

ON BISTABLE POLARITON DISPERSION IN PHOTO-EXCITED CRYSTALS

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Within the Hartree-Fock approximation a possible bistable behaviour of polariton dispersion under photo-excitation is outlined using Hanamura's bosonic Hamiltonian.

Since the first observation of optical bistability (OB) in GaAs [1] and InSb [2] different mechanisms of optical nonlinearity [3-6] and feedback [7] have become the subject of intensive experimental and theoretical research. At moderate excitation and rather low temperature there exists a system of strongly interacting excitons in the crystal which behave as nonboson composite quasiparticles. These may generate giant nonlinearities such as a density-dependent shift and broadening of exciton levels as well as bleaching. Many authors [8-12] have made use of such nonlinearities to predict the occurrence of density bistability (DB) and OB. However, the polariton effect has been disregarded up to now in all above-cited works. As the polariton dispersion can be derived from the dielectric function it should be "multistable" if the latter is a multi-valued function of incident laser intensity. Additional waves and boundary problems of polaritons in nonlinear media are recently dealt with in refs. [13,14] and refs. [15,16], respectively, but how DB governs the polariton dispersion has not been considered anywhere. Also, we shall show anomalies in the reflectivity caused by the bistable polariton dispersion.

Since reliable evidence for the exciton level shift is missing any theoretical approximations serve as an orientation only. We herein use Hanamura's bosonic Hamiltonian [17] which in an effective manner can describe both dynamical and kinematical interactions between excitons coupled to an internal monochromatic classical radiation with the electric field strength $E_k(z,t)$:

$$H = \sum_p \omega_{xp} a_p^+ a_p + \frac{1}{2} \sum_{pq} w(pq) a_{l+q}^+ a_{p-q}^+ a_p a_l - du(k) \left(a_k^+ + a_{-k} + \sum_{pq} g(pq) [a_{k+q}^+ a_{p-q}^+ a_p + a_p^+ a_{p-q} a_{q-k}] E_k(z,t) \right), \tag{1}$$

where a_p destroys an exciton with wave-vector p and energy ω_{xp} ; d is the matrix element of the dipole moment; and w , u and g are the specific integrals of the appropriate exciton envelope functions (see ref. [17]). $E_k(z,t)$ in (1) itself obeys a Maxwell equation:

$$\left(\frac{\partial^2}{\partial z^2} - \epsilon_\infty \frac{\partial^2}{\partial t^2} \right) E_k(z,t) = V^{-1} \frac{\partial^2}{\partial t^2} \langle \Pi_k(z,t) \rangle, \tag{2}$$

where ϵ_∞ , V and Π are the background dielectric constant, the volume and the excitonic polarization operator.

$$\langle \Pi_k \rangle = du(k) \left([\langle a_k \rangle + \langle a_{-k}^+ \rangle] + \sum_{pq} g(pq) [\langle a_p^+ a_{p-q} a_{k+q} \rangle + \langle a_{q-k}^+ a_{p-q}^+ a_p \rangle] \right), \tag{3}$$

$\langle \rangle$ means the average over the eigenstate of H . Setting up the Heisenberg equations of motion for the averages present in (3) and solving them within the Hartree-Fock approximation for a macroscopic homogeneous and stationary situation we obtain the

complex dielectric function ϵ depending on both the incident laser frequency Ω and the intracrystal exciton density $n = V^{-1} \sum_p \langle a_p^+ a_p \rangle$:

$$\epsilon(\Omega, n) = \epsilon_\infty + \frac{\langle \Pi_k \rangle}{VE_k} = \epsilon_\infty - \frac{3e^2 E_g u_0^2 \mu^{-1} (1 - 2g_0 n)^2}{2(\omega_x + 2\omega_0 n - i\gamma) [\Omega^2 - (\omega_x + 2\omega_0 n - i\gamma)^2]} \quad (4)$$

In deriving (4) we neglected the k -dependence of w , u and g and used the following notations: $w_0 = Vw(000) = 26\pi E^b r^3/3$, $u_0 = u(0)V^{-1/2} = \pi^{-1/2} \times r^{-3/2}$ and $g_0 = -Vg(00) = 7\pi r^3$, where E^b , r , μ and γ are the exciton binding energy, the radius, the reduced mass and the phenomenological level damping and E_g is the band gap. Being created by photo-excitation and thanks to the finite life time τ excitons can reach a balance regime in which n is related to the incident laser intensity $I = |E|^2 |1 + \sqrt{\epsilon}|^2/4$ as follows:

$$I = \frac{n |1 + \sqrt{\epsilon(\Omega, n)}|^2}{4\tau\epsilon_2(\Omega, n)} \times \left\{ \frac{1}{2} [\epsilon_1(\Omega, n) + \sqrt{\epsilon_1^2(\Omega, n) + \epsilon_2^2(\Omega, n)}] \right\}^{1/2}, \quad (5)$$

where ϵ_1 and ϵ_2 are the real and imaginary parts of ϵ . Due to the n -dependence of ϵ , eq. (5) may possess more solutions n_i than one for fixed Ω and I yielding a so-called density multistability. Fig. 1 represents n as function of I for various $x = \Omega/\omega_x$ for CdS whose

