

Mott transition and optical bistability in CdS

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We calculate the nonlinear transmission and the intrinsic optical bistability due to induced absorption caused by strong renormalization of the band gap as a result of a Mott transition from a weakly ionized exciton gas to a strongly ionized electron–hole plasma. Models of “abrupt” (accompanied by an ionization catastrophe) and “gradual” Mott transitions are considered. The results account for the low switching thresholds (about 10 kW/cm²) recorded experimentally for CdS in the optical-bistability regime. Investigations of the dependences of the nonlinear transmission of the crystal on the polarization of the incident radiation and on the sample temperature indicate that the Mott transition in CdS is “gradual.”

INTRODUCTION

Optical bistability (OB) is realized in systems with strong nonlinearities in the presence of a positive-feedback (FB) loop. Feedback can be produced either by using an external cavity (cavity OB) or by internal processes that occur in the system itself, such as induced absorption (intrinsic OB).

Intrinsic OB of nonthermal origin in CdS was first observed in Refs. 1 and 2 (at an approximate excitation level 1 MW/cm²) and was attributed³ to a long-wave shift of the semiconductor absorption edge by single particle interaction in an electron–hole plasma (EHP) of high density (to renormalization of the band gap).

An OB regime was observed in Cd crystals (80 K) under stationary excitation by argon-laser pulses of approximate duration 10 μsec. The threshold for switching over to a state with a high absorption coefficient did not exceed 10 kW/cm². By choosing a suitable exciting-pulse duration we were able to prevent sample heating and the ensuing change of absorption with decrease of the semiconductor band gap. Simultaneously recorded luminescence spectra have revealed an exciton–exciton scattering *P* band, indicating that in the OB regime an important role is played by the processes not only in the plasma system but also in the exciton system.

We present here calculation results that account for the low switching thresholds for a bandgap semiconductor in the OB regime^{4–7} under stationary excitation, and the recorded changes of the nonlinear transmission of CdS (80 K) when the laser-emission polarization and the crystal temperature (the initial transmission) are altered by a Mott transition from a weakly ionized gas of excitons into a strongly ionized EHP and back. We analyze the system behavior in Mott transitions of two types: 1) jump of the carrier density when the Mott-transition conditions are met (in this case the total particle densities for the excitons—EHP and EHP—excitons transitions are different); 2) smooth increase of the carrier density during the Mott transition (no hysteresis in the dependence of the carrier density on the total density of the photoexcited particles).

DESCRIPTION OF MODEL

1. Kinetic Equation

To analyze the operating regimes of a nonlinear element with intrinsic feedback we used the following kinetic equation for the total density $n = n_e + n_{ex}$ of the photoexcited

particles in the semiconductor:

$$\frac{dn}{dt} = \frac{S[1 - \mathcal{T}(\hbar\omega_0, n_e, n_{ex})]}{\hbar\omega_0 d} - \frac{n}{\tau}, \quad (1)$$

where n_{ex} and n_e are the densities of the excitons and of the free electron–hole pairs, S is the radiation power density at the entry into the sample, d is the sample thickness, $\hbar\omega_0$ is the laser-emission photon energy, $\mathcal{T}(\hbar\omega_0, n_e, n_{ex})$ is the semiconductor transmission, and τ is the characteristic lifetime of the excited particles, with account taken of all the possible radiative and nonradiative recombination channels. It is assumed in Eq. (1) that the lifetimes of the excitons and of the free electron–hole pairs are equal (they actually differ insignificantly⁸), and the nonuniform distribution of the photoexcited particles over the sample thickness are disregarded (thin samples were used).

With allowance for the experimental conditions, Eq. (1) can be considered in the stationary approximation:

$$n = \frac{\tau S [1 - \mathcal{T}(\hbar\omega_0, n_e, n_{ex})]}{\hbar\omega_0 d} \quad (2)$$

