\mu_1}{\mu\mu_0}\hat{T}\,(\mu_2,\mu)\,\hat{S}_\nu^{\text{R}}(\Omega,\Omega_0)\,\hat{T}\,(\mu_0,\mu_1),$$

where $\Omega_1=\Omega_1(\theta_1,\varphi_0)$, $\mu_2=\cos\theta_2$ and $\mu=\cos\theta$, while μ_1 and μ_2 connected with μ_0 and μ by Snell's law.

We introduce in analogy with Ref. 2 the average damping times of the total intensity $I=I_l+I_r$ and of the intensities $Q=I_l-I_r=IP_{\text{lin}}$ and $V=I_+-I_-=IP_{\text{circ}}$ of the linearly and circularly polarized radiation. These times are defined as

$$\tau_i(n_0, \tilde{\omega}_0, \Omega_2) = \frac{\int_0^\infty I_i(-0, \Omega_2, t)\, t\, dt}{\int_0^\infty I_i(-0, \Omega_2, t)\, dt}, \qquad (10)$$

where $I_1 = I, I_2 = Q$, and $I_3 = V$. Substituting I_i from (9) in (10) we get

$$\tau_i = \frac{\sum_{\nu=1} \nu \tilde{\omega}_0^\nu I_i^{(\nu)}(-0, \Omega_2)}{I_i(-0, \Omega_2)} \tau_{\text{life}}, \qquad (11)$$

where $I_i(-0, \Omega_2)$ is the solution of the stationary transport equation. In (11), $\tilde{\omega}_0^\nu I_i^\nu(-0, \Omega_2)$ is the contribution to the Stokes parameter $I_i(-0, \Omega_2)$ by the ν-fold scattered radiation. It can be seen from (11) that with increase of $\tilde{\omega}_0$ and n_0 the mean damping times of the intensity I and of the components Q and V of the secondary radiation become longer, especially at $n_0 \gg 1$.

To determine the degree of retardation of the intensity and of the polarization of the scattered radiation we introduce the quantity $\gamma_i = (\tau_i - \tilde{\tau}_i)/\tilde{\tau}_i$, where τ_i is determined by Eq. (11), and $\tilde{\tau}_i$ is equal to

$$\tilde{\tau}_i(n_0, \tilde{\omega}_0, \Omega_2) = \frac{\int_0^\infty I_i(-0, \Omega_2, t)\, dt}{I_i(-0, \Omega_2)}. \qquad (12)$$

According to (9), in the case considered here $\tilde{\tau}_i \equiv \tau_{\text{life}}$ and does not depend on the propagation direction Ω_2, and accordingly $\gamma_i = (\tau_i - \tau_{\text{life}})/\tau_{\text{life}}$.

The matrices $\hat{S}$ were calculated numerically with a computer by successive iterations of the corresponding equations of Ref. 7 up to fourth order inclusive. At normal incidence of the exciting light ($\mu_1 = \mu_2 = 1$), for radiation propagating normal to the crystal surface ($\mu_2 = \mu = 1$) we obtain at $n_0 = 1$

$$\begin{aligned} \tau_1/\tau_{\text{life}} &= 1 + 0{,}60\tilde{\omega}_0 + 0{,}46\tilde{\omega}_0^2 + 0{,}29\tilde{\omega}_0^3, \\ \tau_2/\tau_{\text{life}} &= 1 + 0{,}44\tilde{\omega}_0 + 0{,}25\tilde{\omega}_0^2 + 0{,}12\tilde{\omega}_0^3, \\ \tau_3/\tau_{\text{life}} &= 1 + 0{,}29\tilde{\omega}_0 + 0{,}11\tilde{\omega}_0^2 + 0{,}03\tilde{\omega}_0^3, \end{aligned} \qquad (13)$$

and at $n_0^2 = 10$

$$\begin{aligned} \tau_1/\tau_{\text{life}} &= 1 + 0{,}77\tilde{\omega}_0 + 0{,}68\tilde{\omega}_0^2 + 0{,}47\tilde{\omega}_0^3, \\ \tau_2/\tau_{\text{life}} &= 1 + 0{,}52\tilde{\omega}_0 + 0{,}33\tilde{\omega}_0^2 + 0{,}13\tilde{\omega}_0^3, \\ \tau_3/\tau_{\text{life}} &= 1 + 0{,}28\tilde{\omega}_0 + 0{,}10\tilde{\omega}_0^2 + 0{,}01\tilde{\omega}_0^3. \end{aligned} \qquad (14)$$

It can be seen that in multiple scattering the intensities of the polarized radiation Q and V attenuate more rapidly than the total intensity I, and specular reflection from the inner surface of the crystal lengthens the damping time of the linear polarization and shortens that of the polarized one. The latter is due to the fact that the circular polarization reverses sign upon specular reflection.