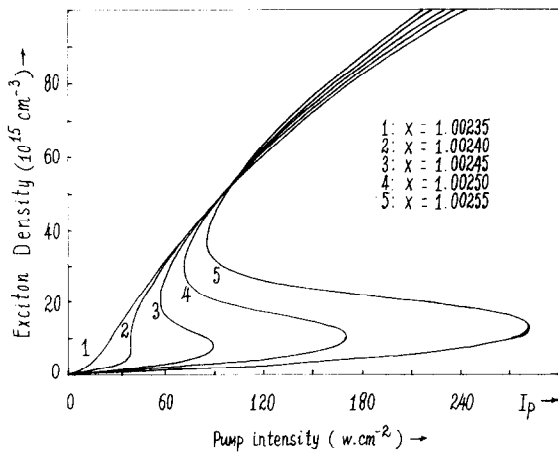


Fig. 1. Exciton density versus incident intensity.

parameters are $\epsilon_\infty = 7.6$, $m_e = 0.205m_0$, $m_h = 1.348m_0$ (m_0 is the free electron mass), $E_g = 2.5857$ eV, $\omega_x = 2.5528$ eV, $E^b = 32.9$ meV, $r = 25.5 \times 10^{-8}$ cm, $\gamma^{-1} = 7$ ps and $\tau = 0.5$ ns. We see that DB appears for $x \geq 1.0024$, i.e. $\Omega - \omega_x \geq 6.4$ meV, with a threshold intensity of about 40 W/cm². It is that DB that dictates the shape of the polariton dispersion which is determined by $\Omega^2 = k^2 \epsilon^{-1}(\Omega, n(I))$ and plotted in fig. 2 for three values $I = 0, 90$ and 150 W/cm². When I increases the whole polariton dispersion is lifted and for I exceeding the threshold value there appears a spectral region where one and the same frequency may correspond to either of three values (among them one unstable) of $\text{Re}(k)$ inside the crystal. This, in turn, must cause anomalies in the reflectivity at the boundary plane of a semi-infinite crystal. In fig. 3 we plot the reflective coefficient R versus x for the three values of I as in fig. 2. It is clear that in the $x \geq 1.0024$ region R might behave bistably if the crystal were pumped by a laser of proper intensity. Finally, as another variant, we display in fig. 4 R as a function of I for different x which again shows bistable behaviour of R for $x \geq 1.0024$.

Two remarks could be made: (1) As polariton theory has been quite successful in interpreting a lot of optical phenomena in unexcited media (see e.g. ref. [18]) it may be hoped that in the case of highly excited materials it will continue to be relevant for both theorists and experimentalists. (2) More realistic

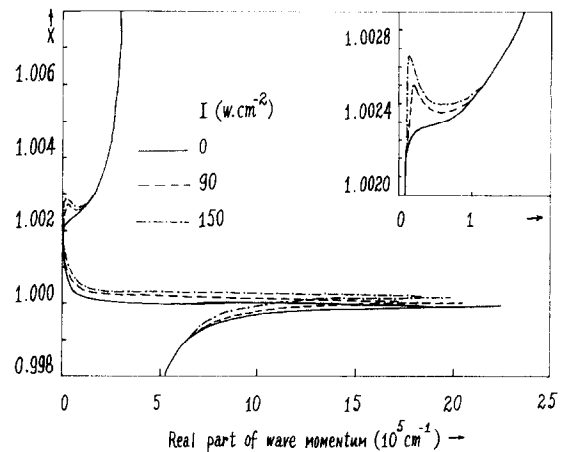


Fig. 2. Polariton dispersions for various intensities. The inset represents the enlarged region of x from 1.002 to 1.003 and $\text{Re } k$ from 0 to 2×10^5 cm⁻¹.

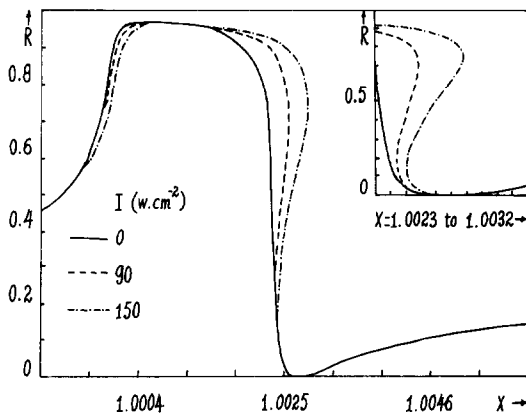


Fig. 3. Reflective coefficient versus normalized frequency for different intensities. The inset enlarges the region of x from 1.0023 to 1.0032.

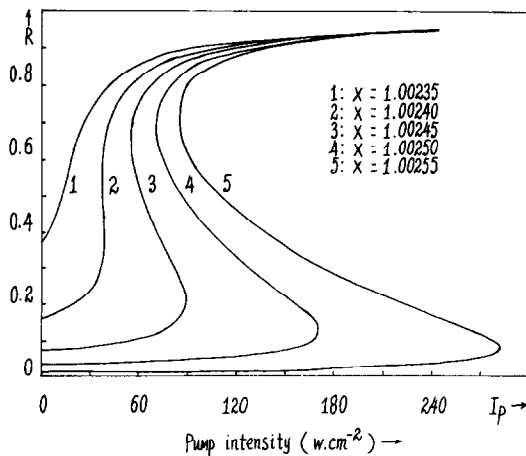


Fig. 4. Intensity-dependent reflective coefficient.

approximations should be restored to in order to account for the non-homogeneous and transient property [8,11], any excited states of the exciton including the scattering one [9], contributions of the

exciton correlation self-energy and excitation-induced broadening [10], etc. Other model calculations may result in another spectral region where bistability of polariton dispersion happens (e.g. the correlation correction [10] leads to a red shift (instead of a blue one) of the exciton level) but do not change the qualitative feature outlined above.

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