2. Makeup of the Photoexcited System

At relatively low densities of the nonequilibrium carriers, when the interaction between them can be neglected, the relative contents of the electron and exciton subsystems is determined by the effective-mass law:⁹

$$n_{ex} = \frac{n_e^2}{N_T} \exp \frac{\varepsilon_{ex}(0)}{kT}, \quad (3)$$

where $N_T = (\mu kT / 2\pi\hbar^2)^{3/2}$, $\mu = m_e m_h / (m_e + m_h)$ is the reduced mass of the electron and hole, and $\varepsilon_{ex}(0)$ is the binding energy of the exciton in the low-carrier-density region. An increase of the density of the photoexcited particles leads to screening of the Coulomb interaction and to a lowering of the binding energy of the exciton. Neglecting the screening action of the dielectric exciton gas,¹⁰ the screening by free carriers can be taken into account within the framework of the Debye approximation with the aid of the expression

$$\Delta\varepsilon_{ex} = -\beta e^2 / \varkappa_0 r_D, \quad (4)$$

where β is a constant on the order of unity, is the static dielectric constant,

$$r_D = (\varkappa_0 kT / 8\pi n_e e^2)^{1/2}$$

is the Debye screening radius. As shown first by Mott,¹¹ at $r_D < a_{ex}$ (more accurate calculations¹² lead to the condition $r_D \leq 0.84a_{ex}$, where a_{ex} is the exciton radius) the screening becomes so strong that binding into excitons becomes impossible, in which case a fully ionized EHP is produced in the system. The carrier density n_e^M needed for the Mott transition is determined from the relation

$$n_e^M = \frac{\kappa_0 kT}{8\pi e^2 (0.84a_{ex})^2}.$$

The effective-masses law for a system of excitons and free carriers, with allowance for screening and for the Mott transition, can thus be written in the form

$$n_{ex} = \begin{cases} (n_e^2/N_T) \exp[\varepsilon_{ex}(n_e)/kT], & n_e \leq n_e^M \\ 0, & n_e > n_e^M \end{cases}, \quad (5)$$

where we have introduced the exciton binding energy

$$\varepsilon_{ex}(n_e) = \varepsilon_{ex}(0) \left(1 - \frac{0.84a_{ex}}{r_D}\right) = \varepsilon_{ex}(0) [1 - (n_e/n_e^M)^{1/2}] \quad (6)$$

which depends on the carrier density n_e (the constant β in (4) was chosen such that $\varepsilon_{ex}(n_e) = 0$ at $n_e = n_e^M$). Note, however, that a Mott transition (in the form of an abrupt jump of the carrier density) has so far not been revealed in experiment by the associated abrupt increase of the photoconductivity or by strong changes of the spectral properties of the semiconductor (see, e.g., Refs. 13 and 14). We shall therefore use hereafter for the analysis of nonlinear transmission and OB in CdS, beside the "abrupt" Mott transition, also the possibility of a "gradual" transition, in which an ionization equilibrium condition in the form

$$n_{ex} = \frac{n_e^2}{N_T} \times \exp\left\{\frac{\varepsilon_{ex}(0)}{kT} \left[1 - \left(\frac{n_e}{n_e^M}\right)^{1/2}\right]\right\} \quad (7)$$

is satisfied in the entire range of the density n_e .

Note the assumption of a gradual Mott transition is of principal significance and alters substantially the form of the dependence of n_e on n . In the case of an abrupt Mott transition the dependence of n_e on n exhibits hysteresis (Fig. 1,

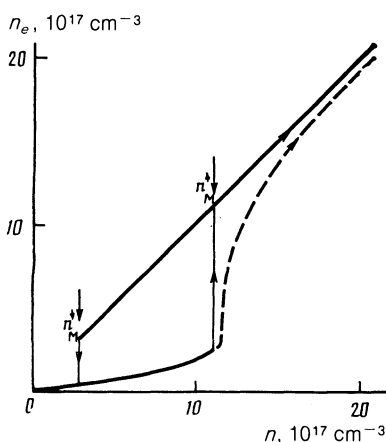


FIG. 1. Dependences of the electron-subsystem density n_e on the total density of photoexcited particles in CdS (80 K) in the cases of abrupt (solid) and gradual (dashed) Mott transitions.

solid line), due to the different screening influences of the excitons and the free carriers. The total photoexcited-pair density n_M needed for the excitons—EHP transition is substantially higher than the density n_M of the reverse transition: $n_M^i = n_e^M(1 + n_e^M/N_T)$, $n_M^r = n_e^M$.

In the case of a smooth Mott transition the density n_e increases rapidly near the value n_M , but there is no hysteresis in the dependence of n_e on n (Fig. 1, dashed line; for $n < n_M^i$ the dependence of n_e on n in the case of a smooth Mott transition coincides with the analogous dependence for an abrupt transition).