Figure 1 shows the time dependences of the light intensities $I^{(\nu)}$ of different scattering orders at excitation geometry $\mu_1 = \mu_0 = 1$ and observation geometry $\mu_2 = \mu = 1$, when the total scattered-light intensity is independent of the incident-radiation polarization. The solid curves describe the change of the total intensity with time, with allowance for the scattering contribution up to fourth order, inclusive:

$$I(-0.t) = \beta I_0 \tilde{\omega}_0 \left[1 + 0.77\tilde{\omega}_0 \frac{t}{\tau_{\text{life}}} + 0{,}63\frac{1}{2!}\left(\tilde{\omega}_0 \frac{t}{\tau_{\text{life}}}\right)^2 + 0{,}49\frac{1}{3!}\left(\tilde{\omega}_0 \frac{t}{\tau_{\text{life}}}\right)^3\right] e^{-t/\tau_{\text{life}}}, \qquad (15)$$

$$\beta = \frac{3}{2\pi} \cdot 0.6664 \cdot 10^{-2}.$$

It can be seen that at $\tilde{\omega}_0 = 0.2$ the main contribution to $I(-0,t)$ is made by $I^{(1)}(0,t)$ and the delay is $\gamma_1^{(4)} \approx 18\%$. With increaseing $\tilde{\omega}_0$ the average time $\tilde{\tau}_1$ increases sharply. Thus, at $\tilde{\omega}_0 = 0.6$ the delay γ_1^4 is already 81%. We note that at $n_0 = 1$ and at the same values of $\tilde{\omega}_0$ the respective delays are $\gamma_1^{(4)} \sim 14\%$ and 59%. It can be seen that allowance for multiple reflection of the scattered light from the crystal boundary increases strongly the average damping time. In

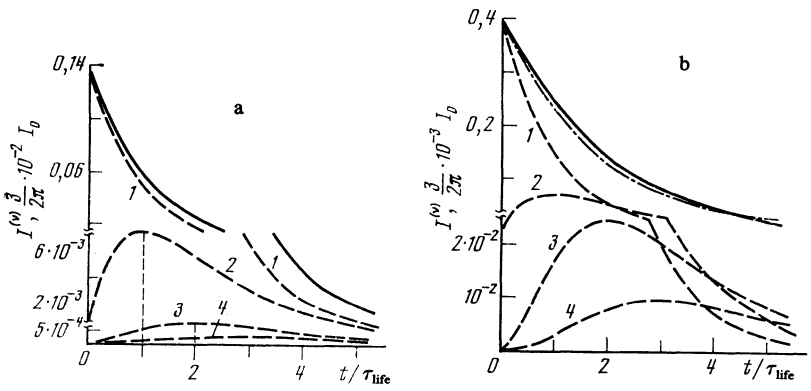

FIG. 1. Time dependences of the radiation intensity at $n_0^2 = 10$, $\mu_1 = \mu_0 = \mu = \mu_2 = 1$, calculated with allowance for the scattering contribution up to fourth order. The dashed curves indicate the contribution to $I(t)$ due to scattering of order ν (curves 1–4 correspond to $\nu = 1$–4, a) $\tilde{\omega}_0 = 0.2$ and b) $\tilde{\omega}_0 = 0.6$. Figure b is a plot of $I(t) = I(0)\exp(-t/\tau_1)$. At $\tilde{\omega}_0 = 0.2$ this curve differs from the exact one by not more than 1%.

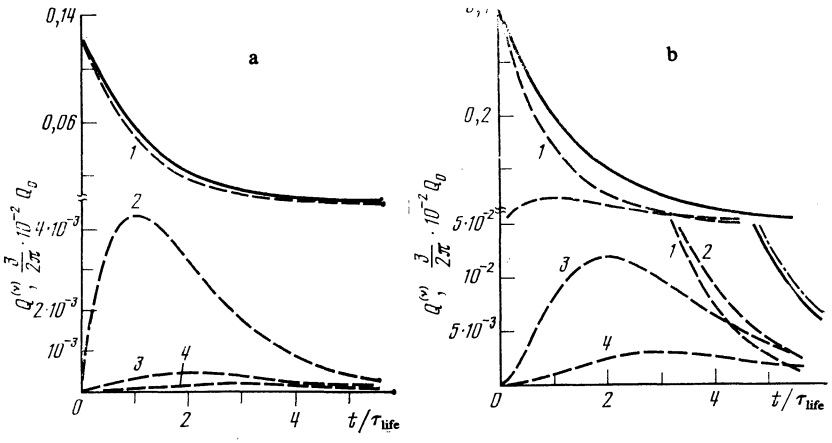

FIG. 2. Time dependences of $Q(t)$ for linearly polarized excitation. The dash-dot lines are plots of $Q(t) = Q(0)\exp(-t/\tau_2)$. The deviation from the exact curve in the remaining sections does not exceed 1% at $\tilde{\omega}_0 = 0.2$ and 5% at $\tilde{\omega}_0 = 0.6$. The remaining parameters and symbols are the same as in Fig. 1.

addition, the character of the time dependence of the intensity is somewhat altered, and the change can be assessed by comparing the calculated $I(t)$ with the exponential curve $I(0)\exp(-t/\tau_1)$ shown dashed in Fig. 1.

