Self-induced transparency of a semiconductor

F. Brückner, Ya. T. Vasilev, V. S. Dneprovskiĭ, D. G. Koshchug, E. K. Silina, and V. U. Khattatov

Moscow State University
(Submitted July 14, 1974)
Zh. Eksp. Teor. Fiz. **67**, 2219–2226 (December 1974)

An investigation was made of the phenomenon of self-induced transparency in semiconductors observed in one-photon interband transitions and in resonance excitation of excitons by ultrashort second-harmonic light pulses emitted by a mode-locked neodymium laser. An analysis was made of the influence of an external electric field and temperature of the semiconductor samples on the self-induced transparency. The optical probing method was used in measurements of the transverse and longitudinal intraband relaxation times and of exciton relaxation times in CdS_xSe_{1-x} crystals.

1. INTRODUCTION

Recently, there have been several theoretical investigations of the coherent interaction of high-power ultrashort light pulses with semiconductors in the case when the pulse duration is less than the transverse relaxation time ($\tau < T_2$) and when the area under a laser pulse is

$$\theta = \frac{\mu}{\hbar}\int_{-\infty}^{\infty} E\,dt > \pi,$$

where E is the slow amplitude of the field of the light pulse on entry into a sample and μ is the dipole matrix element of the transition involved. One-photon[1] and two-photon[2,3] interband transitions have been analyzed and a study has been made of the self-induced transparency due to one-photon resonance excitation of excitons.[4,5] The self-induced transparency in semiconductors was first discovered experimentally in the two-photon excitation of GaAs by ultrashort pulses emitted from a mode-locked neodymium laser.[6,7] Preliminary reports of the observation of the self-induced transparency in semi-conductors of the CdS_xSe_{1-x} type due to one-photon interband transitions and due to resonance excitation of excitons were published earlier.[8,9]

The self-induced transparency represents a considerable reduction in the absorption of ultrashort light pulses traveling at group velocities an order of magnitude lower than the velocity of light in a given semiconductor. A light pulse excites coherently electron-hole pairs (or excitons) as a result of the interband absorption and it loses its energy at the leading edge; this energy then returns to the trailing edge because of the stimulated re-emission of the medium. The condition $\tau < T_2$ imposes certain additional limitations on the relationship between the forbidden band width of the semiconductor E_g and the energy of the exciting photons $h\nu$ in the interband transition case ($h\nu - E_g < E_0$, where E_0 is the energy of an optical phonon), because T_2 depends on the kinetic energy of the created electrons and falls with rising energy due to the interaction with optical phonons.[1]

The present paper describes an experimental investigation of the self-induced transparency which appeared in a semiconducting single crystal of $CdS_{0.75}Se_{0.25}$ due to one-photon interband transitions and due to resonance excitation of excitons. The semiconductor was illuminated with a train of second-harmonic ultrashort pulses ($h\nu$ = 2.34 eV) emitted by a mode-locked neodymium laser (Fig. 1). The pulse duration ($\tau \lesssim 2 \times 10^{-11}$ sec)

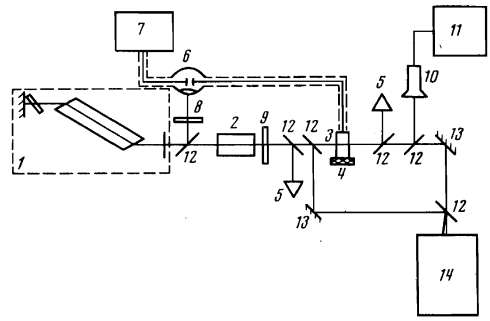

FIG. 1. Schematic diagram of the apparatus: 1) mode-locked neodymium laser; 2) KDP crystal; 3) investigated sample of $CdS_{0.75}Se_{0.25}$; 4) heat sink in a cryostat; 5) calorimeter; 6) nitrogen discharger; 7) high-voltage rectifier; 8) neutral light filters; 9) SZS-22 light filter; 10– coaxial photocell; 11) I2-7 oscillograph; 12) glass plates; 13) mirrors; 14) FÉR-2 image-converter streak camera.

was determined with an FÉR-2 image-converter streak camera whose time resolution was 2×10^{-11} sec. The collision method measurements in the case of two-photon absorption in a semiconducting single crystal of ZnS (E_g = 3.6 eV, T = 300°K) made it possible to estimate the average duration of a pulse in a train of ultrashort pulses ($\tau \approx 5 \times 10^{-12}$ sec).[10] The pulse power density in an unfocused beam could reach 10^9 W/cm.

