Anomalies of polariton dispersion in photo-excited molecular crystals

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Abstract. Green function and unitary transformation techniques are applied to study polariton formation in a molecular crystal placed in an intense external monochromatic light field. Using the exact tri-linear computation relations for Frenkel exciton operators, the energy of polaritons is shown to be dependent on exciton density. Then an effective Hamiltonian is constructed allowing one to obtain the light intensity-dependence of the exciton density. Due to possible bi- or multistability of the density, the calculated intensity-dependent polariton dispersion relations may exhibit different anomalies, such as the splitting of each polariton energy branch into two or several sub-branches and the appearance of supplementary intracrystal polariton-like waves even in the absence of exciton spatial dispersions.

1. Introduction

It is well known that photons and crystalline elementary excitations can mix to form coupled modes called polaritons. One of the most significant tasks in solving the polariton problem is to determine their dispersion relations. Since these relations can be derived rigorously by solving concrete interaction Hamiltonians, they provide the most physically satisfying basis not only for describing many optical effects, among them absorption (Nguyen *et al* 1979), luminescence (Tait and Weiher 1969, Benoit a la Guillaume *et al* 1970), light scattering (Ovander 1962, Nguyen *et al* 1980) and non-linear processes (Ovander 1965, Nguyen *et al* 1981), and also for predicting and interpreting new optical phenomena, such as, e.g. the wavevector dependent absorption occurring in 'weak level' crossing situations (Hopfield and Thomas 1965, Henry 1969) or the appearance of supplementary intracrystal waves due to spatial dispersions of crystalline excitations (Agranovich and Ginsburg 1966). Over the last decade, with the use of laser techniques, the research into highly photo-excited systems has attracted increasing attention. Under intense light the photon–crystalline quasiparticle mixing occurs in a collective manner, i.e. not a photon and a quasiparticle as under weak light but a number of them mix to generate 'collective' polaritons which are also referred to as giant (Haken and Schenzle 1972, 1973) or non-linear (Belkin *et al* 1979) polaritons. Many works have been devoted to the giant polariton. However, in all of these works only the dependence of the energy of the giant polariton on the density of crystalline quasiparticles is considered (see e.g. Inoue 1974, Liu 1983, Avdjugin *et al* 1983, Nguyen 1988a). The density of quasiparticles,

in fact, serves as an internal theoretical uncontrolled parameter, which, generally, can possess more than one stationary solution depending sensitively upon the frequency and intensity of external pumping light field (Toyozawa 1978, 1979). Thus, it would be desirable to have the giant polariton dispersion curves depending directly on frequency and intensity of incident light—the true experimentally controlled parameters. This is precisely the aim of the present paper, where the study is restricted to the case of molecular crystals with Frenkel excitons, being quasiparticles to be mixed with photons. As will be seen, the multistability of the exciton and intracrystal photon densities could cause anomalies in the giant polariton dispersion. A variety of optical anomalies in absorption and resonance scattering under intense light has been observed in CuCl (Nagasawa *et al* 1976, Mita and Ueta 1978) and explained by means of the above-mentioned population multistability (Toyozawa 1978, 1979). The influence of the population instability on the polariton effect, however, has not been considered until now.

We hereafter use the unit system with $\hbar = c = 1$.

2. Hamiltonians and exact commutation relations for Frenkel exciton operators

For simplicity and definiteness, we shall consider a three-dimensional molecular crystal with one molecule in a unit cell and assume that each molecule has only one excited state. Suppose a monochromatic light beam with wavevector q, frequency ω_q and electric field strength E_q of real amplitude A_q

$$E_q(t) = A_q(e^{i\omega_q t} + e^{-i\omega_q t}) \tag{1}$$

is incident onto a surface of the crystal. Let the beam propagating inside the crystal be characterised by its wavevector k. Then, thanks to Maxwell–Frenkel boundary conditions and disregarding the additional boundary conditions (Pekar 1958) and the existence of the dead-layer near the border surface (Ting *et al* 1975), one has

$$\omega_q = q = |q| = k/\sqrt{\varepsilon_\infty} = |k|/\sqrt{\varepsilon_\infty} = \Omega_k \tag{2}$$

where ε_∞ is the background dielectric constant of the crystal.