3. Absorption Coefficient

The absorption coefficient K was calculated by adding the exciton (K_{ex}) and interband (K_g) contributions with account taken of the A and B subbands of the valence band. In the case of an abrupt Mott transition, the exciton absorption at $n_e > n_e^M$ was assumed to be zero.

To take into account the exciton correlations (the Sommerfeld contribution), the interband absorption coefficient was approximated by a hyperbolic tangent. For the A subband, for example,

$$K_g^A = 1/2 K_0^A \{ \text{th} [(\hbar\omega - E_g^A) / \Gamma_g] + 1 \}, \quad (8)$$

where E_g^A is the band gap whose temperature dependence was determined from the relation¹⁵

$$E_g^A = E_{g,0}^A - QT^2 / (\gamma + T)$$

($E_{g,0}^A$ is the width of the band gap at $T = 0$, Q and γ are constants that depend on the type of semiconductor), Γ_g is the width of the absorption edge and is calculated using the formula

$$\Gamma_g = \frac{\hbar\omega_{LO}}{\sigma_0} \text{th}^{-1} \left(\frac{\hbar\omega_{LO}}{2kT} \right),$$

which yields the Urbach limit in the $K_g^A(\hbar\omega)$ dependence on the long-wave section of the absorption edge ($\hbar\omega_{LO}$ is the LO-phonon energy, σ_0 is a dimensionless constant of order unity). The expression for K_g^B is similar to (8).

The constant K_0^A in (8) was calculated using the relation¹⁶

$$K_0^A = \frac{2^{1/2} \mu^{1/2} \kappa_\infty^{1/2} |H_{c,v}^A|^2}{c\hbar^4} [\varepsilon_{ex}(0)]^{1/2},$$

where $H_{c,v}^A$ is a matrix element of the radiative transition between the A -subband of the valence band and the conduction band, and κ_∞ is the high-frequency dielectric constant.

The influence of multiparticle interaction on the interband absorption is taken into account by introducing the band gap $E_g = E_g(n_e)$, which depends on the free-carrier density and whose variation must ensure stability of the position of the exciton level (this last circumstance was confirmed by numerous experiments). The renormalization of the band gap should thus compensate for the decrease of the exciton binding energy, and can be calculated in accordance with (6) from the relation

$$E_g^A(n_e) = E_g^A(0) - \varepsilon_{ex}(0) (n_e/n_e^M)^{1/2}.$$

The complete expression for E_g with allowance for the temperature and density dependences is

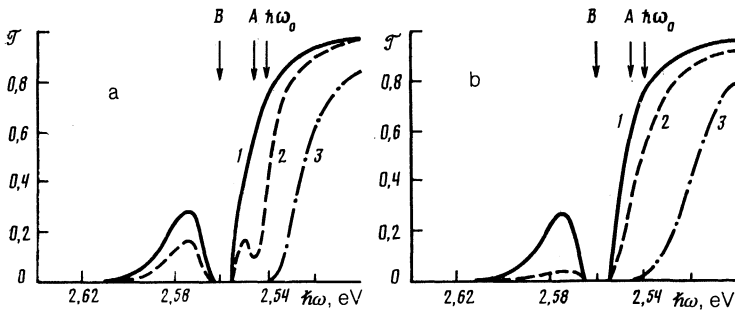


FIG. 2. Calculated transmission spectra of CdS crystal ($T = 80$ K, thickness $d = 1 \mu\text{m}$) assuming a gradual Mott transition: a— in the absence of excitation at $x = 0$ ($\mathbf{E} \parallel \mathbf{c}$) (curve 1), $x = 0.1$ (2), $x = 1$ ($\mathbf{E} \perp \mathbf{c}$) (3); b) in the presence of excitation and $x = 0$ for $n = 0$ (1), $n = 5.6 \times 10^{17} \text{ cm}^{-3}$ ($n_e = 0.3 \cdot n_e^M$) (2), $n = 1.2 \cdot 10^{18} \text{ cm}^{-3}$ ($n_e = 3 \cdot n_e^M$) (3). The arrows mark the spectral position of the laser line and of the absorption lines connected with A and B excitons.

$$E_{g^A}^A(T, n_e) = E_{g,0}^A - \frac{QT^2}{\gamma + T} - \varepsilon_{ex}(0) \left(\frac{n_e}{n_e^M} \right)^{1/2}.$$

Note that under the conditions of the considered experiments⁴⁻⁷ (laser-emission photon energy $\hbar\omega_0 = 2540$ eV, sample temperature $T = 80$ K) the influence of the band filling (of the dynamic Burstein–Moss effect) on the interband absorption coefficient could be neglected, since the condition $\hbar\omega_0 - \mu_p > kT$ (μ_p is the EHF chemical potential) was met even at the maximum experimentally attainable densities n_e .