2. SELF-INDUCED TRANSPARENCY DUE TO INTERBAND TRANSITIONS

The interband self-induced transparency was observed using $CdS_{0.75}Se_{0.25}$ single crystals cooled to 130°K. At this temperature a crystal grown from the gaseous phase had the forbidden band width $E_g \approx 2.335$ eV. Thus, $h\nu - E_g \approx 5$ meV $< E_{TA}$, where E_{TA} is the energy of a transverse acoustic phonon. The giant power and short pulse duration of the selected source and the cooling of a crystal characterized by $E_g \approx h\nu$ were necessary in order to satisfy the conditions for the observation of the self-induced transparency. This transparency did not appear in cooled $CdS_{0.7}Se_{0.3}$ crystals characterized by $h\nu - E_g > E_0$.

In the experimental investigation the attention was concentrated on the phenomena most typical of the self-induced transparency. Figure 2a shows the dependence of the transmission of a crystal ($G \equiv W/W_0$, where W_0 and W are the energies of the incident and transmitted radiation) on W_0 in the case of illumination with a train of ultrashort pulses. The characteristic strong increase in the transparency amounted to more than two

Copyright © 1975 American Institute of Physics

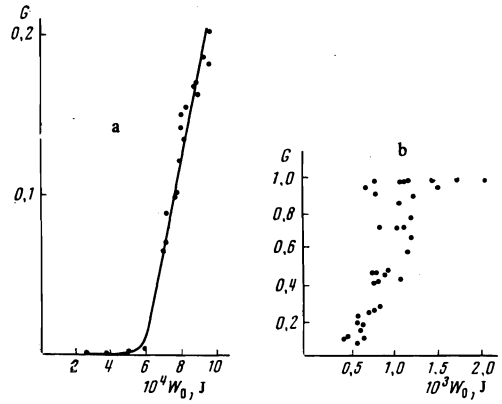

FIG. 2. Dependences of the transmission of a crystal on the energy of incident radiation: a) interband transitions; b) resonance excitation of excitons.

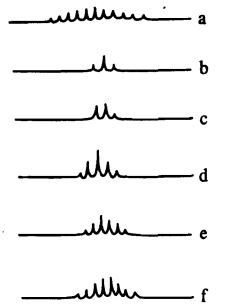

FIG. 3

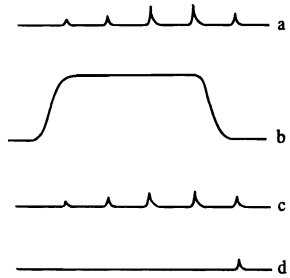

FIG. 4

FIG. 3. Oscillograms of ultrashort light pulses incident on a crystal (a) and transmitted by it: b), c) interband transitions; d) interband transitions caused by focused radiation; e), f) resonance excitation of excitons for $W_0 = 10^{-3}$ J and $W_0 = 1.5 \times 10^{-3}$ J.

FIG. 4. Influence of an external electric field $\mathscr{E}$ on the self-induced transparency: a) oscillogram of ultrashort light pulses at the exit from a sample for $\mathscr{E} = 0$; b) shape of an electric field pulse; c) $\mathscr{E} = 3 \times 10^3$ V/cm; d) $\mathscr{E} = 1.2 \times 10^4$ V/cm.

orders of magnitude and as a result of this increase, G reached 0.2. For the sake of comparison, it should be pointed out that the transmission measured with a DFS-12 double monochromator was $G < 10^{-3}$ (continuous illumination with $\lambda = 0.53\ \mu$). When the source of picosecond pulses was replaced with a laser working under Q-switched conditions ($\tau = 3 \times 10^{-8}$ sec), no transmission was observed even when the incident power was raised up to the damage threshold. The radiation transmitted by a crystal was detected with a special coaxial photocell,[1)] which was matched to an I2-7 oscillograph. The time resolution of the recording system was ~1 nsec. Typical oscillograms of the incident and transmitted radiation are shown in Figs. 3a-3d. These oscillograms show clearly the change in the envelope of a train of pulses transmitted by a crystal (one or two pulses were selected). The transmission was highest for the shortest and strongest pulses (the duration and intensity varied from pulse to pulse in a train[11]). An increase in the density of the incident radiation by focusing raised the amplitudes of the transmitted pulses and gave rise to additional pulses in that part of the train where the duration was shortest.[11] Then, the transmission for isolated ultrashort pulses became $\gtrsim 0.6$.

