spectra, the temperature dependences of various impurity susceptibilities, the specific heat, and others.

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Self-induced transparency is a diphenyl crystal containing pyrene and excited by its own stimulated emission

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The optical self-induced transparency in a diphenyl crystal activated by pyrene is investigated theoretically and experimentally in the 1.6 to 4.1 K temperature range. For the first time in self-induced transparency studies the same crystal has been used as the generator of the stimulated radiation and as the object of the investigation. Optical pumping is carried out at 3530 Å; generation and self-induced transparency occur at 3739 Å which corresponds to the ${}^1B_{2u}\leftrightarrow{}^1A_g$ transition in pyrene. The self-induced transparency in the crystal studied was accompanied by a delay in the pulse (by 8–9 nsec) and by a characteristic kinetics of deformation and its shape (self-broadening, self-compression, self-division, depending on the pulse intensity at the entry into the resonant medium). Both an analytic and a numerical solution of the self-consistent problem of passage under conditions of a nonequilibrium level population density difference are obtained. The electric dipole moment is found by a computer search of the solution for an experimentally determined kinetics of pulse deformation in a resonant medium.

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1. INTRODUCTION

The phenomenon of self-induced transparency (SIT)[1] consists of the bleaching a resonant medium when a laser pulse propagates through the medium (under conditions of nonlinear coherent interaction) and its power exceeds a definite threshold. The SIT is characterized by a substantial delay of the pulse in the resonant medium, by its self-broadening (or self-compression, depending on the signal power entering the sample), and by a definite deformation of the pulse waveform.[2,3] The SIT method makes it possible to determine the irreversible relaxation times, the electric dipole moment of resonant transitions, and other spectroscopic information.[2–6]

In the present paper, the SIT procedure is used to investigate a diphenyl crystal activated with pyrene. This is a member of a new class of objects–impurity molecular crystals. Prior to this investigation, only one publication[7] reported observation of SIT in a similar object, namely in pentacene in *p*-terphenyl, without indication of any particular details on the conditions and singularities of the realization of the effect. The singularity of the SIT in our study lies in the fact that the crystal is excited by its own stimulated emission, i.e., it is simultaneously the radiation generator and the object of the investigation. A numerical analysis of

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the deformation of the pulse waveform with increase of its intensity at the entry into the sample has made it possible to establish the electric dipole moment of the resonant transition.

2. APPARATUS AND MEASUREMENT PROCEDURE. RESULT OF EXPERIMENTAL INVESTIGATIONS

A scheme of those energy levels of pyrene in diphenyl which participate in the light generation and in the formation of the SIT phenomenon is shown in Fig. 1a. The light is absorbed in the transition 1–3 (at the wavelength $\lambda_{13}=3530$ Å), and then, an account of a very rapid nonradiative transition 3–2 (the lifetime of the 3 level is $\tau_{32}\approx 10^{-11}$ sec) the impurity lands on the quasi-equilibrium level 2. The luminescence (generation) in the SIT is effected in the transition ${}^1B_{2u}\rightarrow {}^1A_g(2-1)$ at a wavelength $\lambda_{21}=3739.3$ Å. Under the specified conditions, it was possible to generate light on the (2–4) transition into the state 4, which corresponds to fully symmetrical oscillation at the frequency 1408 cm^{-1}. Inasmuch as absorption on the 4–2 transition (because of the short lifetime of the 4 level) was excluded, a comparison of the kinetics of the generation on the 2–4 transition with the generation on the 2–1 transition has made it possible to conclude that the observed pulse deformation (at the wavelength λ_{21}) is due to the SIT effect.

The diphenyl crystal belongs to the monoclinic system, containing two molecules per unit cell.[8] The pyrene molecule replace the lattice molecules in such a way, that the absorption of light on the resonant 0–0 transition (2–1) turns out to be polarized along the **b** axis, while the luminescence is polarized to an equal degree along two directions: **b** and **c**$'$.[9] Under similar conditions, light can be generated on the resonant 0–0 transition ($\lambda_{21}=3739.3$ Å), polarized along the c' axis and directed perpendicular to this axis.[10] Since the absorption coefficient α on the 1–2 transition is large ($\alpha_{13}\approx 10^2$ cm^{-1}), the conditions for light generation on the 2–1 transition could be satisfied only in a layer $\approx 10^{-2}$ cm thick. The population of the level 2 of the remainder of the sample decreased along the propagation direction z of the λ_{21} radiation like $\exp(-\alpha_{13}z)$, where z is the running coordinate along z. These two parts of the sample are shown schematically in Fig. 1b. According to the foregoing, the absorption of light on the frequencies 0–0 transition is possible only in the case when the axes **b** in both parts of the sample make a certain averaged angle