Figures 2 and 3 show analogous time dependences for $Q(t)$ and $V(t)$ at $n_0^2 = 10$:

$$Q(t)=\beta Q_0\tilde{\omega}_0\left[1+0.52\tilde{\omega}_0\frac{t}{\tau_{life}}+0.30\frac{1}{2!}\left(\tilde{\omega}_0\frac{t}{\tau_{life}}\right)^2+0.15\frac{1}{3!}\left(\tilde{\omega}_0\frac{t}{\tau_{life}}\right)^3\right]e^{-t/\tau_{life}}, \tag{16}$$

$$V(t)=\beta V_0\tilde{\omega}_0\left[1+0.28\tilde{\omega}_0\frac{t}{\tau_{life}}+0.10\frac{1}{2!}\left(\tilde{\omega}_0\frac{t}{\tau_{life}}\right)^2+0.02\frac{1}{3!}\left(\tilde{\omega}_0\frac{t}{\tau_{life}}\right)^3\right]e^{-t/\tau_{life}}. \tag{17}$$

It can be seen that scatterings of higher order contribute less to $Q(t)$ and $V(t)$ than to $I(t)$. At $\tilde{\omega}_0 = 0.2$ we have respectively $\gamma_2^{(4)} = 12\%$, $\gamma_3^{(4)} = 6\%$, while at $\tilde{\omega}_0 = 0.6$ we have $\gamma_2^{(4)} = 46\%, \gamma_3^{(4)} = 21\%$. The $Q(t)$ and $V(t)$ curves are closer to the exponential plots of $Q(0)\exp(-t/\tau_2)$ and $V(0)\exp(-t/\tau_3)$, which are also show dashed in these figures. Therefore if Eq. (15) can be used at $\tilde{\omega}_0 \leqslant 0.6$, expression (16) is valid for $\tilde{\omega}_0 \leqslant 0.8$ and (17) in fact for any $\tilde{\omega}_0$.

Measurement of the temporal characteristics $Q(t)$ and $V(t)$ yields therefore τ_2 and τ_3, from which, knowing $\tilde{\omega}_0$, we can determine τ_{life} and consequently τ_{rad} and τ_0. The value of $\tilde{\omega}_0$, as indicated in Ref. 7, can be determined from the polarization produced by stationary excitation. In the presence of noticeable spin relaxation one can determine independently τ_{life} and the spin-relaxation time τ_s, by comparing the time dependences $I(t)$, $V(t)$, and $Q(t)$, for example by determining $V(t)$ and $Q(t)$ as $t \to 0$. The times τ_{life} and τ_s can be also independently determined by measuring, alongside the time dependences also the dependence of the polarization the magnetic field. The corresponding calculation for the change of the linear polarization of radiation in a longitudinal magnetic field was carried out in Ref. 8.

In concusion, we take the opportunity to thank E. L. Ivchenko for much helpful advice.

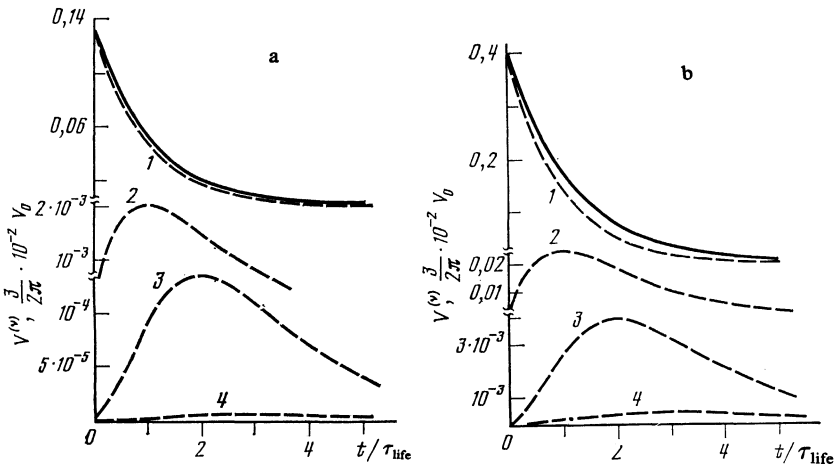

FIG. 3. Time dependences of $V(t)$ for circularly polarized excitation. The plot of $V(t) = V(0)\exp(-t/\tau_3)$ coincides with $V(t)$ accurate to 0.3% at $\tilde{\omega}_0 = 0.2$ and 1% at $\tilde{\omega}_0 = 0.6$. The remaining parameters and symbols are the same as in Fig. 1.

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