The self-induced transparency was characterized by an anomalously low velocity of radiation in the crystals. Figure 1 shows the system used to measure the delay of a pulse transmitted by a crystal, relative to a reference ultrashort pulse. The measurements were carried out using the FÉR-2 streak camera, which was used in the fast-scanning regime (in this case the streak records showed only the axial radiation period). The number of pulses per axial period and their relative intensities varied from flash to flash The recorded delay was $\delta \approx 0.15$–0.2 nsec for the strongest pulses. The measured value of δ allowed us to estimate the velocity of light in the investigated semiconductor from the formula $V = c(1 + c\delta/l)^{-1}$, where $l = 4$ mm is the length of the crystal. We found that $\delta = 0.15$ nsec corresponded to $V = 2.5 \times 10^9$ cm/sec.

The next series of experiments was intended to determine the influence of an external electric field on the self-induced transparency. A high-voltage pulse of ~50 nsec duration was formed using a coaxial line section charged to a voltage of 10 kV. The external field was applied after the breakdown in a discharger filled with nitrogen at 7 atm; the breakdown was caused by part of the radiation of the laser employed in our study. The moment of switching on of the field could be varied by a suitable selection of neutral light filters placed in front by a suitable selection of neutral light filters placed in front of the discharger. The duration of the electric-field pulse was made sufficiently short to avoid heating effects. The field intensity $\mathscr{E}$ in a crystal reached 1.2×10^4 V/cm. Oscillograms of the transmitted radiation in the presence and absence of an external electric field are shown in Fig. 4. A reduction in the transmission of ultrashort light pulses was observed only when the electric pulse coincided in time with that part of the light-pulse train which contained the strongest and shortest pulses. It is clear from Fig. 4 that the transmission could be reduced practically to zero by a sufficiently strong external electric field.

The destruction of the coherent state of excited electron-hole pairs in an external field could be explained by the fact that during an ultrashort light pulse the electrons were capable of acquiring an energy sufficient to destroy their resonance with the exciting radiation[12] $[(e\mathscr{E}\tau)^2/2m^* \gtrsim \Delta E$, where e is the charge and m^* the effective mass of an electron, and ΔE is the photon energy scatter]. If $\Delta E \approx 2$ meV (this value was obtained from the spectral measurements of the second harmonic of the neodymium laser using the ISP-51 spectrograph and a UF-90 camera) and $m^* \sim 10^{-28}$ g, it was found that $\mathscr{E} \gtrsim 10^3$ V/cm.

We also investigated the influence of temperature on the transmission of ultrashort light pulses. These pulses were absorbed completely in a crystal when the tempera-

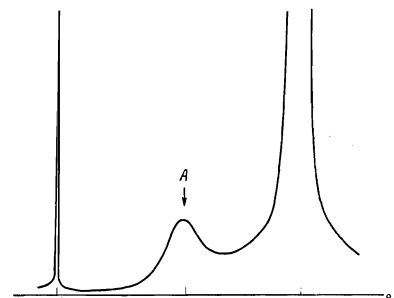

FIG. 5. Luminescence spectrum of a $CdS_{0.75}Se_{0.25}$ crystal excited with ultrashort light pulses (T = 130°K).

ture rose by 15 deg. This could be explained by additional intraband relaxation mechanisms in a semiconductor resulting in the violation of the condition $\tau < T_2$. Obviously, an efficient interaction occurred with transverse acoustic phonons of energy $E_{TA} \approx 0.01$ eV,[13] because when the temperature coefficient of the forbidden band width was taken into account (5×10^{-4} eV/deg[14]), it was found that the temperature rise of 10–15 deg gave $h\nu - E_g > E_{TA}$.

