Self-transparency in gallium arsenide on two-photon interaction with an ultrashort light pulse

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Resonant two-photon interaction between a single picosecond light pulse from a neodymium laser and a GaAs sample is investigated experimentally. At 77°K in the semiconductor the interaction is coherent. Absorption in a 0.2 cm thick crystal is virtually absent for input pulse intensities between 0.05 and 2 GW/cm^2. At 2 GW/cm^2 the velocity of light in the crystal was approximately 30 times smaller than the usual value. The coherent interaction effects completely disappears at room temperature and ordinary two-photon light absorption is observed. Threshold energy densities of the exciting radiation for which destruction of the sample occurs are determined. At nitrogen temperature the threshold is greater than at room temperature by two orders of magnitude.

1. INTRODUCTION

Problems of coherent interaction of laser emission with a resonantly absorbing medium are currently attracting interest. Such an interaction takes place when the duration of the light pulse is less than the transverse relaxation time. The interaction has the remarkable property of bringing about, under certain circumstances, the phenomenon of the so-called "self-induced transparency" when a pulse of electromagnetic radiation passes through the medium practically without attenuation, and the group velocity is lower by more than an order of magnitude than the usual velocity.

In experiments on this effect the resonant medium used so far was either a ruby crystal[1] or a gas[2-4], and the resonant interaction between the emission and matter was of the one-photon type (only one photon is absorbed or emitted in each elementary act). The features of the two-photon interaction of a single ultrashort (picosecond) light pulse (USP) of a neodymium laser with a GaAs semiconductor[1)] were reported for the first time in[5]. Pulse delay in the sample was observed, and the absorption was many times smaller than the two-photon absorption which occurs when GaAs is illuminated by neodymium laser pulses in the nanosecond range[6].

The present paper presents the results of USP-GaAs interaction studies. The main emphasis is on phenomena typical of self-induced transparency. Use of a ultra-high-speed camera in this research made it possible to estimate the duration of the light pulse before and after its passing through the sample. This allowed us to measure the dependency of the intensity of light transmitted through a semiconductor crystal on its intensity at the entry point, to determine the delay associated with the passage through the GaAs, and also to estimate threshold intensities at which the surface of the semiconductor is damaged and their dependence on temperature.

As will be shown below, the results of the experiments prove the correctness of the hypothesis advanced in[5] that USP-GaAs interaction is coherent.

2. MAIN RESULTS OF THE TWO-PHOTON SELF-TRANSPARENCY THEORY

Let a pulse of coherent emission of duration τ act on a two-level medium in which resonant two-photon absorption is possible. If the time of phase coherence is $T_2 > \tau$, and the light intensity is sufficiently high, then coherent effects of light-medium interaction can be observed in such a medium. In the theoretical paper[7], an equation was derived which describes such an interaction and holds true for gases with two nondegenerated energy levels:

$$d\vartheta / dz = -\alpha(1 - \cos\vartheta). \tag{1}$$

Here z is a coordinate plotted along the lines of light propagation;

$$\vartheta(z) = \frac{|r_{cv}|}{2\hbar}\int_{-\infty}^{\infty} \mathscr{E}^2(z,t)\,dt,$$

where $\mathscr{E}$ is the amplitude of the electric field intensity ($\mathscr{E}^2 \sim I/v$, where I is the intensity of the light and v is the propagation velocity); α is the coefficient of nonresonant losses, and r_{cv} depends on the composite matrix element of the transition and the frequency of the exciting radiation[2)]. It can be shown that the solution of Eq. (1) from[7] is of the form

$$\operatorname{ctg}\frac{\vartheta}{2} = \operatorname{ctg}\frac{\vartheta_0}{2} + \alpha z, \tag{2}$$

where $\vartheta_0 = \vartheta(0)$. By selecting a sample of a given length, i.e., by fixing αz, one can plot the dependence of ϑ on ϑ_0. This plot is of the periodic "stepped" type (Fig. 1). The smaller the value of αz, the less pronounced the "steps" and the more the curve approaches the straight line $\vartheta = \vartheta_0$ of complete transparency.