To apply the well-developed second quantisation method it is assumed that the light inside the crystal will be quantised, with the photon creation and annihilation operators denoted by c_k^+ and c_k. The coupling between the external driving field (1) and the intracrystal photon can be described by (Steyn–Ross and Gardiner 1983)

$$H_{\text{ext}}(t) = -(2\varepsilon_\infty V \Omega_k)^{1/2} A_q\, e^{i\omega_q t} c_k + \text{HC} \tag{3}$$

where V is the sample volume. If the light–matter interaction occurs via excitons which are generated by absorbing intracrystal photons, the Hamiltonian of a system of many interacting excitons and photons will be of the form

$$H_{xy} = (\Omega_k + B_k)c_k^+ c_k + \Omega_{xk} a_k^+ a_k + (Q_k/V)c_k^+ c_k^+ c_k c_k + (F_k/N)a_k^+ a_k a_k^+ a_k$$
$$+ R_k(a_k^+ c_k + c_k^+ a_k) \tag{4}$$

where $a_k^+(a_k)$ creates (destroys) an exciton with energy Ω_{xk}. It is different from the low excitation case, in that both photon–photon ($\propto Q_k$) and exciton–exciton ($\propto F_k$) collisions are involved in H_{xy}. $B_k = 2\pi e^2 N/mV\Omega_k$ with e and m being the charge and mass of a free electron respectively; $R_k = (2\pi N/V\Omega_k)^{1/2}\Omega_{xk}P_k$, where P_k is the photon–exciton transition dipole matrix element and N the total number of unit cells. Since the incident

light is assumed to be monochromatic and all relaxation and damping processes are neglected (they may be phenomenologically accounted for, say, in the work of Nguyen 1988a), the state inside the crystal will be that of coherent photons and excitons with the same wavevector k. That is the reason why the symbol Σ_k disappears in (4) and we can, for brevity, drop all the wavevector characters everywhere in what follows. The overall Hamiltonian of the problem under study, thus reads

$$H = H_{ext} + H_{xy}. \tag{5}$$

A very delicate matter to be emphasised is that photons are ideal bosons, but Frenkel excitons are not. There are different approaches to the non-bosonic nature of the exciton (see e.g. Agranovich 1968, Hanamura 1974). For us, that explored by Kaplan (1976) and Kaplan and Ruvinskii (1976) seems to be best suited to the treatment of many-exciton systems with arbitrary exciton density, because they have been successful in finding a closed and exact set of commutation relations for Frenkel exciton operators, although some of them are trilinear but not bilinear as usual, namely

$$[[a_p(t), a_q^+(t)], a_k(t)] = (2/N)a_{p+k-q}(t) \tag{6}$$

$$[[a_p(t), a_q^+(t)], a_k^+(t)] = -(2/N)a_{q+k-p}^+(t) \tag{7}$$

$$[a_p(t), a_q(t')] = [a_p^+(t), a_q^+(t')] = 0. \tag{8}$$

Now we are going to study the intensity-dependence of the dispersion relations of the eigenmodes of the coupled photon–exciton system subjected to excitement by the external light field (1). To do this we shall follow two stages:

(i) We anticipate that the whole system is approaching its steady state with certain stationary exciton and photon densities corresponding to given frequency and intensity of exciting incident light. Then, we could forget H_{ext} for the time being and handle only H_{xy} to find the giant-polariton dispersion relations, which, as a rule, should contain exciton and photon densities as theoretical parameters (see § 3).

(ii) We use the overall Hamiltonian $H = H_{ext} + H_{xy}$ to determine the dependence of the above-mentioned theoretical parameters on the experimentally controlled parameters—the frequency and intensity of the incident light (§ 4).

By combining the results of the two stages we shall have the desired polariton dispersion curves which may exhibit anomalies with respect to those of the usual linear polaritons (§ 5).

3. Density-dependent polariton energy

We define the following retarded Green functions

$$G_1(t) = \theta(t)\langle[a(t), a^+(0)]\rangle \tag{9}$$

$$G_2(t) = \theta(t)\langle[c(t), a^+(0)]\rangle \tag{10}$$

$$G_3(t) = \theta(t)\langle[a^+(t)a(t)a(t), a^+(0)]\rangle \tag{11}$$

$$G_4(t) = \theta(t)\langle[a(t)a^+(t)a(t), a^+(0)]\rangle \tag{12}$$

$$G_5(t) = \theta(t)\langle[a^+(t)a(t)c(t), a^+(0)]\rangle \tag{13}$$

$$G_6(t) = \theta(t)\langle[c^+(t)c(t)c(t), a^+(0)]\rangle \tag{14}$$

where $\langle \ldots \rangle \equiv \langle \ldots \rangle_{H_{xy}}$ means the average over the eigenstate of H_{xy} and $\theta(t)$ is the step function. Taking into account the commutators (6)–(8) we can determine the time-derivatives of $G_1(t)$ and $G_2(t)$ which are expresed in terms of $G_3(t), \ldots, G_6(t)$