An investigation of the self-induced transparency due to interband transitions was carried out also in the case of resonance excitation with single ultrashort pulses. A single pulse was selected with a Pockels cell which was subjected to a high-voltage control pulse of 20 nsec duration (the separation between ultrashort light pulses in a train emitted by the neodymium laser was 14 nsec). However, in this case the stability of the system used to select single pulses had to be high. It is clear from Fig. 3b that the change in the serial number of a selected pulse by unity could destroy the self-induced transparency.

In all the above experiments involving the use of trains of ultrashort pulses we allowed for the heating of a sample and for the change in the forbidden band width due to the absorption of a fraction of the second harmonic of the neodymium laser radiation. We investigated also the luminescence spectrum emitted as a result of excitation with a train of ultrashort pulses (Fig. 5). The excess of the photon energy over the forbidden band width was deduced from the position of the spontaneous luminescence line of the A excitons allowing for their binding energy.

3. SELF-INDUCED TRANSPARENCY DUE TO EXCITONS

The self-induced transparency of the exciton states was investigated in $CdS_{0.75}Se_{0.25}$ crystals cooled to 80°K. In this case the second-harmonic radiation ($\lambda = 0.53\ \mu$, $\Delta\lambda \approx 5$ Å) of the mode-locked neodymium laser (Fig. 1) was in resonance with the line representing the free A excitons.[2)] We determined the transmission of a sample with a coaxial photocell matched to the I2-7 oscillograph. Oscillograms of the pulses transmitted by a sample and the dependence of G on the excitation level for a single (strongest) pulse are shown in Figs. 3e, 3d, and 2b. It is worth noting the selective properties of the semiconductor: only the strongest and shortest pulses were transmitted; moreover, additional pulses appeared behind the sample when the pump intensity was increased (these pulses corresponded to the situation in which conditions for the self-induced transparency were satisfied) when the maximum pulse amplitude was kept constant. Figure 2b shows the nonlinear dependence of the transmission on the excitation rate. At high excitation rates the transmission reached saturation rising to $G \sim 1$ for some ultrashort pulses in a train. This suggested that the anomalously high value of the transmission was not due to the saturation effect. It is clear from Fig. 2 that the excitation threshold of the self-induced transparency due to excitons was lower than the corresponding threshold for interband transitions. The special features of the nonlinear absorption of ultrashort light pulses and the values of the measured relaxation times (see below) indicated that the phenomena observed in a semiconducting single crystal of $CdS_{0.75}Se_{0.25}$ at 80°K were due to the exciton self-induced transparency.

4. MEASUREMENT OF INTRABAND RELAXATION TIMES AND OF EXCITON LIFETIME USING SELF-INDUCED TRANSPARENCY

The relaxation times were measured as follows: a weak probing pulse was used to determine the state of a medium at different times after the passage of a high-power ultrashort light pulse which caused the self-induced transparency. The probing pulse (which was a part of the second-harmonic radiation of the mode-locked neodymium laser) was produced by a wedge formed by two plane mirrors (Fig. 6). A device of this kind allowed us to vary smoothly the delay between the probing and ultrashort light pulses within the range 1–10 psec (when the wedge was moved at right-angles to the incident beam) and within the range $10–10^3$ psec (when one of the mirrors in the wedge was moved longitudinally).[15] The small angle between mirrors (30′) ensured that the beams overlapped in the samples but were spatially separated at a sufficiently long distance (~1 m) from the sample. The probing pulse power was below the self-induced transparency threshold. A sensitive (at least 10^{-6} J) calorimeter was used to measure the energy of a probing pulse transmitted by a crystal and this was done for different delays Δt relative to a high-power ultrashort light pulse.

The obtained dependences of the energy of the probing beam on Δt are shown in Fig. 7a for one-photon interband transitions in a sample of $CdS_{0.75}Se_{0.25}$ cooled to 130°K ($h\nu - E_g \approx 5$ meV); Fig. 7b shows the corresponding dependences for the resonance excitation of excitons (T = 80°K, $h\nu = E_{ex}$, where E_{ex} is the exciton energy). The characteristic scatter of the energy of the probing beam in the case of short delay times could be attributed to the T_2 relaxation. The random nature of the laser radiation could result in a considerable variation of the area under the exciting pulse θ_{10} and the

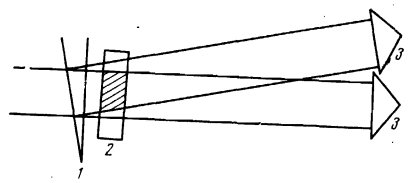

FIG. 6. System used to measure relaxation times: 1) mirror wedge; 2) investigated sample; 3) calorimeter.