Comparing Eq. (2) with the solution

$$\operatorname{ctg}\frac{\vartheta}{2} = \operatorname{ctg}\frac{\vartheta_0}{2}\exp\left(\frac{1}{2}\alpha' z\right)$$

of the corresponding equation for one-photon self-transparency[1], we see that in two-photon self-transparency the stationary pulse is produced at a much larger distance and the transient process fills the greater part of the medium if, of course, $\alpha = \alpha'$. (Note that in the one-photon case ϑ is proportional to the amplitude $\mathscr{E}$ of the light intensity and in the two-photon case $\vartheta \propto \mathscr{E}^2 \propto I$.) The produced stationary pulse must propagate with a velocity significantly lower than v/r, where v is the velocity of light in vacuum and r is the refractive index[7].

From (2) it also follows that pulses with large initial intensity ($2\pi > \vartheta_0 > 0$) must traverse a substantially greater length in order to attain a given value of ϑ. A pulse $\vartheta_0 \approx 2\pi$ will pass through the entire sample

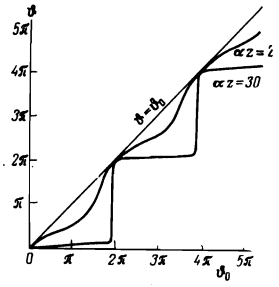

FIG. 1. Theoretical relation $\vartheta = f(\vartheta_0)$ for various values of $z\alpha$.

practically without attenuation, and since its speed is minimal, it will be the last to leave the medium. The rather strong dependence of ϑ on ϑ_0 (see Fig. 1) can cause pulses that are close in intensity at the entry point to differ appreciably at the exit point. Therefore, if the distance required for the formation of a stationary pulse comprises a significant part of the sample, then a strong scatter of the light delay time in the sample will be observed in the experiment.

An equation describing two-photon coherent interaction of light with a semiconductor was obtained for ϑ in[8]. For large values of ϑ this equation takes the form $d\vartheta/dz = -\alpha$, i.e., two-photon absorption does not depend on intensity. As noted in[8], such saturation of the two-photon absorption is a consequence of coherent interaction of light with the medium. In other words, if the light intensity is so great that $\vartheta_0 \gg \alpha z$, then the light passes through the substance practically without absorption.

The time T_2 of the relaxation of polarization in GaAs at 77°K is 10^{-11} sec[9,10]. This means that coherent interaction with USP of duration $\tau \sim 10^{-11}$ sec can occur only at T = 77°K, since $\tau \lesssim T_2$ only in this case. At temperatures higher than that of liquid nitrogen the coherent effects must vanish with increasing temperature, because $T_2 \ll \tau$.

3. THE APPARATUS

The block diagram of the apparatus is shown in Fig. 2. A pulse of energy 4×10^{-3} J was separated with the aid of a Kerr cell from a series of ultrashort pulses of a neodymium laser operating under self-mode-locking conditions.[11]. This pulse passed through a saturating filter, and was then amplified to 8×10^{-2} J by two neodymium-glass rods. A spherical lens L_1, of a focal length of 165 mm focused the light on an n-GaAs sample with carrier density 10^{17} cm^{-3} and mobility 4200 cm^2/sec · V. The crystal could be either at liquid-nitrogen (T = 77°K) or room temperature. The emission intensity was regulated by moving the sample between confocal lenses, and the illumination area was determined photographically. The energy pulse was measured with calorimeters C_1 and C_2 of sensitivity 40 and 25 erg, respectively. The energy measurement error did not exceed 20%. The use of a photochronograph (FÉR-2) made it possible to estimate the upper limit of the duration of the light pulses at the point of entry to and exit from the GaAs, and to observe the time delay of the USP passing through the semiconductor.

The signals that followed the paths ADE and ABC (without the sample) arrived at the FÉR-2 simultaneously to within 2×10^{-11} sec. The upper part of the slit, 20 μ wide and located at the entrance of the photo-

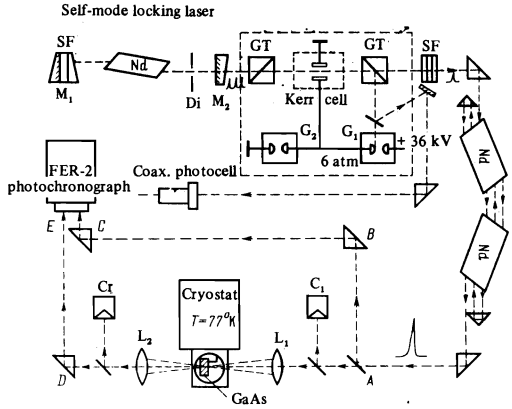

FIG. 2. Block diagram of apparatus: M_1 and M_2—mirrors with reflection coefficients 1.0 and 0.6 at $\lambda = 1.06$ μ, SF—saturating filters, Di—diaphragm, GT—analyzer and polarizer using Glan-Thompson prisms, G_1 and G_2—discharge gaps and C_1 and C_2—calorimeters, L_1 and L_2—confocal spherical lenses with focal lengths 165 and 155 mm, respectively.