$$\dot{G}_1(t) = [1 - 2(N_x/N)]\delta(t) - i\Omega_x G_1(t) - iRG_2(t) + (i/N)(2\Omega_x - F)G_3(t)$$

$$- i(F/N)G_4(t) + (2iR/N)G_5(t) \tag{15}$$

$$\dot{G}_2(t) = -i(\Omega + B)G_2(t) - iRG_1(t) - (2iQ/V)G_6(t) \tag{16}$$

where

$$N_x = \langle \hat{N}_x \rangle \equiv (N/2)\{\langle [a^+(0), a(0)] \rangle + 1\} \tag{17}$$

is the total exciton number. The specific form of the total exciton number operator $\hat{N}_x$ as seen from (17) is due to the non-bosonic nature of excitons (see Kaplan 1976). Now going to the energy representation by means of the transformation

$$G_i(t) = \frac{1}{2\pi} \int G_i(E) \exp(-iEt)\, dE \tag{18}$$

we get from (15)–(16)

$$(E - \Omega_x)G_1(E) = i[1 - 2(N_x/N)] + RG_2(E) + (1/N)(F - 2\Omega_x)G_3(E)$$

$$+ (F/N)G_4(E) - 2(R/N)G_5(E) \tag{19}$$

$$(E - \Omega - B)G_2(E) = RG_1(E) + 2(Q/V)G_6(E). \tag{20}$$

To solve (19) and (20) for $G_1(E)$ and $G_2(E)$ we have to set up equations of motion also for $G_3(E), \ldots, G_6(E)$, which, in turn, will involve further new unknown functions, say, $G_7(E)$, $G_8(E)$, and so on. In what follows we resort to the well-known random-phase approximation (RPA) which formally gives us in the case of coherent excitons

$$G_3(E), G_4(E) \to N_x G_1(E) \tag{21}$$

$$G_5(E) \to N_x G_2(E) \qquad G_6(E) \to N_y G_2(E) \tag{22}$$

with $N_y = \langle c^+ c \rangle$ being the total number of internal photons. With (21) and (22), (19) and (20) become

$$\{E - \Omega_x[1 - (2n_x/n)] - (2n_x/n)F\} G_1(E) + R[(2n_x/n) - 1]G_2(E)$$

$$= i[1 - (2n_x/n)] \tag{23}$$

$$-RG_1(E) + (E - \Omega - B - 2Qn_y)G_2(E) = 0 \tag{24}$$

where the corresponding densities are $n = N/V$, $n_x = N_x/V$ and $n_y = N_y/V$. From (23) and (24) we immediately obtain

$$G_1(E) = i(1 - 2n_x/n)(E - \Omega - B - 2Qn_y)/\{(E - \Omega_x(1 - 2n_x/n) - 2Fn_x/n)$$

$$\times (E - \Omega - B - 2Qn_y) - R^2(1 - 2n_x/n)\} \tag{25}$$

$$G_2(E) = iR(1 - 2n_x/n)/\{(E - \Omega_x(1 - 2n_x/n) - 2Fn_x/n)(E - \Omega - B - 2Qn_y)$$
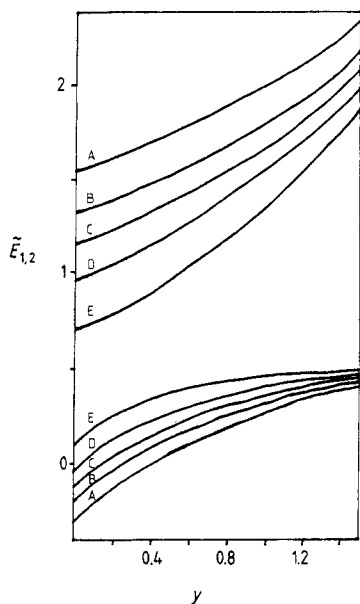
$$- R^2(1 - 2n_x/n)\}. \tag{26}$$

Figure 1. $\tilde{E}_{1,2} = E_{1,2}\Omega_x^{-1}$ versus $y = K\varepsilon_\infty^{-1/2}\Omega_x^{-1}$. Curve A, $\rho_x = 0$; curve B, $\rho_x = 0.3$; curve C, $\rho_x = 0.5$; curve D, $\rho_x = 0.7$; curve E, $\rho_x = 0.9$.