The exciton-absorption coefficient was approximated by Lorentzian profile:

$$K_{ex^A}(\hbar\omega) = \frac{\pi\alpha_A [E_{ex^A}^A(T)]^2 \Gamma_{ex}}{\hbar\omega_\infty^{1/2} c \{ [E_{ex^A}^A(t) - \hbar\omega]^2 + (\Gamma_{ex}/2)^2 \}}, \quad (9)$$

where α_A is the A -exciton polarizability, $E_{ex^A}^A(T)$ is the A -exciton ground-state energy and has the same temperature dependence as $E_{g^A}^A$, and Γ_{ex} is the width of the exciton absorption line. The nonlinear changes of the exciton absorption can be due to the following: change of the exciton polarizability as a result of screening of the Coulomb interaction, shift of the exciton level, broadening of the exciton absorption peak. According to the results of Ref. 17, the exciton polarizability changes insignificantly even near Mott carrier densities, and the absence of a shift of the exciton level with change of the pump power was confirmed by experiment more than once. The most probable cause of the nonlinearity of the exciton absorption is thus the broadening of the exciton line, which was taken into account with the aid of the expression

$$\Gamma_{ex} = \Gamma_{ex,0}(T) + 2\pi\hbar(2a_{ex})^2 [\sqrt{2} \langle v_{ex} \rangle (n_{ex} + n_e) + \langle v_e \rangle n_e], \quad (10)$$

where $\langle v_{ex} \rangle$ and $\langle v_e \rangle$ are respectively the average velocities of the excitons and electrons, and $\Gamma_{ex,0}(T)$ is the tempera-

ture dependent width of the exciton line in the region of low densities of photoexcited carriers (densities calculated from the data of Ref. 18 with allowance for scattering by acoustic and optical phonons).

Transitions connected with the A subband of the valence band are forbidden in the polarization $\mathbf{E} \parallel \mathbf{c}$, whereas for transitions with participation of the B subband the oscillator strength is practically independent of the incident-radiation polarization, a fact taken into account by introducing in the final expression for the total absorption coefficient K an additional parameter x that ranges from zero to unity ($x = 0$ at $\mathbf{E} \parallel \mathbf{c}$ and $x = 1$ for $\mathbf{E} \perp \mathbf{c}$). The final expression for K is

$$K = (xK_{g^A}^A + K_{g^B}^B) + m(xK_{ex^A}^A + K_{ex^B}^B), \quad (11)$$

where $m = 1$ for $n_e \leq n_e^M$ and $m = 0$ for $n_e > n_e^M$, and in the case of an abrupt Mott transition and $m = 1$ for all values of n_e for a gradual one.

The semiconductor transmission \mathcal{T} , which enters in Eq. (2) for the total density of the photoexcited particles, was calculated with neglect of interference effects (the interference-induced modulation of the transmission from the short-wave side of the laser line was insignificant even at a polarization $\mathbf{E} \parallel \mathbf{c}$). Here $\mathcal{T} = \exp(-Kd)$ (d is the sample thickness). Figure 2 shows an example of the calculated transmission spectra of CdS ($T = 80$ K, $d = 1 \mu\text{m}$) in the absence (a) and presence (b) of excitation, assuming a gradual Mott transition. The CdS parameters used in the calculations were: $\varepsilon_{ex}(0) = 29.5$ meV (Ref. 19), $a_{ex} = 30 \text{ \AA}$ (Ref. 20), $E_{g,0}^A = 2.585$ eV, $E_{g,0}^B = 2.600$ eV, $Q = 0.54$ meV/K, $\gamma = 279$ K (the temperature constant Q and γ , and also the energies $E_{g,0}^A$ and $E_{g,0}^B$, were determined from the temperature restructuring of the exciton-reflection spectra in the regions of the A and B resonances²¹), $m_e = 0.185m_0$, $m_h = 1.105m_0$ (m_0 is the free-electron mass)²², $\kappa_0 = 8.58$, $\kappa_\infty = 5.26$ (Ref. 22), $\alpha_A = 1.08 \cdot 10^{-3}$, $\alpha_B = 0.5 \cdot 10^{-3}$,

