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Self-induced transparency in two-photon resonance on an inhomogeneously broadened line

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Results are presented of an experimental and theoretical investigation of the interaction of ultrashort laser pulses with an inhomogeneously broadened two-photon resonance transition. The experiments were performed on neodymium glass. At low temperature the interaction is coherent, as manifest by a partial bleaching of the sample and by a decrease of the pulse propagation velocity; in some cases the pulse broke up into two subpulses on leaving the sample. With rising temperature, these phenomena disappeared. The results of a numerical analysis have shown that the evolution of the laser pulse as it propagates in the sample depends substantially on the Stark shift of the levels and on the width of the resonance line. The experimental results agree qualitatively with the calculations.

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1. INTRODUCTION

Processes of coherent interaction of radiation with matter, such as optical nutation, photon echo, and self-induced transparency, have been intensively investigated in the last few years (see, e.g., Ref. 1). For the interaction to be coherent it is necessary that the duration of the radiation pulse τ_p be shorter than the time or irreversible relaxation of the polarization T_2^* of the medium. These processes were customarily investigated under conditions of single-photon resonance. However, the development of two-photon resonance spectroscopy² raises the natural question of how the coherent interaction is changed under these conditions.

If the interaction is coherent, then when a pulse passes through a resonant medium self-induced transparency (SIT) sets in. This phenomenon was first predicted and experimentally investigated for one-photon resonance by McCall and Hahn.³ The possibility of observing such an effect in two-photon resonance is not self-evident. Nonetheless, if the scatter of the levels of the resonant atoms can be neglected (i.e., if the time T_2^* of reversible relaxation of the polarization is infinitely long), and if the doubled frequency of the radiation field coincides exactly with the frequency of the resonant atoms, then the effect of self-induced transparency in two-photon absorption (TPSIT) is possible, as was theoretically shown by Belenev and Poluëktov.⁴ It turned out that the additional nonlinearity introduced by the two-photon interaction leads to new consequences, such as narrowing and peak enhancement of high-energy pulses.^{5,6}

In a real experimental situation one encounters more frequently a line with inhomogeneous broadening. In

contrast to homogeneous broadening, this situation has been investigated in less detail. The particular case when the homogeneous broadenings are equal ($T_2' = T_2^* < \infty$) was considered theoretically.⁷ Under these conditions the coherent interaction can take place only when the spectral line width is less than the spectrum of the pulse (narrow line).

TPSIT was experimentally observed in media in which the excited levels were inhomogeneously broadened: in semiconductors (on interband transitions)^{7,8} and in potassium vapor (the resonance was produced here by a sum of two photons of unequal energy).⁹

Anomalously weak absorption of the pulse energy and a decrease in its propagation velocity were reported in Refs. 7 and 8. The impossibility of resolving the pulse time structure prevented the authors of these papers from investigating this phenomenon fully.

In the present paper we present the results of an experimental and theoretical investigation of TPSIT on an inhomogeneously broadened line. We investigated experimentally a sample of neodymium glass, where it is easy to realize the case of a broad line, i.e., to satisfy the condition $T_2^* < \tau_p$. Allowance for the inhomogeneous broadening greatly complicates the theoretical analysis. In this case there are no analytic solutions and computer calculations are necessary, but we did not perform them. The influence of inhomogeneous broadening manifests itself noticeably in the case of a broad line. Under these conditions, owing to the high-frequency Stark effect, the excited atoms are those whose frequency scatter exceeds the spectrum of the pulse. The damping of the induced polarization by the dephasing of the atomic radiators is accelerated, thereby significantly influencing the evolution of the pulse.