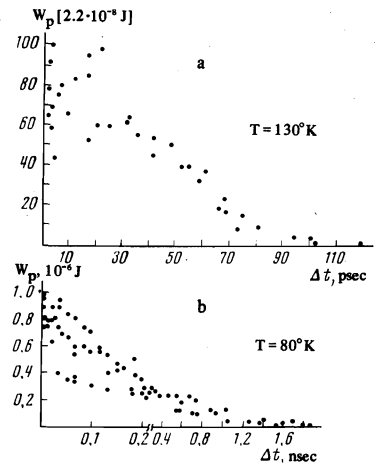

FIG. 7. Dependences of the energy of a probing beam on the delay time relative to a high-power ultrashort light pulse in the case of interband transitions (a) and resonance excitation of excitons (b).

medium crossed by the probing pulse could be either amplifying ($\theta_{10} \approx \pi$), or transparent ($\theta_{10} \sim \pi/2$), or strongly absorbing ($\theta_{10} < \pi/2$). Therefore, the energy of the probing beam should change in the $\Delta t \lesssim T_2$ case.[3)] The delay Δt corresponding to a strong fall of the fluctuations of the energy of the probing beam set the upper limit of the transverse relaxation time.

The intraband relaxation time of a $CdS_{0.75}Se_{0.25}$ crystal was $T_2 \leq 20$ psec for $h\nu - E_g \approx 5$ meV at T = 130°K. In the case of resonantly excited excitons we found that $T_2 \lesssim 0.15$ nsec (Fig. 7b). In the $\Delta t > T_2$ case the interaction became noncoherent. In this range the scatter of the experimental values of the transmitted energy was of the same magnitude as the experimental error. The smooth fall in Fig. 4b could be attributed to the T_1 relaxation effect. The exponential fall of the probing-beam energy W_p in the range $\Delta t > 0.15$ nsec could be used to estimate the relaxation time: $T_1 \approx 0.8$ nsec. This value was in agreement with the results of Kulevskiĭ,[16] who measured the time T_1 for the exciton effect by investigating the luminescence kinetics. The strong fall of the curve in Fig. 7a in the $\Delta t > 20$ psec range (absence of saturation) could be explained by the fast (characteristic time $\sim 10^{-11}$ sec) exciton formation process.[17]

5. CONCLUSIONS

The experiments carried out demonstrated the extensive potential applications of the self-induced transparency in the physics of semiconductors, which include the determination of the intraband relaxation times and mechanisms and of exciton relaxation times, measurement of the effective masses and dipole matrix elements of interband transitions (μ) far from the band edge on destruction of coherent states by external fields,[2] determination of μ from the excitation threshold of the self-induced transparence (in such investigations we have to know the shape of the exciting pulses and measurements of this kind are as yet very few[18]), and so on. There is also a promising application of the excitation of the self-induced transparency in lasers emitting highly compressed extremely short ($\sim 10^{-13}$ sec) radiation pulses,[19] which should make it possible to extend the class of semiconductors that can be investigated and to carry out studies in the range $h\nu - E_g > E_0$, where E_0 is the optical phonon energy.

We used a semiconductor with a suitable forbidden band width because the source of ultrashort light pulses was a mode-locked neodymium laser. It would be preferable to excite the self-induced transparency in semiconductors using radiation emitted by mode-locked organic lasers with a wide frequency tuning range.

The self-induced transparency in semiconductors can find interesting applications in several technological devices such as selectors of ultrashort pulses (for example, a semiconductor can be used to separate–as in Fig. 2b–the strongest and shortest pulse from a train of ultrashort pulses), small-size optical delay lines, optoelectronic circuit elements controlled by external fields, nonlinear bleachable filters, etc.