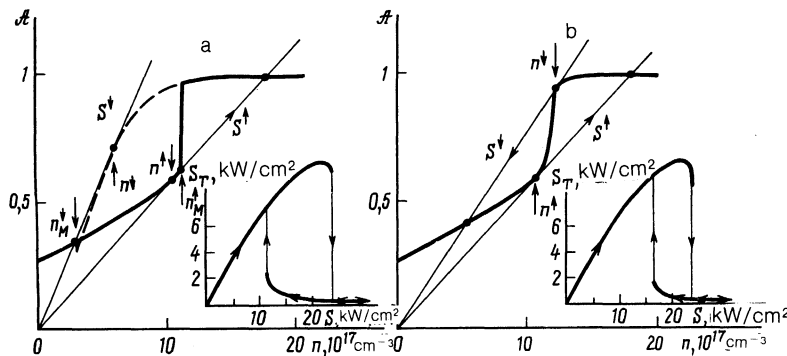


FIG. 3. Dependences of the loss coefficient on the total density of the photoexcited particles for CdS ($T = 80$ K, $x = 0$) in the case of abrupt (a) and gradual (b) Mott transitions. The thin straight lines show the tangents to the $\mathcal{L}(n)$ plot, corresponding to the threshold values S^1 and S^2 of the switching power. The insets show the corresponding hysteretic dependences of S_T on S .

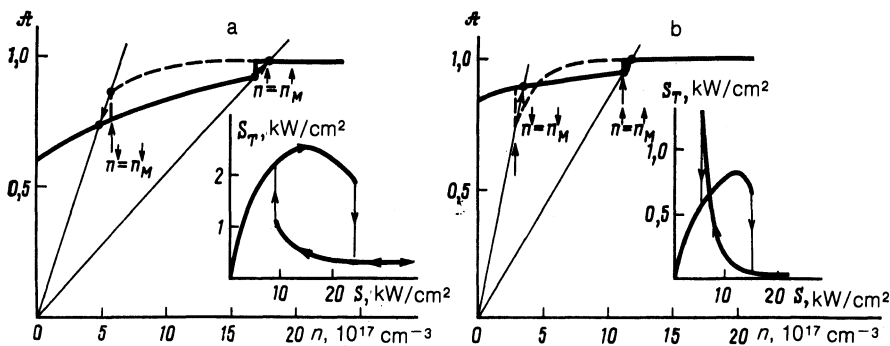


FIG. 4. Dependences of the loss coefficient \mathcal{A} on the total density n of the photoexcited particles and the corresponding hysteresis plots of $S_T(S)$ for CdS in the case of an abrupt Mott transition: a— $x = 0$, $T = 150$ K, b— $x = 0.3$, $T = 80$ K.

$|H_{c,v}^A| = 8.32 \cdot 10^{-46}$ erg 2 cm 3 , $|H_{c,v}^B| = 3.88 \cdot 10^{-46}$ erg 2 cm 3 (the exciton polarizabilities and the matrix elements of the interband transitions were calculated for the following oscillator-strength values: $f_{ex}^A = 3.22 \cdot 10^{-3}$ (Ref. 20), $f_{ex}^B = \frac{1}{2}(f_{ex,B}^{\perp} + f_{ex,B}^{\parallel}) = 1.5 \cdot 10^{-3}$ (Ref. 19), $\hbar\omega_{LO} = 38$ meV, 22 and $\sigma_0 = 2.22$ (Ref. 23).

CALCULATION RESULTS AND THEIR DISCUSSION

Equation (2) for the total density of photoexcited particles is compatible with expressions (5) or (7), which determine the partial makeup of the electron-hole system, and Eqs. (8), (9), and (11) for the calculation of the coefficients of interband, exciton, and total absorptions (with allowance for the expressions for E_g and Γ_{ex}) constitute the complete set of equations for the determination of the nonlinear response of a semiconductor to incident laser radiation.