2. EXPERIMENT

In the present study the TPSIT was realized in a system where glass activated with Nd^{+3} ions was used both in the laser and as the resonantly absorbing medium.¹⁰ The radiation of the laser with frequency ω_L (the transition ${}^4F_{3/2} - {}^4I_{11/2}$) is at a two-photon resonance with the transition from the ground state ${}^4I_{9/2}$ to the ${}^4G_{7/2}(\omega_0)$ level. The absorption coefficient for the two-photon transition under discussion was measured in Ref. 11 and found to be $\gamma = 5 \times 10^{-12}$ cm/W. The inhomogeneous broadening of the ${}^4G_{7/2}$ level (which is connected with the amorphous structure of the glass matrix) amounts to $\Delta\omega_p = 4.7 \times 10^{13}$ sec⁻¹, which is larger than the detuning $\omega_0 - 2\omega_L = 10^{13}$ sec⁻¹.

To satisfy the interaction coherence condition $\tau_p < T'_2$ we used in the experiment a single ultrashort pulse (USP, $\tau_p \approx 100$ psec) from a mode-locked laser, while the investigated resonantly absorbing sample, of length 300 mm, was cooled to 4.6 K. Investigations have shown¹² that the energies of the discussed levels of Nd^{+3} in glass are not noticeably shifted when the temperature is varied in the range 4–300 K.

To increase the intensity of the laser radiation, the cryostat with the sample was placed at the center of a telescope made up of two lenses with focal length $f_1 = f_2 = 1333$ mm. The intensity was 2×10^{10} W/cm², thus excluding nonlinear scattering and self-focusing of the radiation in the glass.¹¹

The set investigations included both energy and time measurements.

The results of the energy measurements are shown in Fig. 1, which illustrates the dependence of the transmission R of neodymium glass on the intensity of the incident radiation at temperatures 300 and 4.6 K.¹¹ The measurements were made with two high-sensitivity calorimeters. The sample transmission was determined from the ratio of the energies W_0 and W_{tr} of the pulses incident on the sample and passing through it: $R = W_{tr}/W_0$.

As seen from Fig. 1 (case $T = 300$ K) at laser radiation intensities $I < 10^9$ W/cm² the losses in the neodymium glass are linear: the transmission does not depend on the intensity of the incident radiation (here $R \approx 0.9$, in good agreement with the official specifications of the sample). At higher laser intensities, the losses to two-photon absorption come into play: the transmission of the sample decreases to 0.6.

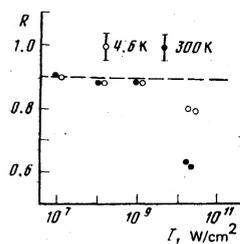


FIG. 1. Dependence of the transmission of the neodymium glass on the intensity of the incident laser radiation.

Measurements performed at 4.6 K show the following results: the linear losses remain constant ($R = 0.9$ at $I < 10^9$ W/cm²), but with increasing intensity the contribution of the nonlinear losses decreases noticeably. In this case the transmission is $R = 0.8$.

To carry out time investigations, the laser radiation was split by a glass plate into two parts, one of which (reference pulse) was directly incident on a streak camera (time resolution 3×10^{-11} sec) and the other passed first through the sample. The system was so adjusted that at room temperature of the sample both pulses were incident on the recorder simultaneously. Inasmuch as the pulses on the camera screen were spatially separated in this case, the described procedure made it possible to investigate the change of both the velocity and of the time structure of the pulse.

When the temperature was changed from 300 to 4.6 K, the maximum-intensity pulse passing through the sample was seen to be broken up into two subpulses. The most characteristic result of the photometry of the recordings of the reference pulse and of the pulse that passed through the sample is shown in Fig. 2. In this case, as shown by an estimate,¹⁰ $\theta_0 \approx 4\pi$, i. e., the 4π pulse is broken up into two subpulses.

At $T = 4.6$ K we have also observed a delay of the pulse, with a maximum value $\Delta t = 150$ psec. The delay depended on the intensity of the pulse incident on the sample and became equal to zero when the light flux was decreased by a factor of 5. A delay of 150 psec corresponds to an average pulse propagation velocity $v = 0.9c/n$, where c is the speed of light in vacuum and $n = 1.52$ is the refractive index of the neodymium glass.