Obviously, $G_1(E)$ and $G_2(E)$ possess two common poles determining the dispersion laws of the two energy branches of giant polaritons

$$E_{1,2}(k) = \tfrac{1}{2}[\![\Omega_{xk} + (F_k - \Omega_{xk})\rho_x + k/\varepsilon_\infty^{1/2} + B_k + nQ_k\rho_\gamma$$
$$\pm \{[\Omega_{xk} + (F_k - \Omega_{xk})\rho_x - k/\varepsilon_\infty^{1/2} - B_k - nQ_k\rho_\gamma]^2$$
$$+ 4R_k^2(1 - \rho_x)\}^{1/2}]\!] \tag{27}$$

where the relative densities $\rho_x = 2n_x/n$ and $\rho_\gamma = 2n_\gamma/n$ are introduced for convenience. As in other works, we have got the giant polariton dispersion relations depending on ρ_γ and ρ_x. However, the expressions for $E_{1,2}$ derived here seem to be more general and correct. In all other works, for example, Q was put equal to zero, while in the work of Liu (1983) the multiplier of the term $\propto 4R^2$ was $(1 - \rho_x)^2$ instead of $(1 - \rho_x)$ that should be incorrect (see Nguyen 1988b, c). Neglecting the k-dependence of Ω_{xk}, F_k, Q_k, P_k and putting $\Omega_k = \Omega_{x0}$ in B_k and R_k we can plot in figure 1 the polariton dispersion curves for various values of ρ_x. The data used for plotting are (Agranovich 1968) $\Omega_x \simeq 5$ eV, $n \simeq 10^{22}$ cm^{-3}, $F \simeq 2.5$ eV, $Q = 0$ and $|P| \simeq 2e \times 10^{-8}$ cm. From figure 1 it is clear that the upper polariton branch E_2 shifts down and the lower one E_1 rises for increasing ρ_x. Negative values of E_1 for small $y = k\varepsilon_\infty^{1/2}\Omega_x^{-1}$ are caused by the fact that antiresonant terms were ignored in writing $H_{x\gamma}$.

Before going on to the next section note that such correct expressions for $E_{1,2}$ as in (27) could also be derived by a Bogolubov diagonalisation method, if instead of $H_{x\gamma}$ one used the following effective Hamiltonian $\tilde{H}_{x\gamma}$

$$\tilde{H}_{x\gamma} = \tilde{\Omega}c^+c + \tilde{\Omega}_x a^+a + \tilde{R}(a^+c - c^+a) \tag{28}$$

$$\tilde{\Omega} = \Omega + B + nQ\rho_\gamma \tag{29}$$

$$\tilde{\Omega}_x = \Omega_x + (F - \Omega_x)\rho_x \tag{30}$$

$$\tilde{R} = R(1 - \rho_x)^{1/2}. \tag{31}$$

The advantage of using $\tilde{H}_{xy}$ is that in it both photon and exciton operators are ideally bosonic. In the work of Liu (1983) an *ad hoc* effective Hamiltonian H^{eff} was constructed so that the correct equations of motion for operators a and c could be obtained and then such an H^{eff} should be made diagonal yielding the giant polariton energy. Unfortunately, the H^{eff} of Liu, being Hermitian, is not self-consistent: equation (16b) of Liu (1983) cannot be derived from his H^{eff} as is required. To meet the purpose pursued in the work of Liu (1983) the effective Hamiltonian must be non-Hermitian as has been provided by Nguyen (1988c) and then the polariton dispersion curves may be drawn by a step-by-step Bogolubov transformation method developed by Nguyen (1988b). The effective Hamiltonian written in (28) is Hermitian because here we require it to be such that the correct dispersion laws (27) (but not the equations of motion for Green functions (23) and (24)) should be obtained. In § 4 $\tilde{H}_{xy}$ is used to derive the dependence of ρ_γ and ρ_x upon the frequency and the intensity $I = 2\varepsilon_\infty A^2$ of the incident light.