The solutions of Eq. (2) can be obtained by graphical means, as the points where the plot of the loss coefficient \mathcal{A} vs n ($\mathcal{A} = 1 - \mathcal{T} = 1 - \exp(-Kd)$) intersect the straight line $n\hbar\omega_0 d / \tau S$ (Figs. 3a, b). In the case of an "abrupt" Mott transition (Fig. 3a), owing to the hysteresis in the dependence of n_e on n (see Fig. 1), the form of the dependence of \mathcal{A} on n in the density interval $n_M^l < n < n_M^h$ is different for increasing and decreasing input intensities (Fig. 3a; the solid line corresponds to increase of n on the leading front of the incident pulse, and the dashed line corresponds to decrease of n on the trailing edge). For both abrupt and gradual Mott transitions there exists a range of incident-radiation intensities at which Eq. (2) has more than one solution, meaning bistability of the system. The critical values of the power density needed to switch over the sample into a state with high (S^h) and low (S^l) absorption correspond to tangents, shown by thin straight lines in Figs. 3a and 3b, to the plot of

$\mathcal{A}(n)$ (n^l and n^h are the critical values of the total density). The hysteresis in the plots of the intensity S_T of the transmitted light against the intensity S of the light incident have no qualitative differences at $T = 80$ K and $E \parallel e$ for abrupt and gradual Mott transitions (insets in Figs. 3a and 3b). These plots begin to differ substantially when the crystal temperature is raised or when the polarization of the incident radiation is changed (with increase of the parameter x).

According to the calculation results, for an abrupt Mott transition a rise of the sample temperature broadened the range of intensities S in which a bistable transmission regime exists (Fig. 4a). Note that whereas at $T = 80$ K the absorption was turned on and off before the Mott densities n_M^l and n_M^h were reached (Fig. 3a; $n^l < n_M^l, n^h > n_M^h$), for $T > 110$ – 120 K the crystal switching was due to the jump of the density n_e , due to an ionization catastrophe in the abrupt Mott transition (Fig. 4a; $n^l = n_M^l, n^h = n_M^h$).

We note one more feature of the calculated hysteretic dependences observed in the temperature interval 100–120 K. When the crystal is switched over in this case, the transmission does not increase (as, for example, at $T = 80$ or 150 K, Figs. 3a and 4a), but decreases, owing to the turning-on of the exciton absorption on going from EHP to excitons. Note that a similar feature was observed at $T = 80$ K in the hysteretic dependence of S_T on S when the polarization differed from $E \parallel c$ ($x = 0$), as illustrated in Fig. 4b ($T = 80$ K, $x = 0.3$).

The changes of the calculated dependences of S_T on S for a gradual Mott transition were entirely different. When the temperature was raised the intensity region in which the solution of Eq. (2) is not single-valued narrowed down rapidly (see Fig. 5a and its inset), while at temperatures $T \geq 100$ K the dependence of S_T on S no longer had hysteresis (Fig.

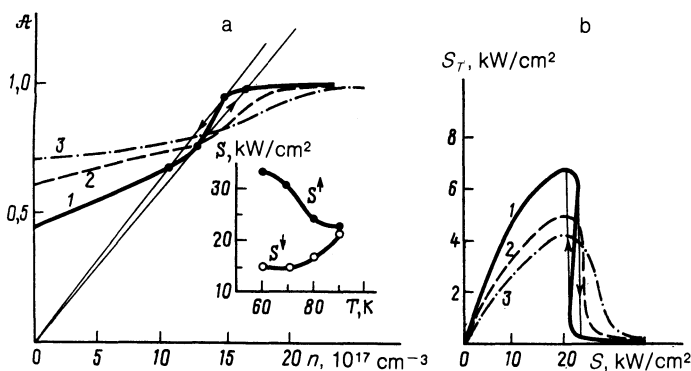


FIG. 5. Dependences of the loss coefficient \mathcal{A} on the total density n of the photoexcited particles (a) and the corresponding $S_T(S)$ dependences (b) for CdS ($x = 0$) at $T = 90$ K (1), 100 K (2), 110 K (3) in the case of a gradual Mott transition. The inset shows the dependence of the threshold values S^h and S^l of the switching power on the sample temperature.

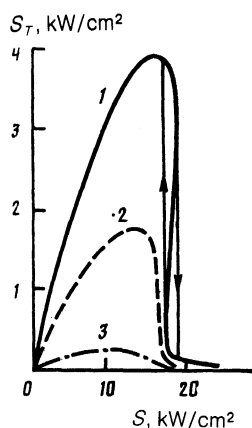


FIG. 6. Plots of S_T against S for CdS ($T = 80$ K) in the case of a gradual Mott transition for $x = 0.1$ (1), 0.2 (2), 0.5 (3).