By way of control, the time measurements were made with a glass sample having no Nd^{+3} ions. Neither splitting nor delay of the pulse was observed in that case.

Thus, the partial bleaching of two-photon resonant absorption, the splitting of a 4π pulse into two subpulses, and the delay of the pulse all indicate that the interaction of the radiation with the neodymium glass at 4.6 K is coherent. The threshold intensity for the onset of TPSIT is close to 10^{10} W/cm².

A rise in the sample temperature should lead to a decrease in the time T'_2 of the irreversible relaxation of the polarization, leading in final analysis to violation of the interaction coherence condition. Figure 3 shows the dependence of the maximum delay of the pulse on the sample temperature. With increasing temperature, the delay decreases, and at 60 K it is practically non-

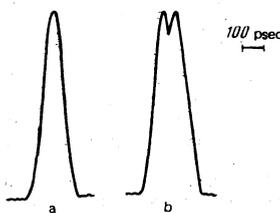


FIG. 2. Waveform of the reference pulse (a) and of the pulse passing through the neodymium glass (b).

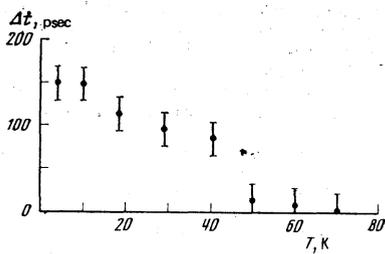


FIG. 3. Dependence of the maximum delay of the pulse on the temperature of the neodymium glass.

existent. The experimental results allow us to state that in the system under consideration $T_2^* > 10^{-10}$ sec at 4.6 K and $T_2^* < 10^{-10}$ sec at 60 K.

It is interesting to note that the nonlinear character of the onset of the pulse delay was pointed out also in Ref. 8. The difference in the degree of decrease of the pulse propagation velocity in the case of coherent bleaching in neodymium glass and in GaAs,⁸ where the decrease was by a factor of 30, is apparently due to the different absorptivities of these media.

Incomplete bleaching of the two-photon resonant absorption (this is particularly clearly pronounced in Ref. 8) at $I \geq 10^{10}$ W/cm² is apparently due to the presence of a broad inhomogeneously broadened line. This, as well as the question of how deep the modulation of the 4π pulse can be when split into subpulses, calls for a theoretical analysis of this phenomenon.

3. NUMERICAL ANALYSIS

The system of equations describing the propagation of an ultrashort pulse under conditions of two-photon resonance takes within the framework of the vector model,¹³ in dimensionless variables, the form¹⁴

$$\begin{aligned} \partial \varepsilon / \partial \xi &= i \{ \alpha \langle N - N_0 \rangle \varepsilon + \langle U \rangle \varepsilon^* \}, \\ \partial U / \partial \tau &= -i(\Delta + \Delta_{st})U + iN\varepsilon^2, \\ \partial N / \partial \tau &= -\text{Im}\{U\varepsilon^{*2}\}, \end{aligned} \quad (1)$$

where $\varepsilon = E / \max E(t, z=0)$, E is the slowly varying amplitude of the electric field, N is the population difference between the upper and lower levels, U is the slowly varying part of the off-diagonal element of the density matrix, $\Delta = (\omega_0 - 2\omega_L) / \Omega$ is the normalized frequency detuning ($\Omega = 2|q| | \max E(t, z=0) |^2$ is the Rabi two-photon precession frequency, q is the two-photon coupling coefficient), $\Delta_{st} = \alpha |\varepsilon|^2 = (a/2q) |\varepsilon|^2$, and a is the Stark coefficient. The independent variables were taken to be $\xi = z/L$ and $\tau = \Omega(t - z/v)$, where $L^{-1} = (2\pi\omega_L^2 \hbar |q| n_a / kc^2)$, n_a is the density of the resonant atoms, and v and k are respectively the velocity and the wave vector of the electromagnetic wave in the medium. The quantity L has the meaning of the nonlinear absorption length. The angle brackets in the amplitude equation denote averaging over the spread of the frequency ω_0 due to the inhomogeneous broadening of the resonant line. The initial conditions for the variables describing the medium are the following: $U=0$ and $N=1$ at $\tau=-\infty$.