4. Density multistability

As was stated in § 2, we are dealing with the coherent exciton–photon state. Let the averaged value over the coherent state be denoted by $\langle\!\langle \ldots \rangle\!\rangle \equiv \langle \ldots \rangle_H$, then according to Glauber (1966) and Bogolubov (1971) we have

$$\langle\!\langle a \rangle\!\rangle = N_x^{1/2} \qquad \langle\!\langle c \rangle\!\rangle = N_\gamma^{1/2}. \tag{32}$$

Averaged values of such a kind are termed averaging over the coherent state or quasi-averaging. It can be proved that if the Hamiltonian H conserves total number of quasiparticles, then usual averaged values $\langle a \rangle \equiv \langle a \rangle_H$ exactly equal to zero. To achieve non-zero averaged values it is necessary to have Hamiltonians changing the total number of quasiparticles. In our problem, the overall Hamiltonian $\tilde{H} = H_{ext} + \tilde{H}_{xy}$ is exactly the required one with H_{ext} as a term changing the total number of excitons and photons. To eliminate the explicit time-dependence in H_{ext}, we cast $\tilde{H}$ into a rotating frame of reference as below

$$\tilde{H} = \mathbb{H} = (\tilde{\Omega} - \omega)c^+c + (\tilde{\Omega}_x - \omega)a^+a + \tilde{R}(a^+c + c^+a) + \tilde{A}(c^+ + c) \tag{33}$$

where $\tilde{A} = -(2\varepsilon_\infty V\Omega)^{1/2}A$. By means of a unitary transformation

$$\mathbb{H} \rightarrow \tilde{\mathbb{H}} = S^+\mathbb{H}S \tag{34}$$

with

$$S = \exp[\mu(c^+ - c) + \nu(a^+ - a)] \tag{35}$$

and real coefficients μ and ν defined by

$$\nu = \tilde{R}\tilde{A}[(\tilde{\Omega} - \omega)(\tilde{\Omega}_x - \omega) - R^2]^{-1} \tag{36}$$

$$\mu = -(\tilde{\Omega}_x - \omega)\tilde{A}[(\tilde{\Omega} - \omega)(\tilde{\Omega}_x - \omega) - R^2]^{-1} \tag{37}$$

we can get $\tilde{\mathbb{H}}$ in the form

$$\tilde{\mathbb{H}} = (\tilde{\Omega} - \omega)c^+c + (\tilde{\Omega}_x - \omega)a^+a + \tilde{R}(a^+c + c^+a) + \mu^2(\tilde{\Omega} - \omega) + \nu^2(\tilde{\Omega}_x - \omega)$$
$$+ 2\mu\nu\tilde{R} + 2\mu\tilde{A}. \tag{38}$$

We see, in contrast to $\tilde{\mathsf{H}}$, that $\check{\mathsf{H}}$ conserves the total number of quasiparticles. Now, if the eigenstates of H and $\check{\mathsf{H}}$ are $|\Phi\rangle$ and $|\check{\Phi}\rangle = S^+|\Phi\rangle$, respectively, we shall have

$$N_x^{1/2} = \langle\langle a\rangle\rangle = \langle a\rangle_{\mathsf{H}} \equiv \langle\Phi|a|\Phi\rangle = \langle\Phi|SS^+aSS^+|\Phi\rangle = \langle\check{\Phi}|S^+aS|\check{\Phi}\rangle$$

$$= \langle S^+aS\rangle_{\check{\mathsf{H}}} = \langle a\rangle_{\check{\mathsf{H}}} + \nu \tag{39}$$

$$N_y^{1/2} = \langle\langle c\rangle\rangle = \ldots = \langle c\rangle_{\check{\mathsf{H}}} + \mu. \tag{40}$$