$$\varphi=\frac{1}{n}\sum_{i}^{n}\varphi_i,$$

where n is the number of layers in the second part of the sample, and the position of the b axis of each of them is specified by the angle φ_i. The existence of these layers, their rotation relative to each other, and the possibility of introducing the average angle φ was proved by an x-ray structure analysis. Thus, the intensity of the electric field **E** of the pulse of the wavelength λ_{13} at the point z is equal to $\mathbf{E}_0\exp(-\alpha_{13}z)\cos\varphi$, where $\mathbf{E}_0$ is the intensity of the electric field of the λ_{13} pulse at the point $z\approx 0$.

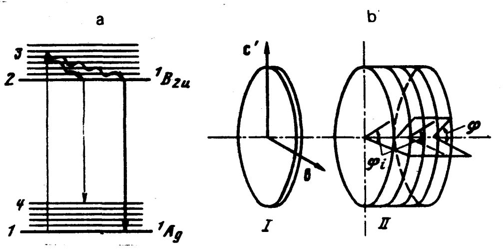

FIG. 1. Working energy levels of pyrene in diphenyl (a) and the parts of the sample (I and II) in which the generation of the stimulated radiation and the self-induced transparency develop (b); b and c' are the preferred axes of the diphenyl crystal with pyrene.

We used diphenyl single crystals with relatively high concentration of pyrene molecules ($\approx$0.10–0.15 mol.%), this being a necessary condition for the onset of stimulated emission in the sample. The crystals were grown from the melt by the Bridgman method. In the investigations we used plates of thickness 0.2 cm. The plates were placed in an optical cryostat at a temperature 1.6–4.2 K. A block diagram of the setup is shown in Fig. 2. A special unit made it possible to rotate the crystal in the cryostat about an axis that coincided with the excitation direction, and thus choose the optimal conditions for the onset of the λ_{21} generation and the SIT. To excite the crystal at the wavelength λ_{13}, we used the third harmonic of a $Y_3Al_5O_{12}$: Nd laser with pulse duration $\Delta t\approx 10$ nsec. In the experiment we investigated both the waveform of the pulse and the energy of the output radiation at two luminescence frequencies of the pyrene, corresponding to the 0–0 transition and to the fully symmetrical 1408 cm^{-1} oscillation. The system for recording the pulses and their intensity is described in detail in Ref. 11. The registration was through a monochromator by a coaxial photomultiplier, the signal from which was photographed from the screen of a high-speed oscilloscope.

We recall that a dimensionless characteristic of a laser pulse in a resonant medium is the pulse area

$$\theta(z,t)=\frac{d_{12}}{\hbar}\int_{-\infty}^{t}\varepsilon_0(z,t')\,dt', \qquad (1)$$

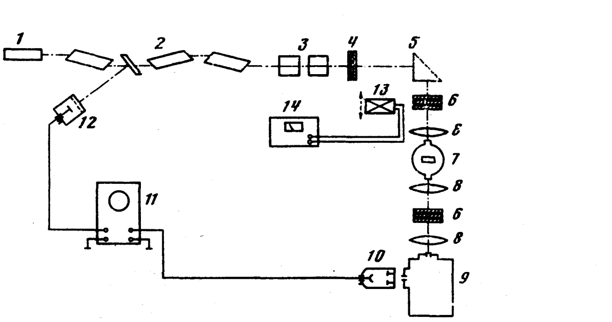

FIG. 2. Block diagram of the setup for the investigation of self-induced transparency: 1—LTIPCh-8 laser, 2—amplifiers, 3—frequency converters, 4—filters for separation of $\lambda_{13}=3530$ Å, 5—rotating prism, 6—set of neutral light filters, 7—cryostat in which the sample is placed, 8—lens, 9—ZMR-3 monochromator, 10—ÉLU-FT coaxial photomultiplier, 11—12-7 oscilloscope, 12—FÉK-09 coaxial photocell, 13—KOD-6 calorimeters, 14—microvolt-microammeter F-116/1.

where d_{12} is the modulus of the electric dipole moment of the resonant transition (1–2); $\varepsilon_0(z, t')$ is the amplitude of the electric field intensity of the light wave at the instant of time t' at the point z. In the estimates it is customary to use the following expression for the area of a rectangular pulse of duration Δt:

$$\theta = \hbar^{-1} d_{12} \varepsilon_0 \Delta t, \tag{2}$$

where $\varepsilon_0 = (8\pi W/cS)^{1/2}$; W is the laser pulse power, c is the speed of light, and S is the beam cross section area.

