Lasing on a transition between quantum-well levels in a quantum dot

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Lasing has been observed on transitions between quantum-well levels in a quantum dot (consisting of microscopic semiconductor crystals in a glassy matrix).

The energy spectrum of a quantum dot (a quasi-zero-dimensional structure) is determined by its characteristic size. There is obvious interest in achieving lasing on transitions between the quantum-well levels of a quantum dot, since by varying the size of the structure one could tune the wavelength of the laser. In some previous experiments[1] we detected a decrease in absorption at frequencies corresponding to transitions between quantum-well levels of quantum dots during excitation of microscopic semiconductor crystals by intense ultrashort light pulses. The size of the effect suggested that a gain regime might be attainable.

In this letter we are reporting a study of the dynamic changes in the absorption and gain (!) spectra in microscopic crystals of CdSe (at 80 K) of various radii (3–10 nm) during excitation by ultrashort light pulses. The test samples which exhibited a gain have been used to achieve lasing.

We studied the time evolution in the absorption (or gain) spectra by a standard two-pulse excitation-probing procedure. The samples were pumped with ultra short pulses at the second harmonic of a Nd:YAG laser (the pulse length was about 20 ps, the energy W was varied up to 0.5 mJ). The probing was carried out with ultrashort pulses of "white" light, generated by tapping part of the main laser beam into a cell holding heavy water. Spectra of the probing light before and after passing through the sample were measured with an OVA-284 multichannel analyzer.

Figure 1 shows absorption spectra $\alpha(\lambda)$ for various values of the delay Δt between the exciting and probing pulses for samples in which the microscopic crystals had different average radii, $\overline{R} = 3.5$ nm (sample 1) and 6 nm (sample 2). The size of the microscopic crystals was determined from the positions of the bleaching peaks in the differential transmission spectra.[1]

The ultrashort pump pulses caused bleaching near the absorption edge of the samples with peaks at frequencies corresponding to allowed transitions between quantum-well levels in the valence and conduction bands.[1] The value of $|\Delta\alpha|/\alpha_0$ (α_0 is the absorption coefficient for the unexcited sample; $\Delta\alpha = \alpha - \alpha_0$) for sample 1 reached a maximum of 0.7 (at $\Delta t = 0$), but a gain regime ($\alpha < 0$) was not reached even at the maximum pumping level. The relaxation times of the induced change in absorption coefficient did not exceed 100 ps.

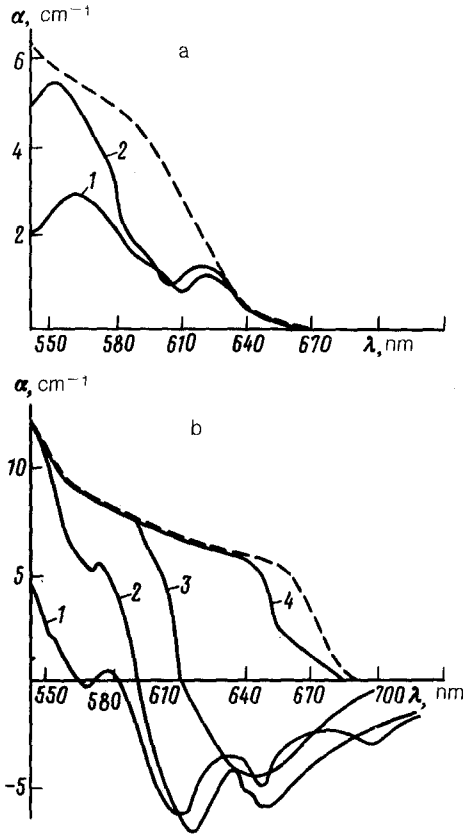

FIG. 1. Absorption (gain) spectra of unexcited (dashed lines) and excited (solid lines) microscopic crystals of CdSe (at 80 K) for various values of the delay Δt between the exciting and probing pulses. a: Sample 1, $W = 0.25$ mJ. 1—$\Delta t = 0$; 2—33 ps. B: Sample 2, $W = 0.12$ mJ. 1—$\Delta t = 0$; 2—66 ps; 3—133 ps; 4—2.85 ns.

In sample 2, the ultrashort pump pulses caused not merely a bleaching but also the appearance of a fairly broad spectral region in which a gain ($\alpha < 0$) arose. The gain peaks corresponded to the energies of $1s$–$1s$ ($\lambda_1 = 649$ nm) and $1p$–$1p$ ($\lambda_2 = 616$ nm) transitions. The maximum values of the gain (about 5–7 cm^{-1}) were comparable to the absorption coefficient of the unexcited sample. This result corresponds to the attainment of a complete population inversion for these levels. The gain at the wavelength λ_2 relaxed over a time of about 100 ps, while negative values of α near λ_1 persisted for a relatively long time (at least 1 ns).

A resonator was formed from sample 2 in order to arrange lasing. The thickness of the resonator was about 1 mm (the reflectances of the mirrors at a wavelength of 650 nm were 100% and 95%). Longitudinal pumping was provided by an unfocused second-harmonic beam sent through one of the mirrors (the transmittance of the mirror at the pump wavelength was 30%). At low pump levels, a broad band with a

peak near 682 nm was visible in the luminescence spectra (spectrum 1 in Fig. 2). This peak had the same position and the same shape as the emission line of sample 2 without reflecting coatings.[1] With increasing energy of the ultrashort pump pulses, a stimulated emission appeared in the direction perpendicular to the resonator mirrors. The spectrum of this stimulated emission (spectrum 2 in Fig. 2) was shifted in the short-wave direction from the peak of the spontaneous band. At excitation levels of about 4 μJ, we detected an intense, directional emission with a wavelength $\lambda_g = 644$ nm (the width of the line was $\Delta\lambda = 7$ nm), which corresponds to a transition between low-lying quantum-well levels. This intense emission (see the inset in Fig. 2) appeared abruptly. With increasing excitation level, the emission line broadened and shifted in the long-wave direction ($\lambda_g = 650$ nm and $\Delta\lambda = 14$ nm at $W = 0.1$ mJ).

