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Short Communication

Intensity-dependent absorption coefficient and refractive index near the band gap of highly excited semiconductors

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Résumé. — Nous étudions la dépendance du coefficient d'absorption α et de l'indice de réfraction n en fonction de l'intensité de lumière incidente au voisinage de la résonance excitonique. Le comportement non-bosonique des excitons peut constituer un mécanisme de non-linéarité optique conduisant à des formes bistables de α et n au-dessus de cette résonance.

Abstract. — The dependence of absorption coefficient α and refractive index n on the intensity of incident light are studied near the exciton resonance. The non-boson behaviour of excitons may serve as a mechanism of optical nonlinearity leading to bistable shapes of α and n above the resonance.

Two important branches of research of optical properties in highly laser-excited semiconductors near the absorption edge are the derivation of the dependence of the dielectric function ε on exciton density σ [1, 2] and the determination of the conditions for the occurrence of density bistability (DB) [3-5]. As the intensity I of incident pumping laser is, among other quantities, an experimentally controlable parameter, it is reasonable and even necessary to have concrete optical characteristics dependent directly on I rather than on σ. The aim of this communication is to study such a dependence which in a sense can be seen as a bridge connecting the two branches of research mentioned above. Similar task has been done recently in the spectral region near the two-photon biexciton resonance [6].

Let us start from an effective bosonic Hamiltonian of Hanamura [7] describing a system of many interacting excitons placed under the action of an externally driven monochromatic classical laser field with frequency ω : ($\hbar = c = 1$ being used hereafter)

$$
\begin{aligned}
H &= \sum_p \omega_{xp} a_p^+ a_p + \frac{w}{2V} \sum_{lpq} a_{l+q}^+ a_{p-q}^+ a_p a_l - \\
&- du V^{1/2} \left\{ a_k^+ + a_{-k} + \frac{\mu}{V} \sum_{pq} \left[a_{k+q}^+ a_{l-q}^+ a_l + a_l^+ a_{l-q} a_{q-k} \right] \right\} E_k(t)
\end{aligned}
\tag{1}
$$

where $E_k(t)$ the electric field strength of the radiation inside the crystal, a_p^+ the creation operator of an exciton with wave vector p and energy ω_{xp}, V the sample volume, d the matrix element of dipole moment, $w = 26\pi E^b r^3/3$, $u = \pi^{-1/2} r^{-3/2}$, $\mu = 7\pi r^3$ with E^b and r being the binding energy and radius of an exciton. The dielectric function (ε_∞ the background dielectric constant)

$$
\varepsilon = \varepsilon_\infty + \frac{< P_k >}{E_k} \tag{2}
$$

can be derived from the knowledge of the averaged (over the eigenstate of H) value of the polarization operator P_k which within the Hartree-Fock approximation is of the form :

$$
P_k = du V^{-1/2} (1 - 2\mu\sigma)\,(a_k + a_{-k}^+) \tag{3}
$$

Using (1) to set up the equations of motion for a_k and a_{-k}^+ which will then be solved in the framework of the Hartree-Fock approximation for a stationary regime. Putting the solutions of a_k and a_{-k}^+ into (3) and then (3) into (2) we obtain :

$$
\varepsilon(\omega, \sigma) = \varepsilon_\infty - \frac{2e^2 \Pi_{cv}^2 m^{-1} u^2 (1 - 2\mu\sigma)^2}{(\omega_x + 2w\sigma - i\gamma)\left[\omega^2 - (\omega_x + 2w\sigma - i\gamma)^2\right]} \tag{4}
$$

where Π_{cv} the interband matrix element of the wave-vector operator, e and m the free electron charge and mass and γ the exciton inverse dephase time being phenomenologically introduced. The absorption coefficient α and the refractive index n of CdS obtained from (4) are plotted as functions of $x = \omega / \omega_x$ in figures 1 and 2, resp., for $\sigma = 0$; 9×10^{15} and 1.8×10^{16} cm^{-3}. The

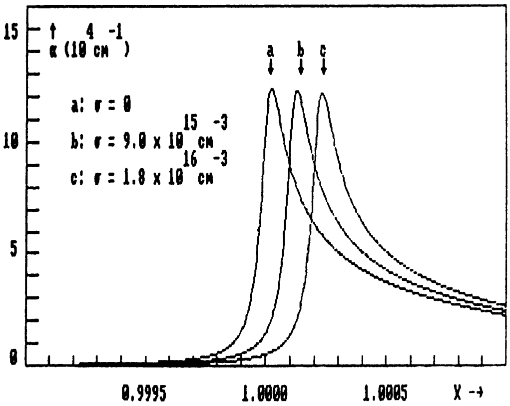

Fig.1.

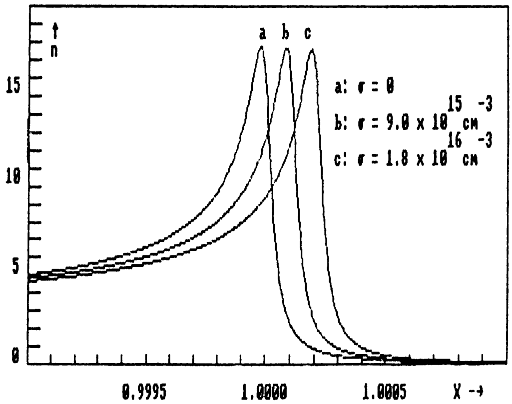

Fig.2.

Fig.1 – Absorption coefficient as functions of x.