The evolution of the pulse is characterized by the value of the reduced Stark coefficient α , by the inhomogeneous

relaxation time T_2^* , and by a function $E(t, z=0)$ that describes the envelope of the pulse of duration τ_p at the entrance into the resonant medium. In the performed computer experiment, the input pulse had a Gaussian profile with no frequency modulation, and its amplitude was chosen such that the parameter θ_0 , defined by

$$\theta_0 = (1 + \alpha^2)^{1/2} \int_{-\infty}^{\infty} e^{\xi} (\xi=0, \tau) d\tau,$$

was equal approximately to π , 3π , and 5π . The reduced Stark coefficient α was assumed to be 1 and 0.1. The contour of the resonant line was assumed Gaussian with a width equal to $2/T_2^*$. The cases of a narrow line ($\tau_p/T_2^* \approx 0.2$) and a broad line ($\tau_p/T_2^* \approx 10$) were investigated.

The choice of the independent variables connected with the coordinate system moving with the velocity of the initial pulse makes it possible to reduce the system of nonlinear partial differential equations (1) to a system of ordinary equations with separable variables, and to use a numerical method suitable for the latter. It turns out that the Runge-Kutta method yields a satisfactory accuracy (of the order of 1%) up to a propagation depth equal to two nonlinear absorption lengths, after which the error begins to increase noticeably. A more economical prediction method, namely a three-point correction method with the same step as in the Runge-Kutta method, was unstable. Decreasing the integration step increases the computer operation time but does not yield a considerable improvement in the accuracy, nor does it make it possible to go to depths exceeding one absorption length. Preference was therefore given to the Runge-Kutta method.

It must be emphasized that although the investigated problem was already considered by others,⁵⁻⁷ we present here a combined account of the Stark effect and inhomogeneous broadening (in contrast to Refs. 5 and 6), and we consider here the limiting case of substantially differing polarization relaxation times, namely $T_2^* < T_1^*$, whereas in Ref. 7 these times were equal. Allowances for the incoherent processes that lead to damping of the total polarization and to the population difference⁶ reduces to introducing the corresponding relaxation terms into the equation for the variables U and N in (1). Calculations performed at $T_2^* = 2T_1 = 400\tau_p$ and $T_2^* = 2T_1 = 50\tau_p$ (T_1 is the characteristic time of the energy relaxation in the medium) did not lead to a noticeable difference in the results. Therefore in all the subsequent calculations these times were assumed to be infinite.

The results of a numerical analysis for a narrow inhomogeneously broadened line recall qualitatively those obtained at $T_2^* = \infty$.⁵ There were observed damping of the weak pulse, formation of a sharp peak at the center of the 3π pulse, and the splitting of the 5π pulse into two narrow subpulses [Fig. 4(a)]. The depth of the dip between these subpulses was approximately 0.8 of the maximum pulse amplitude. Depending on θ_0 , the difference of the populations N at each depth of the medium during the time of action of the excitation executed