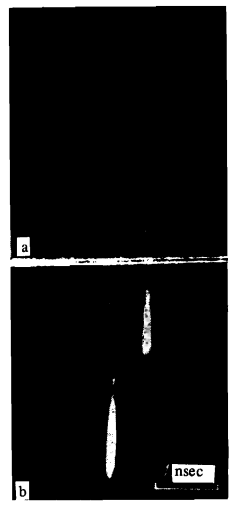

FIG. 3. Photochronograms of light pulses: a—coinciding pulses (without a sample in ADE (Fig. 2)); b—the pulse transmitted through the sample (upper) is delayed relative to the reference pulse (lower).

chronograph perpendicular to the image scan direction, was illuminated by a pulse which had followed the path ADE, while the lower part of the slit was illuminated by a pulse which had followed the path ABC, so that in the absence of a sample the images of both parts of the slit fell on the same straight line. Moreover, an unilluminated segment remained between them on the screen. The shifting of the upper part of the image to the right relative to the lower part when a sample was present testified to the delay of the USP transmitted through the sample. The measurement error of the delay time was ±30% because of the nonlinearity of the photochronograph scan.

4. EXPERIMENTAL RESULTS

Figure 3 shows photochronograms taken with the aid of the FÉR-2. Reduction of the photographs has shown that pulse duration at the base under self-mode-locking conditions does not exceed 2×10^{-11} sec (resolution time of the apparatus). It is known from[12] that such pulses can be of several picosecond duration. Henceforth, for the sake of specificity, we assume everywhere that $\tau = 10^{-11}$ sec.

The velocity v of the light in the medium can be estimated from the formula[5] $v = c\,(1 + c\delta t/l)^{-1}$, where δt is delay time of the USP in a crystal whose thickness is l. The maximum delay time δt for a sample 0.2 cm

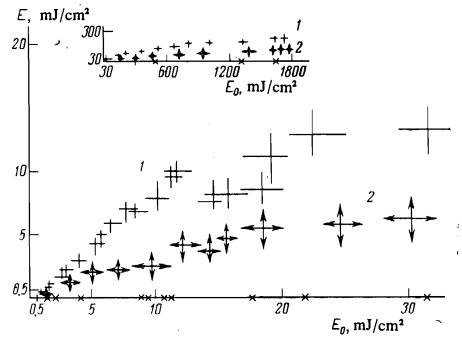

FIG. 4. Output energy E vs input energy E_0 at various sample thicknesses l: 1) l = 0.2 cm, 2) l = 0.8 cm. At room termpature the sample with l = 0.2 cm is opaque (crosses on the abscissa axis).

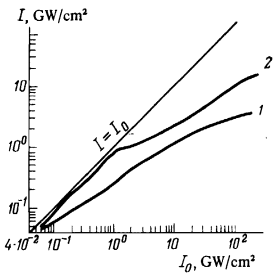

FIG. 5. Output intensity I of a sample vs. input intensity I_0. The light pulse duration is assumed to equal 10^{-11} sec. 1 – l = 0.8 cm; 2 – l = 0.2 cm.

thick reached 0.6 nsec at an incident-light intensity of 1–3 GW/cm^2. This means that the propagation velocity of solitary USP decreased down to 3.3×10^8 cm/sec from the expected value $c/n \approx 9 \times 10^9$ cm/sec, where n = 3.3 is the refractive index at $\lambda = 1.06\ \mu$. At input intensities lower and higher than 1–3 GW/cm^2, the delay rapidly diminished.