The authors are grateful to D. N. Klyshko and R. V. Khokhlov for discussing this investigation, to V. T. Platonenko, Yu. M. Popov, and M. K. Sheĭnkman for valuable advice, and to M. S. Kraevskiĭ for his help in some of the experiments.

[1)] The authors are grateful to A. V. Naumov for supplying a prototype of this photocell.

[2)] An additional check of the position of the exciton line was made using the cathodoluminescence spectra of the investigated samples.

[3)] These predictions were confirmed by a theoretical analysis of the dynamics of propagation of two ultrashort light pulses based on a model of an inhomogeneously broadened resonant medium composed of identical two-level atoms. The calculations were carried out by S. M. Zakharov and É. A. Manykin.

[1] I. A. Poluéktov and Yu. M. Popov, ZhETF Pis'ma Red. 9, 542 (1969) [JETP Lett. 9, 330 (1969)].

[2] Yu. P. Lisovets, I. A. Poluéktov, Yu. M. Popov, V. S. Roĭtberg, Kvant Elektron. (Mosc.) No. 5, 28 (1971) [Sov. J. Quantum Electron. 1, 455 (1972)].

[3] N. Tzoar and J. I. Gertsen, Phys. Rev. Lett. 28, 1203 (1972).

[4] A. Schenzle and H. Haken, Opt. Commun. 6, 96 (1972).

[5] V. V. Samartsev, A. I. Siraziev, and Yu. E. Sheĭbut, Izv. Akad. Nauk SSSR Ser. Fiz. 37, 2175 (1973).

[6] T. L. Gvardzhaladze, A. Z. Grasyuk, I. G. Zubarev, P. G. Kryukov, and O. B. Shatberashvili, ZhETF Pis'ma Red. 13, 159 (1971) [JETP Lett. 13, 111 (1971)].

[7] T. L. Gvardzhaladze, A. Z. Grasyuk, and V. A. Kovalenko, Zh. Eksp. Teor. Fiz. 64, 446 (1973) [Sov. Phys-JETP 37, 227 (1973)].

[8] F. Bruckner, V. S. Dneprovskiĭ, D. G. Koshchug, and V. U. Khattatov, ZhETF Pis'ma Red. 18, 27 (1973) [JETP Lett. 18, 14 (1973)].

[9] F. Bruckner, V. S. Dneprovskiĭ, and D. G. Koshchug, ZhETF Pis'ma Red. 20, 10 (1974) [JETP Lett. 20, 4 (1974)].

[10] V. V. Arsen'ev, V. S. Dneprovskiĭ, D. N. Klyshko, and L. A. Sysoev, Zh. Eksp. Teor. Fiz. 60, 114 (1971) [Sov. Phys.-JETP 33, 64 (1971)].

[11] V. S. Dneprovskiĭ, D. G. Koshchug, and V. U. Khattatov, Kvant. Elektron. (Mosc.) No. 2, 84 (1973) [Sov. J. Quantum Electron. 3, 144 (1973)].

[12] V. S. Roĭtberg, Candidate's Dissertation, Lebedev Physics Institute, Academy of Sciences of the USSR, M., 1974.

[13] M. Aven and J. S. Prener, Physics and Chemistry of II-VI Compounds, North-Holland, Amsterdam, 1967 (Russ. Transl., ed. by S. A. Medvedev, Mir, M., 1972).

[14] R. H. Bube, Photoconductivity of Solids, Wiley, New York, 1960 (Russ. Transl., IIL, M., 1962).

[15] F. Bruckner, V. S. Dneprovskiĭ, D. G. Koshchug, and V. U. Khattatov, Modulyatsionnaya spektroskopiya poluprovodnikov (Modulation Spectroscopy of Semiconductors), Tbilisi, 1973.

[16] L. A. Kulevskiĭ, Candidate's Dissertation, Lebedev Physics Institute, Academy of Sciences of the USSR, M., 1969.

[17] M. S. Brodin, Kvant. Elektron (Kiev) No. 8, 33 (1973).

[18] S. D. Franchenko and B. A. Frolov, ZhETF Pis'ma Red. 16, 147 (1972) [JETP Lett. 16, 101 (1972)].

[19] E. B. Treacy, IEEE J. Quantum Electron. QE-5, 454 (1969).

Translated by A. Tybulewicz
236