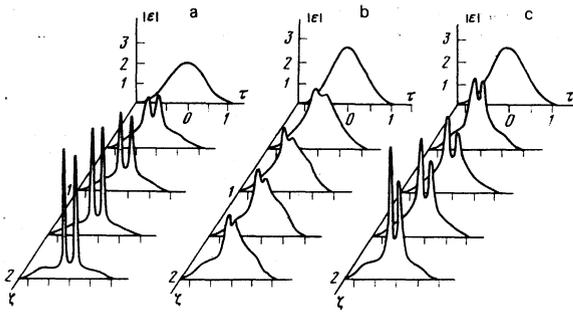


FIG. 4. Evolution of the 5π pulse in the case: a) $\tau_p/T_2^* = 0.2$, $\alpha = 1$; b) $\tau_p/T_2^* = 10$, $\alpha = 1$; c) $\tau_p/T_2^* = 10$, $\alpha = 0.1$.

a definite number of oscillations, from a maximum $N_{\max} = 1$ to a minimum $N_{\min} \approx (\alpha^2 - 1)/(\alpha^2 + 1)$.¹⁴ Although the population difference did not return to its initial value when the action of the pulse was stopped, the deviation from $N = 1$ was negligible $N(\xi, \tau \rightarrow \infty) > 0.8$, thus indicating that the absorbing medium returns energy to the pulse. The 3π pulse experienced a delay $\Delta t/\tau_p \approx 0.1$. The described phenomena did not depend on the value of α .

In the case of a wide inhomogeneously broadened line ($\tau_p/T_2^* \approx 10$), the propagation of the 3π and 5π pulses depended substantially on the reduced Stark coefficient. At $\alpha = 1$, a sharp peak was produced on the envelope of the 3π pulse, as was the case in the preceding calculations. Although a splitting of the 5π pulse does take place [Fig. 4(b)], the produced dip is approximately 0.1 of the maximum wave amplitude. Just as before, the USP is delayed and its magnitude increases to $\Delta t/\tau_p \approx 0.3$. In the case of excitation by a 5π pulse, the change of the population with time was a periodic and with increasing propagation depth of the USP it tended to a monotonically decreasing dependence on τ . Thus, the fraction of the pulse energy that remains in the medium is larger than for the case of a narrow line.

On the other hand, at $\alpha = 0.1$ the picture of the propagation of the ultrashort pulse is similar to that occurring in the case of small inhomogeneous broadening: a high peak is produced at the center of the 3π pulse and a relatively narrow dip is produced when the 5π pulse is split. The delay of the 3π pulse was $\Delta t/\tau_p \approx 0.1 - 0.15$.

Regardless of the value of the inhomogeneous broadening and of the reduced Stark coefficient, the π pulse attenuated and showed no delay.

Both in the case of the inhomogeneously broadened line and for a broad line, the transmission coefficient determined in Sec. 2 of this article depends on the energy of the initial pulse. Thus, for example, after passing over two absorption lengths at $\tau_p/T_2^* = 0.2$ and $\alpha = 1$, its value was 0.4 for the π pulse, 0.8 for the 3π pulse, and 0.9 for the 5π pulse. Under the same conditions but for a broad line, the transmission coefficient was smaller (thus, $R \approx 0.85$ for the 5π pulse).

We present a qualitative explanation of the obtained numerical results. The observed difference between the

evolution of the USP when propagating in a broad line and in a narrow line (Fig. 4) was accompanied by additional energy losses of the pulse in the former case. Since the relaxation times T_1 and T_2^* were assumed to be infinite in the calculations, these losses may be connected with the damping of the macroscopic polarization as a result of the dephasing of the radiators. The possibility of dephasing is due to the presence of the inhomogeneously broadened line. In the case $\tau_p \ll T_2^*$ the spectrum of the pulse exceeds the line widths, therefore all the radiators are excited independently of the Stark shift. The dephasing, which occurs within a time equal to the reciprocal value of the spectrum of the excited radiators (i.e., in this case after a time T_2^*) is negligible during the time of action of the USP. By the same token, the macroscopic polarization of the medium does not manage to relax in the time τ_p , and coherent return of energy from the medium to the pulse is possible. On the other hand, if the spectrum of the pulse is smaller than the line width ($\tau_p \gg T_2^*$ —broad line), what is excited is a packet of radiators with a width that spans the spectrum of the pulse $1/\tau_p$ plus the Stark line shifts $\Delta\omega_{st}$, i.e., $1/\tau_p + \Delta\omega_{st}$. The dephasing of the excited emitters takes place within a time $(1/\tau_p + \Delta\omega_{st})^{-1}$, and if $\Delta\omega_{st} > 1/\tau_p$, then the macroscopic polarization attenuates before the end of the action of the pulse.