Denoting by θ the area of the pulse at the entry into the investigated part of the sample, we note that the SIT phenomenon is formed at $\theta_0 > \pi$. In the case when $2\pi > \theta_0 > \pi$, the formation of a stationary 2π pulse (soliton) is accompanied by an increase of the pulse duration, and at $3\pi > \theta_0 > 2\pi$ by self-compression of the pulse. The group velocity of the propagation of such a pulse is much less than the phase velocity of the light, while the delay time of the pulse in the resonant medium can be estimated from the formula

$$\Delta t_d = {}^1/_2 \alpha_{12} l \Delta t_r, \tag{3}$$

where α_{12} is the coefficient of resonant absorption, $1/\Delta t_r$ is the spectral width of the pulse, t_r is the time of the resonant absorption, and l is the sample length.

A set of oscillograms that illustrate the character of the deformation of the pulse in the diphenyl crystal with pyrene with increasing pulse power at the entrance into the sample is shown in Fig. 3a. Oscillogram *a* shows the waveform of the excitation pulse (λ_{13}), while oscillograms *b*–*h* correspond to pulses emerging from the sample at the wavelength of the 0–0 transition (λ_{21}). Oscillograms *b* and *c* demonstrate the transformation of spontaneous luminescence into stimulated emission and makes it possible to estimate the longitudinal relaxation time $T_1 \approx 10^{-7}$ sec. Pulse *c* in Fig. 3 has a small area and (as shown by investigations of samples of various lengths l) its waveform remains practically unchanged; therefore the waveform of this signal can be regarded as the waveform of the pulse at the entry into the investigated part of the sample. Its duration is $\Delta t = 8$ nsec and is commensurate with the time of transverse irreversible relaxation $T_2 = 8$ nsec,[7] so that in our experiments the condition of nonlinear coherent interaction $\Delta t \leqslant T_2, T_1$ was satisfied in our experiment.

An increase of the pulse power by one order of magnitude ($2\pi > \theta_0 > \pi$) caused an increase in the pulse duration and formation of soliton. The lengthening of the pulse in conjunction with the observed relatively large delay of the pulse ($\Delta t_d = 8$–9 nsec) is a characteristic attribute of SIT, which distinguishes this phenomenon from the saturation effect. Further increase of the power ($3\pi > \theta_0 \geqslant 2\pi$) is accompanied by a substantial deformation of the pulse in the resonant medium and its self-compression. This phenomenon is an additional confirmation of the fact that the observed kinetics of the deformation of the pulse waveform is connected with SIT. The results of a numerical analysis of the pulse deformation in the medium are shown in Fig. 3B.

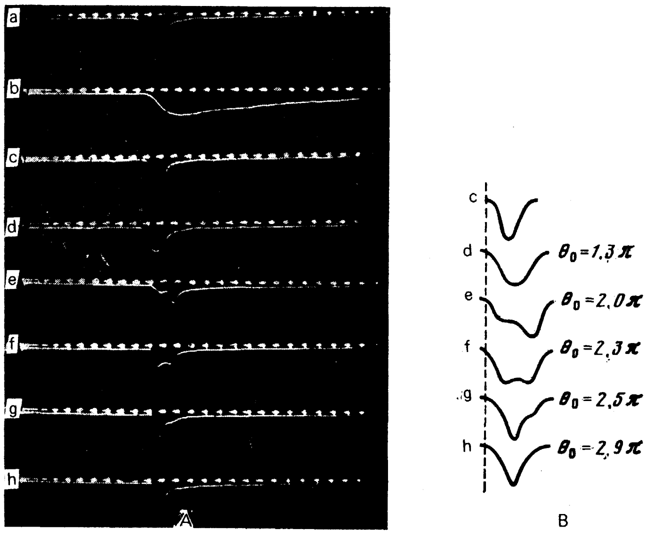

FIG. 3. *A*—oscillograms demonstrating the kinetics of deformation of the waveform of the pulse in a diphenyl crystal with pyrene (pulse duration 10 nsec, markers—10 nsec); a) excitation pulse (λ_{13}), b) pulse waveform (at wavelength λ_{21}) at a pump power $W_{13} = 3.8 \times 10^{23}$ photons/cm² sec, c) $W_{13} = 1.5 \times 10^{24}$ photons/cm² sec, d) $W_{13} = 1.0 \times 10^{25}$ photons/cm² sec, e) $W_{13} = 2.4 \times 10^{25}$ photons/cm² sec, f) $W_{13} = 3.1 \times 10^{25}$ photons/cm² sec, g) $W_{13} = 3.77 \times 10^{25}$ photons/cm² sec, h) $W_{13} = 4.8 \times 10^{25}$ photons/cm² sec. *B*—result of numerical analysis of the pulse waveform at the exit from the resonant medium.