Fig.2 – Refractive index as functions of x.

used parameters for CdS are $\varepsilon_\infty = 7.6$, $\omega_x = 2.5528$ eV, $E^b = 32.9$ meV, $r = 25.5 \times 10^{-8}$ cm, $m_e = 0.205\ m$, $m_h = 1.348\ m$, E_g (band gap) $= 2.5857$ eV and $\gamma^{-1} = 7$ ps. We see that the peaks of the curves are shifting to higher frequency region for increasing exciton density. This is due to the blue shift of the exciton energy level which in turn is caused by the non-boson character of excitons ($w = 26\pi E^b r^3/3$ is the contribution of purely kinematical exciton-exciton interactions ; the dynamical ones vanish in the $k = 0$ limit used here. For details see [7]). As a matter of fact, the excitons are generated optically and σ must be governed by the input laser intensity $I = |E|^2/T$ with $T = 1 - |1-\sqrt{\varepsilon}|^2/|1+\sqrt{\varepsilon}|^2$ being the transmition coefficient at the vacuum-crystal boundary surface. Thus, α and n as functions of I depend parametrically on σ. The $(\sigma - I)$-relation is followed from the balance condition in a stationary situation :

$$I\tau T\varepsilon_2 = \sigma n \tag{5}$$

where $\varepsilon_2 = \mathrm{Im}\ \varepsilon$ and τ the exciton life time equal to about 0.5 ns for CdS. As T, ε_2 and n are all dependent on σ equation (5) becomes a very complicated one stronly nonlinear in σ. The dependences of σ on I are numerically evaluated for CdS and represented in figure 3 which show the

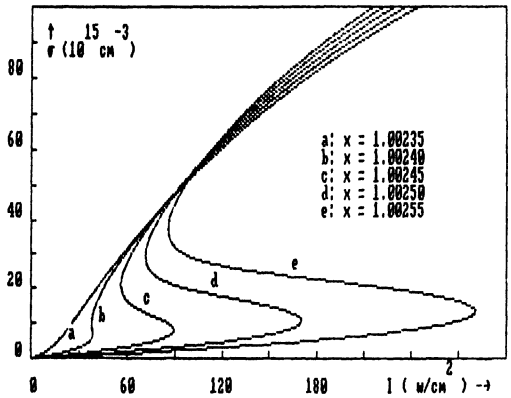

Fig.3 – Exciton density as functions of I.

occurrence of DB for $x \geq 1.0024$. The threshold intensity for the DB is of the order of about 40 w/cm^2. Figures 4 and 5 display α and n as functions of I for several values of x. According to figure

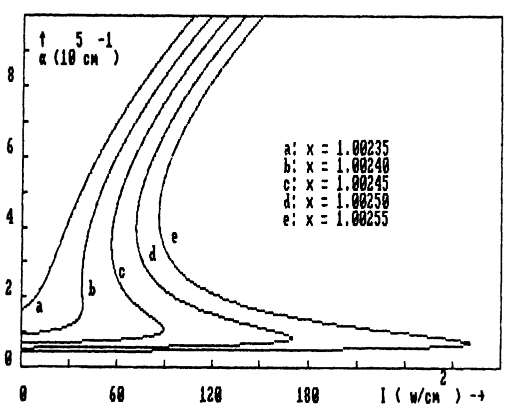

Fig.4.

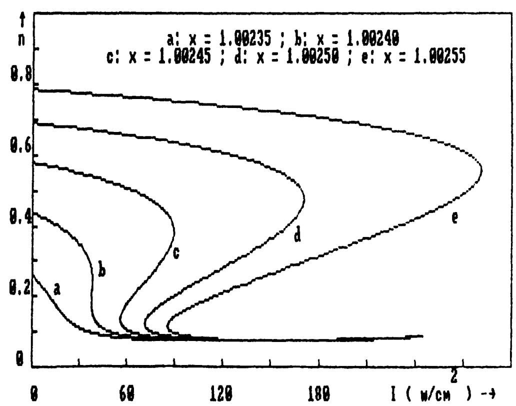

Fig.5.

Fig.4 – Absorption coefficient as functions of I.
Fig.5 – Refractive index as functions of I.

3 the behaviour of α and n is bistable for $x \geq 1.0024$, too. Note that $x \geq 1.0024$ means $(\omega - \omega_x) \geq$ 6.4 meV which is well smaller than $(E_g - \omega_x) = E^b = 32.9$ meV. This makes possible experimental measurements in the exciton resonance spectral region. Theoretical curves of CdS for the intensity of reflected beam $I_r = (1 - T)I$ *versus* the pumping intensity I are drawn in figure 6 as a possible reference for experimentalists.

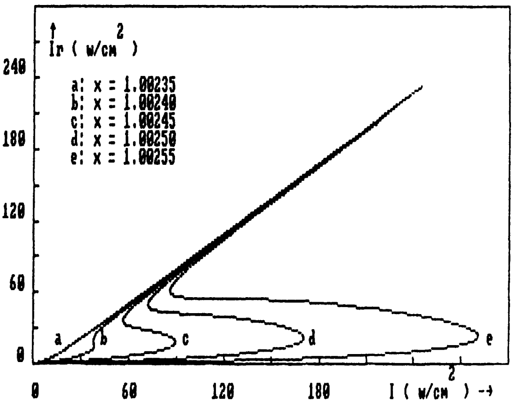

Fig.6 – Reflected intensity *vs.* incident intensity.

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