Figure 4 shows the experimental dependence of the output energy density E on the input energy E_0 for crystals 0.2 cm (crosses) and 0.8 cm (crosses with arrows) thick at 77°K. The experimental points take into account the reflection losses at both the front and rear end faces of the sample (30% for each face). As can be seen from Fig. 4, a sample 0.2 cm thick is almost completely transparent when the input energy density ranges from 5×10^{-4} J/cm^2 to 8×10^{-3} J/cm^2, but then the curve has several bends at E_0 ranging from 8×10^{-3} J/cm^2 to 2 J/cm^2. The output energy changes density slightly with further increase of the input energy density. It can be seen from plot 2 that the absorption increases several fold with increasing sample thickness. Plots 1 and 2 demonstrate that the coefficient of two-photon absorption $K^{(2)}$ is three orders of magnitude less than the value received for GaAs[6] with nanosecond neodymium-laser pulses.

Figure 5 shows plots of $I = f(I_0)$ for samples at liquid nitrogen temperature. At $I_0 > 2$ GW/cm^2 the theory fails to account for the behavior of the curves: they deviate from the straight line corresponding to total transparency but, as before, the two-photon absorption coefficient remains almost three orders of magnitude less than the one obtained in the experiment with nanosecond pulses[6].

The transparency of GaAs diminishes sharply at room temperature. Results of the corresponding measurements are shown by crosses on the abscissa axis in Fig. 4. This indicates that at room temperature the usual two-photon absorption takes place, as expected, since $T_2 \ll \tau$ at $T \approx 300$°K, and no coherent interaction between the radiation and the medium should take place.

It is noted in[13] that damage is due to the concentration of light energy $KI\tau$, where K is the absorption coefficient (in cm^{-1}). Therefore, by decreasing the duration τ, it is possible to increase the pump intensity without damaging the semiconductor. In the two-photon case $K^{(2)} = bI$, and the damage is determined by the quantity $bI^2\tau$. Thus, for two-photon absorption, the threshold intensities $I_{thr}^{(1)}$ and $I_{thr}^{(2)}$, which correspond to durations τ_1 and τ_2 satisfy the relation

$$I_n^{(1)}/I_n^{(2)} = (\tau_2/\tau_1)^{1/2}. \qquad (3)$$

At intensity levels of 20 MW/cm^2, a pulse of 30 nsec duration damaged the GaAs cooled to 77°K. It follows from (3) that at $\tau = 10^{-11}$ sec the damage threshold will increase to I = 1.5 GW/cm^2. Experiments have shown that at room temperature the front face of the GaAs sample was damaged at a USP intensity $I_{thr} \sim 1$ GW/cm^2. This is in accord with the estimate given above. The value of I_{thr} for a sample cooled to 77°K was significantly greater, about 200 GW/cm^2. The increase of I concomitant with the decrease in temperature indicates that in the case of cooled GaAs, the USP energy does not have time to become transferred to the lattice. It should be noted that when a sample is acted upon by a series of picosecond pulses, the damage threshold is several times lower. In the case of a cooled crystal this is apparently due to absorption of "background" pulses that heat the crystal.

CONCLUSION

The experiments have shown that the following phenomena, characteristic of coherent interaction of light with a substance, occur when an individual USLP passes through GaAs at 77°K.

1. A time delay of the USLP, which depends on its intensity I_0. This delay reaches its maximum at 2 GW/cm^2 and corresponds to a light propagation velocity in GaAs $v = 3.3 \times 10^8$ cm/sec (as against $v = c/n = 9 \times 10^9$ cm/sec, where n = 3.3 is the refractive index of the GaAs at 1.06 μ wavelength).

2. In the USLP intensity range 0.05 GW/cm^2 < I_0 < 2 GW/cm^2, GaAs 0.2 cm thick absorbs practically no light. At $I_0 > 2$ GW/cm^2, absorption does take place, but however, the absorption coefficient $K^{(2)} = bI$ is roughly three orders of magnitude less than the corresponding value of $K^{(2)}$ for nanosecond pulses[6].

3. Anomalously low two-photon absorption is accompanied by a significant rise in the damage threshold (from 1 GW/cm^2 at T = 300°K to 200 GW/cm^2 at T = 77°K).

The above phenomena depend on temperature. In particular, the two-photon absorption observed at 300°K is close to the usual one[6].

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[1)]The energy of a photon of an Nd-laser is $\hbar\omega$ = 1.17 eV; the width of the forbidden band of the GaAs is Δ = 1.49 eV at a temperature of 77°K, i.e. $2\hbar\omega > \Delta > \hbar\omega$.

[2)]In the case of a 2π pulse, $r_{CV}(\text{cm}^3) \approx 10^{-21}/(\text{cm}^2)\tau$ (psec).

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