The SIT can be realized if the diffraction losses are small. This occurs if the beam diameter is $D > 2(\lambda_{21} l)^{1/2}$. At the values $D = 0.2$ cm and $l = 0.2$ cm, in this experiment, this inequality was satisfied.

The dependence of the pulse power W_{21}^{out} at the exit from the sample on the pump pulse power W_{13}^{in} (which is proportional to W_{21}^{in}) is shown in Fig. 4. The presence of a flat section at $W_1 > 0.9$ MW characterizes the bleaching of the resonant medium and demonstrates, in conjunction with the other described singularities of the propagation of the pulse in the sample, that the SIT effect has been observed.

3. THEORETICAL ANALYSIS OF THE RESULTS

Returning to the model described at the beginning of Sec. 2, we obtain the difference between the populations of levels 2 and 1 as a function of the angle φ. The derived expression will be used as the initial condition

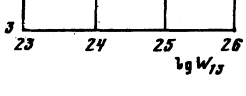

FIG. 4. Logarithmic dependence of the pulse power W_{21} on the pump pulse power W_{13}.

in the analysis of the SIT effect and for an estimate of φ. The system of kinetic equations for the level population N_n (n is the number of the level) takes the form

$$dN_n/dt=\sum_{n'}[P_{n'n}N_{n'}-P_{nn'}N_n]+\tilde{\omega}_{nn'}(N_{n'}-N_n), \tag{4}$$

where $P_{n'n}$ is the probability of an energy transition from the level $|n'\rangle$ to the level $\langle n|$,

$$\tilde{\omega}_{nn'}=\hbar^{-2}\mathscr{P}^2_{nn'}g(\omega), \quad \mathscr{P}_{nn'}=-\mathbf{d}_{nn'}\mathbf{E}_0\exp(-\alpha_{12}z)\cos\varphi.$$

$\mathbf{E}_0$ is the pulse electric field intensity, $g(\omega)$ is the normalized form function of the 32 line, and $\mathbf{d}_{nn'}$ is the electric dipole moment of the nn' transition. From the solution of this system we find that the ratio of the population difference of levels 2 and 1 (ΔN_{21}) to the total number of active particles (N) is equal to

$$n_0=\frac{\Delta N_{21}}{N}=\frac{\tilde{\omega}_{31}\tau_{31}(\tau_{21}-\tau_{32})-\tau_{31}-\tau_{32}}{\tilde{\omega}_{31}\tau_{31}(\tau_{21}+2\tau_{32})+\tau_{31}+\tau_{32}}, \tag{5}$$

where $\tau_{nn'}$ is the time of the longitudinal relaxation on the $|n\rangle\to\langle n'|$ transition. Since $\tau_{32}\approx10^{-11}-10^{-12}$ sec and $\tau_{21}\approx10^{-7}$ sec, it follows that

$$n_0\approx\frac{A\exp(-\alpha_{13}z)-1}{A\exp(-\alpha_{13}z)+1}=-\mathrm{th}\left(\frac{\alpha_{13}z}{2}+\frac{\gamma}{2}\right), \tag{6}$$

where $A=\hbar^{-2}d^2_{13}E^2_0(0)\tau_{21}\tau_{32}\cos^2\varphi$, $\gamma=-\ln A$. Obviously, the inverted population difference in the first part of the crystal is realizable at $n_0>0$, as was the case under the condition of the present experiment. The populations of levels 2 and 1 of the pyrene in the investigated part of the diphenyl crystal deviates in our case from the equilibrium value and is determined by formula (6). We write down the system of differential equations for the components of the nonequilibrium polarization (u, v, n), due to the nonlinear coherent interaction of the pulse with the pyrene molecules at the 2–1 transition frequency. In the interaction representation, these equations take the form[1]

$$du/dt=-\Delta\omega v, \quad dv/dt=\Delta\omega u+\varkappa\varepsilon(z,t)n,$$
$$dn/dt=-\varkappa\varepsilon(z,t)v, \tag{7}$$