Since $\langle a\rangle_{\check{\mathsf{H}}} = \langle c\rangle_{\check{\mathsf{H}}} = 0$, (39) and (40) give us $N_x = \nu^2$ and $N_y = \mu^2$, which can be rewritten as follows

$$\rho_x = [\![2\Omega\Omega_x R^2(1-\rho_x)\check{I}/\{[(B+nQ\rho_y)f+R^2]\rho_x+(B+nQ\rho_y)\Delta-R^2\}^2]\!] \tag{41}$$

$$\rho_y = [\![2\Omega\Omega_x(\Delta+f\rho_x)^2\check{I}/\{[(B+nQ\rho_y)f+R^2]\rho_x+(B+nQ\rho_y)\Delta-R^2\}^2]\!] \tag{42}$$

where $\check{I} = I(n\Omega_x)^{-1}$, $f = F - \Omega_x$ and $\Delta = \Omega_x - \omega$. Dividing (42) by (41) we have ρ_y expressed only in terms of ρ_x, namely

$$\rho_y = \rho_x(\Delta + f\rho_x)^2(1-\rho_x)^{-1}R^{-2}. \tag{43}$$

Insert (43) into (41) and then recast the latter into the form

$$\check{I} = [\rho_x/2\Omega\Omega_x R^6(1-\rho_x)^3]\{2nQ(\Delta+f\rho_x)^3\rho_x$$

$$+ R^2(1-\rho_x)[B(\Delta+f\rho_x)-R^2(1-\rho_x)]\}^2 \tag{44}$$

which reveals that under certain conditions one value of $\check{I}$ might correspond to several values of ρ_x (and ρ_y through (43)), indicating the occurrence of density multistability. Since the photon–photon scattering is, in fact, very weak, we can henceforth put $Q = 0$. Then (44) becomes

$$\check{I} = \beta\rho_x(\rho_x+\alpha)^2(1-\rho_x)^{-1} \tag{45}$$

where

$$\alpha = [4z(1-y)-u^2]/[4z(x-1)+u^2]$$

$$\beta = [4z(x-1)+u^2]^2/yu^2 \tag{46}$$

$$x = F\Omega_x^{-1} \qquad z = B\Omega_x^{-1} \qquad u = 2R\Omega_x^{-1} \tag{47}$$

are normalised dimensionless quantities introduced for convenience in numerical calculations. Equation (45) is a simplified form of the more general one treated in the works of Nguyen (1986, 1988a) according to whose results density bistability would occur if the following inequality is met:

$$-9 < 8\alpha < 0. \tag{48}$$

5. Polariton dispersion anomalies

We have shown in § 3 that polariton dispersion curves are subject to changes in their shapes depending on how highly the crystal is excited, i.e. $E_{1,2}$ depend on ρ_x and ρ_y. On the other hand, from § 4 follows that ρ_x and ρ_y, themselves, are governed by I and ω. Generally speaking, for fixed I and ω there may be more than one corresponding pair of values of ρ_x and ρ_y, so we can write $\rho_j^i \equiv \rho_j^i(I, \omega)$, where $i = 1, 2, 3, \ldots$ and $j = x$ or γ. Inserting the multivalued densities $\rho_j^i(I, \omega)$ into $E_{1,2}(k, \rho_j^i)$, one would get anomalous
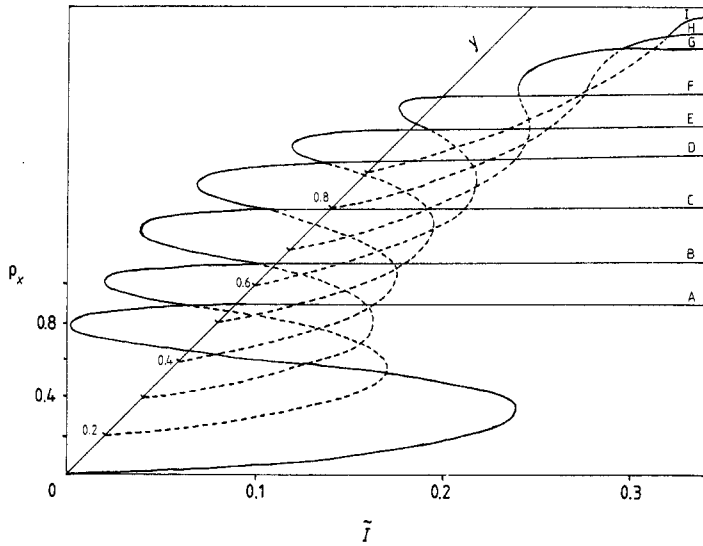
Figure 2. ρ_x versus $\tilde{I} = I(n\Omega_x)^{-1}$. Curve A, $y = 0.1$; curve B, $y = 0.2$; curve C, $y = 0.3$; curve D, $y = 0.4$; curve E, $y = 0.5$; curve F, $y = 0.6$; curve G, $y = 0.7$; curve H, $y = 0.8$; curve I, $y = 0.9$.
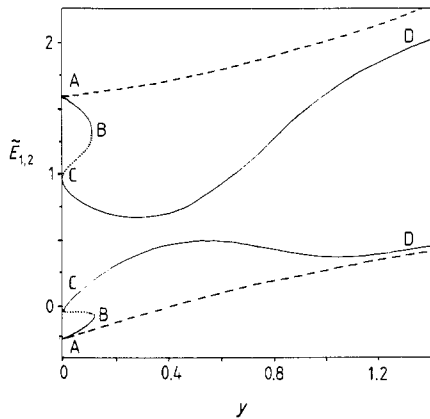