In the case of the small-radius microscopic crystals, in which the exciton correlations are negligible, and the spectrum of states is determined by the independent quantization of the energies of the electron and the hole, the dipole matrix element of an allowed transition for any pair of valence-band and conduction-band levels does not depend on the dimensions of the microscopic crystal. It is equal to the dipole moment of the bulk semiconductor,[2] d_{cv}. The electric susceptibility of the excited sample, with a microscopic-crystal concentration N_m ($N_m = \xi / V_m$, where ξ is the concentration by volume of the semiconductor in the sample, and V_m is the volume of an individual microscopic crystal), can be found by summing the contributions of all the quantum-well levels and the three valence subbands ($i = A,B,C$), if inhomogeneous broadening is ignored:

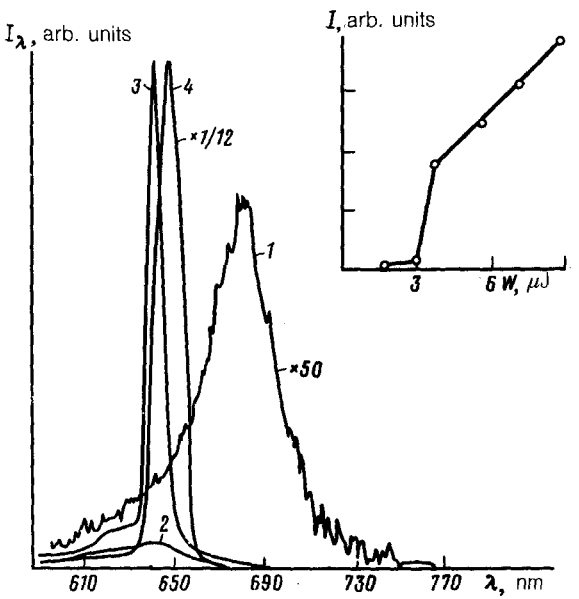

FIG. 2. 1—Spontaneous luminescence spectrum of CdSe microscopic crystals (Sample 2) at $W = 0.5$ μJ; 2–4—spectra of the emission directed perpendicular to the resonantor mirrors at $W = 3$, 5, and 100 μJ, respectively. The inset shows the intensity of the emission in the direction perpendicular to the resonantor mirrors versus the energy of the ultrashort pump pulse.

$$\chi(\hbar\omega) = N_m \sum_{i,l,n} |d_{cv}^i|^2 \frac{2(2l+1)(n_{l,n}^e + n_{i,l,n}^h - 1)}{(\hbar\omega - E_{i,l,n}) + i\hbar\Gamma_{i,l,n}} . \quad (1)$$

Here $E_{i,l,n}$ is the energy of the transition between the levels with quantum numbers l and n (Ref. 3) for valence subband i; $\Gamma_{i,l,n}$ is the homogeneous width of the transition; and $n_{l,n}^e$ and $n_{i,l,n}^h$ are the occupation numbers of the electron and hole levels, respectively. The expression $(n_{l,n}^e + n_{i,l,n}^h - 1)$ was introduced in (1) to deal with the filling of states caused by the exciting light. The absorption coefficient (or the gain) is determined by the imaginary part of the susceptibility: $\alpha(\hbar\omega) = [4\pi \mathrm{Im}\chi(\hbar\omega)]/en_0$, where n_0 is the refractive index. The gain at the frequency ω, corresponding to the energy for the first transition ($\hbar\omega = E_{A,0,1}$) reaches a maximum value in the case $n_{0,1}^e = n_{A,0,1}^h = 1$. This maximum value is given by

$$g_{max} = \frac{16\pi^2 N_m |d_{cv}^A|^2}{\lambda n_0 (\hbar\Gamma_0)}, \quad (2)$$

where $\Gamma_0 = \Gamma_{A,0,1}$. For the particular parameter values of sample 2 [$\xi = 0.1\%$, $\bar{R} = 6$ nm, and $\hbar\Gamma_0 = 44$ meV (determined from the width of the peak in the differential transmission spectra[1])] and for $d_{cv}^A = 2.4 \times 10^{-17}$ cgs units (according to the data of Ref. 4), we find $g_{\max} = 8\ \mathrm{cm}^{-1}$. This result is close to the value measured experimentally (Fig. 1b). It can be seen from (2) that the gain $g_{\max}$ increases with increasing concentration of the microscopic crystals and thus with decreasing radius of these crystals at a fixed concentration by volume of the semiconductor, ξ. In the samples with the microscopic crystals of the smaller radius, the measurements show that it is a more complicated matter to achieve the gain regime, because of the higher threshold for the transition to this regime (the threshold energy is proportional to the concentration of microscopic crystals) and also because of the dominant role played by radiationless recombination in small-radius crystals. The relaxation time mentioned above (100 ps) for the microscopic crystals with $\bar{R} = 3.5$ nm is much shorter than the radiative-recombination time.[5]

The lasing efficiency could be raised, and lasing could be achieved at room temperature, by increasing the concentration of the microscopic crystals.

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