where $\Delta\omega=\omega_{21}-\omega$, ω is the central frequency of the pulse spectrum, ω_{21} is the frequency of the resonant 2–1 transition of the molecule, $\varkappa=2d_{12}/\hbar$, $\varepsilon=E_1\sin\varphi$, E_1 is the amplitude of the electric field intensity of the pulse generated by the first part of the crystal at the instant of time t at the point z, $u=u(t,z,\Delta\omega)$, $v=v(t,z,\Delta\omega)$, $n=n(t,z,\Delta\omega)$. In the case of exact resonance ($\Delta\omega=0$) the solution of the system (7) is written in the form

$$u(t,z,0)=u_0, \quad v(t,z,0)=n_0\sin\theta(t)+v_0\cos\theta(t),$$
$$n(t,z,0)=-v_0\sin\theta(t)+n_0\cos\theta(t), \tag{8}$$

where

$$\theta=\varkappa\sin\varphi\int_{-\infty}^{t}E_1(z,t')dt',$$

u_0, v_0, and n_0 are the initial values of the polarization components.

The specific polarization of the medium **P**, which is connected with the transverse components u and v, is given by

$$\mathbf{P}(t,z)=N\mathbf{d}_{12}\int_{-\infty}^{\infty}g(\Delta\omega)d(\Delta\omega)[u\cos(\omega t-kz)-v\sin(\omega t-kz)],$$

where $g(\Delta\omega)$ is the distribution function of the dephasing parameter $\Delta\omega$, k is the wave function of the radiation of the frequency $\omega\approx\omega_{21}$. Substituting the obtained solutions (8) in Maxwell's equation, we obtain a differential equation for the pulse area:

$$\frac{d\theta(z)}{dt}=\frac{\alpha_{12}}{2}[n_0\sin\theta(z)+v_0\cos\theta(z)], \tag{9}$$

the solution of which takes the form

$$\theta(z)=2\mathrm{arctg}\left(\mathrm{tg}\frac{\theta_0+\psi_0}{2}\right)\times\exp\left\{-\frac{\alpha_{12}}{2}z(n_0^2+v_0^2)^{1/2}\right\}-\psi_0, \tag{10}$$

where $\sin\psi_0=-v_0/(n_0^2+v_0^2)$. Since τ_{32} is much less than the duration of the pump pulse λ_{13}, it follows that in the considered case $u_0=0$ and $v_0=0$. Under these conditions, Eq. (9) is transformed into

$$\frac{d\theta(z)}{dz}=-\frac{\alpha_{12}}{2}\mathrm{th}\left[\frac{\alpha_{13}z}{2}+\frac{\gamma}{2}\right]\sin\theta(z); \tag{11}$$

and its integration leads to the result

$$\theta(z)=2\mathrm{arctg}\left\{\mathrm{tg}\left(\frac{\theta_0}{2}\right)\left(\mathrm{ch}\frac{\gamma}{2}\right)^{\beta}\Big/\left[\mathrm{ch}\left(\frac{\alpha_{13}z+\gamma}{2}\right)\right]^{\beta}\right\}, \tag{12}$$

where $\beta=\alpha_{12}/\alpha_{13}$. A plot of the function (12) is shown in Fig. 5, where branch I shows the behavior of the area of the pulse under conditions of the existence of a nonequilibrium population difference, and branch II in the absence of this difference (this branch corresponds to the McCall-Hahn solution[1]). At $\theta_0<\pi$ branch I first reaches a maximum at $z=\ln A/\alpha_{13}$, and then decreases as it tends to $\theta(\infty)=0$. In the case $2\pi>\theta_0>\pi$ the branch I first decreases to the minimum point at $z=\ln A/\alpha_{13}$, and then increases and tends to $\theta(\infty)=2\pi$. The plot can be periodically continued to higher values of θ_0. Thus, the difference in the behavior of the two branches should be felt at distances z up to $\ln A/\alpha_{13}$, which under the conditions of the present experiment is much less than $l=0.2$ cm. The dependence of $\theta(z)$ reflects the possibility of SIT at $\theta_0\geqslant\pi$ in the case under consideration. The condition $\theta_0=\pi$ has made it possible to estimate the value of the angle φ, which in the investigated sample turned out to be 15°, somewhat higher than the angle obtained by x-ray structure analysis.