Figure 3. Polariton dispersion curves for $\tilde{I} = 0$ (broken curves) and $\tilde{I} = 0.15$ (full curves). The dotted lines are unstable.
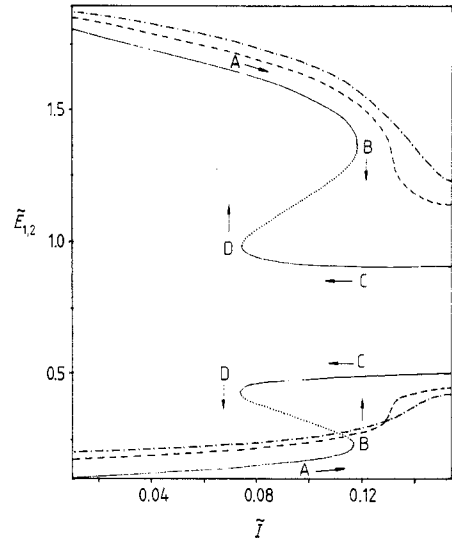

Figure 4. $\tilde{E}_{1,2}$ versus $\tilde{I}$ for $y = 0.6$ (full curve), 0.75 (chain curve) and 0.8 (broken curve). Unstable states are shown as dotted curves.

curves for the dispersion of the giant polariton which will be displayed in figures 3 and 4 below. Figure 2 represents ρ_x as a function of $\tilde{I}$ for various values of y. We see that for $y \geqslant y_c \simeq 0.75$ ρ_x is a monotonically increasing function of $\tilde{I}$, while for $0 < y < y_c$ the $(\tilde{I} - \rho_x)$-correspondence is one-to-three. The larger y the narrower the interval of $\tilde{I}$ between which density bistability takes place. In figure 3 we draw dispersion curves of

the polariton for a fixed $\bar{I} = 0.15$. For comparison, those for the unexcited case $\bar{I} = 0$ are also plotted in it. Figure 3 shows, at least, two kinds of anomalies: (i) Each polariton energy branch splits into two sub-branches—the solid lines AB and CD—because the dotted lines BC correspond to unstable solutions of ρ_x (Toyozawa 1978). (ii) For a fixed $\bar{I}$ one incident light with a certain frequency ω might generate inside the crystal more than one polariton wave of same frequency, but different refractive indices, i.e. different wavevectors (see figure 3). Note that the origin of the above-mentioned supplementary intracrystal polariton waves comes from the intensity-dependence of the polariton dispersion relations, but not from the spatial dispersion of excitons as it is already well-known (Agranovich and Ginsburg 1966). Finally, the explicit $\bar{I}$-dependence of the polariton energy is represented in figure 4 for three values of y. For certain values of y (say, $y = 0.6$) the anomalous property may manifest itself in splitting two polariton branches into four sub-branches (full curves in figure 4). When $\bar{I}$ is increasing and then decreasing, the hysteresis effect of polariton energy will occur, but the hysteresis loop $A \rightarrow B \rightarrow C \rightarrow D \rightarrow A$ of the upper branch goes clockwise, while that of the lower one goes counter-clockwise.

6. Conclusions

We have dealt with possible anomalies associated with the intensity-dependence of the energy of the giant polariton within the simplest model. We wonder could any predicted effects in fact be checked experimentally? If such anomalies as demonstrated in figures 3 and 4 were responsible, they must influence all optical resonant processes under intense light when the photon–exciton interaction is of importance. Hopefully, this paper can aid experimentalists in measuring such processes. Of course, to be in possible agreement with observations, one has to generalise the theoretical model to account for the dephase and depopulation times of both internal photons and excitons as well as the spatial dispersion of the latter, etc.

Acknowledgments

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