5b). Similar dependences (transition from bistable transmission to ordinary nonlinear one) appeared also upon increase of the parameter x connected with the polarization of the incident radiation and with the corresponding change of the initial absorption (Fig. 6).

Comparison of the calculation with the experimental results⁴⁻⁷ favors the assumption that the Mott transition is gradual. For an abrupt Mott transition the OB regime should exist in a wide range of temperatures and polarizations, contrary to the experimental data (Fig. 7). Hysteresis on the dependence of S_T on S could be obtained experimentally only in a polarization close to $E \parallel c$ (polarization angle $\varphi = 0$) at minimum heating of the sample (using pulses of duration not longer than 10–15 μsec). Even an insignificant increase of the polarization angle (Fig. 7, curves 1 and 2) or lengthening of the light pulses (Fig. 7, curves 1 and 3), have led to vanishing of the hysteresis and to a transition from the OB regime to the regime of nonlinear transmission with induced absorption. Neither was there ever observed in experiment a decrease of the sample transmission on the trailing edge of the incident pulse, predicted in the abrupt Mott transition model in the temperature range 100–120 K ($x = 0$) or for an incident-radiation polarization other than $E \parallel c$ (Fig. 4b).

Note that the calculated values of the switchover thresholds S^+ range from 20 to 30 kW/cm^2 , somewhat higher than the measured values⁴⁻⁷ ($S \leq 10 \text{ kW}/\text{cm}^2$), but much lower than the switching thresholds for the OB due exclusively to the renormalization of the band gap for excitation of high-density EHP without allowance for the Mott transition.¹⁻³

It was noted in Ref. 4 that at excitation intensities close to the threshold values for the OB regime, the P -band of exciton–exciton scattering dominated in the luminescence spectra of CdS (80 K). This pointed to a high efficiency of the exciton–exciton collision process, which leads to broadening of the exciton absorption line. The presence of this process alone cannot lead to a bistable behavior of the system,²⁴ but the induced absorption connected with the collision broadening contributes to development of the OB regime in a Mott transition. Calculations made for CdS ($T = 80$ K, $x = 0$), show that when account is taken of the collision broadening the switching threshold S^+ is lower by approximately 1.5 times than without this account.

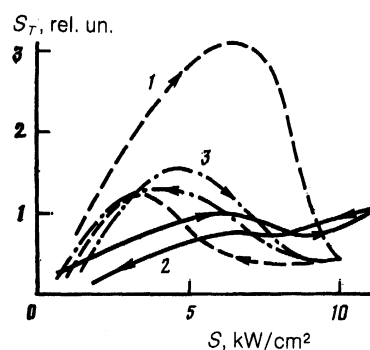


FIG. 7. Experimental dependences of the intensity S_T of the transmitted light on the intensity S of the incident light for a CdS crystal ($T = 80$ K) at different polarization of the incident radiation (φ is the angle between the vector E in the laser beam and the c axis of the crystal) and at different durations τ_p of the pulses employed: $\tau_p = 10 \mu\text{sec}$, $\varphi = 0^\circ$ (1), $\tau_p = 10 \mu\text{sec}$, $\varphi = 20^\circ$ (2); $\tau_p = 24 \mu\text{sec}$, $\varphi = 0^\circ$ (3).

We note in conclusion that the nature of the Mott transition in semiconductors remains unclear to this day.^{9,25,26} Moreover, there is even no simple experimental criterion that distinguishes the plasma state of an electron–hole system from the exciton state. In recent investigations of the dynamics of evolution of stimulated emission in CdSe and CdS (Ref. 27) we succeeded in recording a transition from a plasma to excitons when the density of photoexcited carriers was decreased, as a result of their recombination, as revealed by an abrupt increase of the induced-radiation pulse delay time relative to a picosecond excitation pulse. The results of the present papers suggest that the Mott transition proceeds smoothly and is not connected with an ionization catastrophe in the entire excited volume of the crystal. Note that in addition to the smooth Mott transition model proposed above, the absence of jumps in the dependence of the crystal transmission on the density of the photoexcited particles can be attributed to spatial lamination of the excited volume into regions occupied by exciton gas and plasma.²⁸ An increase of the excitation intensity should lead to an increase of the volume occupied by the EHP, and to a gradual change of the transmission of the crystal. In this case the Mott transition is a first-order phase transition, but this is at variance with experimental results.¹⁴

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