The use of the SIT phenomenon to obtain information

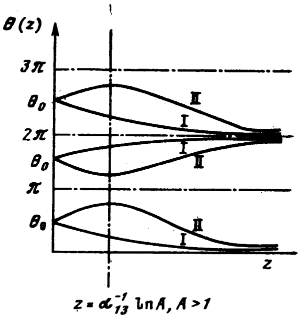

FIG. 5. Plot of the function $\theta(z)$.

on a resonant medium calls for allowance for the influence of the relaxation processes. In this case the analytic solution of the joint system of Maxwell and block equations is impossible. Therefore in the present study a computer search was made for the unknown parameters on the basis of the experimentally known kinetics of deformation of the waveform of the pulse with increasing θ_0 and the known values $T_1 = 10^{-7}$ sec and $T_2 = 8\times 10^{-9}$ sec. The analysis is based on a system of differential equation given in Ref. 2. If the field E and the polarization P are defined in the following manner:

$$\begin{aligned} E &= {}^1/_2\varepsilon(t, z)\exp\{i[\varphi(t, z)+\omega t-kz]\}+\text{c.c.},\\ P &= {}^1/_2\mathcal{P}(t, z)\exp\{i[\psi(t, z)+\omega t-kz]\}+\text{c.c.} \end{aligned} \qquad (13)$$

(where $\varphi(t, z)$ and $\psi(t, z)$ are the corresponding phases), then the foregoing system of equations takes the form[2]

$$\begin{aligned} &\frac{\partial\varepsilon}{\partial t}+\frac{c}{\eta}\frac{\partial\varepsilon}{\partial z}=\frac{2\pi\omega}{\eta^2}\mathcal{P}\sin(\psi-\varphi),\\ &\varepsilon\left(\frac{\partial\varphi}{\partial t}+\frac{c}{\eta}\frac{\partial\varphi}{\partial z}\right)=-\frac{2\pi\omega}{\eta^2}\mathcal{P}\cos(\psi-\varphi),\\ &\frac{\partial\mathcal{P}}{\partial t}+\frac{\mathcal{P}}{T_2}=\frac{d_{12}^2}{\hbar}\Delta N_{12}\varepsilon\sin(\psi-\varphi),\\ &\left[\frac{\partial\psi}{\partial t}+\Delta\omega\right]\mathcal{P}=\frac{d_{12}^2}{\hbar}\Delta N_{12}\varepsilon\cos(\psi-\varphi),\\ &\frac{\partial\Delta N_{12}}{\partial t}+\frac{1}{T_1}(\Delta N_{12}-\Delta N_{12}^0)=-\frac{1}{\hbar}\mathcal{P}\varepsilon\sin(\psi-\varphi), \end{aligned} \qquad (14)$$

where ΔN_{12}^0 is defined by expression (5) ($\Delta N_{12}^0 = n_0 N$), and η is the refractive index of the light in the sample.

In the computer search we used the universal high-level programming language PL/1 (Ref. 12). The program was produced with a computer. In the calculation we took into account the fact that with increasing pulse power the temperature increased somewhat and the relaxation time T_2 decreased. In addition, we took into consideration the exponential decrease of the population of level 2 as the pulse propagates along the z axis. The results of the calculation of the waveform of the process at different values of θ_0 are shown in Fig. 3B. The computer search has shown that the experimentally observed and calculated pulse waveforms agree at $d_{12} \approx 10^{-20}$ cgs esu. This value of d_{12} leads, if Δt_d is estimated by formula (3), to a value 13–14 nsec, which is somewhat longer than the experimentally observed delays (≈9 sec). In analogy with Ref. 6, this small discrepancy can be attributed to the non-single-mode spectrum of the pulse.

4. CONCLUSION

Thus, we have demonstrated in the present paper the possibility of using the SIT phenomenon to investigate a new class of objects–impurity molecular crystals. By a computer search we have established the value of d_{12}. For the first time ever, we have realized in the SIT method a situation wherein the crystal serves simultaneously as a source of stimulated emission and as the object of investigation. Such an approach to SIT offers advantages connected with the fact that both the radiation generator and the sample are at the same temperature (in the present study–at liquid-helium temperature). This practically eliminates the problem of attaining resonance conditions, a problem encountered with ruby.[1,6] In addition, since both the generation in the SIT develop in the same crystal, we do not encounter the difficulties due to the different optical qualities of the active element of the generator and of the investigated sample.

The existence of a nonequilibrium population of the working levels has led to modified solutions of the self-consistent problem. We have also obtained a solution [see (10)] which is valid under conditions when not only the population but also nonequilibrium polarization of the resonant medium is in disequilibrium.

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