It is seen that the Stark shift influences substantially the evolution of the USP in the inhomogeneously broadened medium. Loss of coherence can be observed only when the spectral width of the pulse is less than the inhomogeneous line width, while the Stark shift and the duration of the pulse are connected by the relation $\tau_p \Delta\omega_{st} \geq 1$.

On the other hand, when the level shift in the field of the pulse is much less than the spectral width of the USP, $\Delta\omega_{st} \ll 1/\tau_p$, the coherent propagation will take place in analogy with the case of a narrow line.

It should be noted that the Stark dephasing due to the inhomogeneous broadening can be the factor that limits the growth of the peaks of the subpulses. The growth of the intensity at the maximum of the subpulse, as it passes through the medium, leads to a growth of the Stark shift, even if the reduced Stark coefficient is small.

4. DISCUSSION

1. The most pronounced demonstration of coherent interaction of USP in two-photon resonance is the splitting of the input pulse into two subpulses, as shown on the photoelectronic recording (Fig. 2).

A numerical calculation of the evolution of a Gaussian pulse shows that under conditions close to the experimental ones the pulse likewise splits into two subpulses. As seen from Fig. 4, the time interval between the subpulses does not exceed the initial pulse duration, in good agreement with experiment.

It must be emphasized that in the case of a broad inhomogeneously broadened line the depth of modulation of the pulse when it breaks down depends substantially

on the value of the Stark shift of the levels. Unfortunately, we do not know the Stark coefficient a of the investigated level of the neodymium ion Nd^{3+} in the glass. A comparison of the theoretical results (Fig. 4) with the experimentally observed breakdown indicates that the reduced Stark coefficient is $\alpha = a/2q \approx 0.5$. Using the value of the two-photon absorption in neodymium glass¹¹ we can estimate the order of magnitude of q , which turns out to be $\sim 10^2$ cgs esu, so that $a \sim 10^2$ cgs esu.

2. In an experiment performed at USP intensities $I \geq 10^{10}$ W/cm², a delay of the pulse in the sample was observed, by a time $\Delta t \approx 1.5\tau_p$. A numerical calculation also demonstrates the effect of the pulse delay ($\Delta t \leq 0.3\tau_p$) and, just as in the experiment, its appearance has a threshold character, i. e., it occurs at $\theta_0 \geq 2\pi$. The quantitative difference in the value of the delay can be connected with the unequal shape of the input pulse in the calculations and in the experiment.

3. *Partial bleaching* of two-photon absorption in neodymium glass was observed in the experiment.

A numerical calculation also shows that the USP undergoes *anomalously low energy losses*, but somewhat less than in the experiment, this apparently being due to the additional losses on the inhomogeneities and defects in the glass.

4. We investigated experimentally the dependence of the pulse delay on the sample temperature (Fig. 3). This made it possible to estimate the characteristic times of irreversible relaxation of the polarization T_2' in the neodymium glass. The nature of the irreversible relaxation in this temperature region is known.¹⁵ In principle, the temperature dependence can be taken into account by introducing an additional relaxation term into the system of equations (1). In the numerical analysis, however, we confined ourselves to the case $\tau_p \ll T_2'$, by the same token concentrating our attention on the role of the inhomogeneous broadening in the TPSIT.

Thus, the physics of coherent interaction is determined essentially by the rephasing and dephasing of the radiators. The use of photon echo, which was recently realized in experiment under conditions of two-photon resonance,¹⁶ would be quite promising here.

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