Optical nonlinearity of ionic crystals and its increasing absorption optical bistability

1996 J. Phys.: Condens. Matter 8 6695

(http://iopscience.iop.org/0953-8984/8/36/020)

J. Phys.: Condens. Matter **8** (1996) 6695–6703. Printed in the UK

Optical nonlinearity of ionic crystals and its increasing absorption optical bistability

Bao-Hua Wei†‡§, Fa Ou†, K W Yu‡ and Tin-Wan Wu†

† Department of Applied Physics, South China University of Technology, Guangzhou 510641, People's Republic of China

† Department of Physics, The Chinese University of Hong Kong, Shatin, New Territories, Hong Kong

§ International Centre for Material Physics, Shengyang 110015, People's Republic of China

Received 29 March 1996, in final form 28 May 1996

Abstract. This paper presents an approach to the intracavity interaction of electromagnetic waves with anharmonic lattice vibrations of ionic crystals. The nonlinear lattice dynamics and an expression for the nonlinear polarization are derived in terms of optical phonon modes. In the rotating-wave approximation the total coherent Hamiltonian of the system is derived. Using the corresponding equation of steady states, the increasing absorption optical bistability (IAOB), as a nonlinear effect, is demonstrated. The results show that optical nonlinearities due to the interaction of photons with various elementary excitations of bosonic type such as phonons and excitons may be the origin of IAOB, which is consistent with our previous works.

1. Introduction

The study of optical bistability and multistability in optical nonlinear systems has been a subject of interest over the last two decades. In the past, most of the optical systems concerned were based on the light–matter interaction and involved using the two-level atomic model or other atomic models which possess saturated absorptive characteristics [1–11]. In recent years, a new kind of optical bistability—increasing absorption optical bistability (IAOB)—has aroused great interest in scientists [12–15]. Since the dynamical behaviour of IAOB is substantially different from saturated absorption optical bistability (SAOB), we must update our knowledge to treat this new subject. In recent years, we have proposed a dynamical model for the IAOB, which is just like the Maxwell–Bloch equations for saturated absorptive nonlinearity including the SAOB [12–14]. On the basis of this IAOB model, we have suggested that there exist some kinds of elementary excitation, which obey the boson statistics, in optical nonlinear systems; IAOB will arise from the interaction of photons with these boson elementary excitations including the electron–hole pair exciton in semiconductors.

This paper attempts to provide an example to justify the use of the Hamiltonian of the IAOB model mentioned above, which is obtained in a phenomenological way. It will be demonstrated that in addition to the exciton in semiconductors [16], the phonon of anharmonic lattice vibration in ionic crystals may play the role of the boson elementary excitation with which the light interacts. The layout of the paper is follows: section 2 presents the Hamiltonian of the ionic crystal; the nonlinear polarization of the crystal will be discussed in section 3; section 4 gives the Hamiltonian of the ionic crystal coupled with

0953-8984/96/366695+09$19.50

an electromagnetic field; and the semiclassical equations of motion and the IAOB will be given in section 5; finally a concise discussion is presented in section 6.

2. The Hamiltonian of the crystal

Using the normal coordinate Q and its conjugate momentum P, the Hamiltonian of anharmonic motion of the crystal can be written as [17]

$$
\begin{aligned}
H_{crystal} = {} & \frac{1}{2}\sum_{\alpha}(P_\alpha^2 + \omega_\alpha^2 Q_\alpha^2) + \frac{1}{2}\sum_{\alpha\beta\gamma}\mu_{\alpha\beta\gamma}(P_\alpha P_\beta Q_\gamma + (Q_\alpha P_\beta P_\gamma) \\
& + \frac{1}{3}\sum_{\alpha\beta\gamma}\eta_{\alpha\beta\gamma}Q_\alpha Q_\beta Q_\gamma + \frac{1}{2}\sum_{\alpha\beta\gamma\delta}\lambda_{\alpha\beta\gamma\delta}(P_\alpha P_\beta Q_\gamma Q_\delta + Q_\delta Q_\gamma P_\beta P_\alpha) \\
& + \frac{1}{4}\sum_{\alpha\beta\gamma\delta}\epsilon_{\alpha\beta\gamma\delta}Q_\alpha Q_\beta Q_\gamma Q_\delta
\end{aligned}
\tag{1}
$$

where we retain only the nonlinear terms of the first order (μs and ηs) and the second order (λs and ϵs) which will be adopted in later discussions. In order to ensure the hermiticity of the Hamiltonian after the quantization, every product of P and Q is added to its cyclic permutation in the round brackets of the second and fourth summations in equation (1).

The quantized normal coordinates and momenta obey the following commutation relations:

$$
[Q_\alpha, P_\beta] = \mathrm{i}\hbar\delta_{\alpha\beta} \qquad [Q_\alpha, Q_\beta] = [P_\alpha, P_\beta] = 0. \tag{2}
$$

The annihilation and creation operators of the phonon mode, b_α and b_α^+, are introduced by the following transformation:

$$
Q_\alpha = \left(\frac{\hbar}{2\omega_\alpha}\right)^{1/2}(b_\alpha + b_\alpha^+) \qquad P_\alpha = \frac{1}{\mathrm{i}}\left(\frac{\hbar\omega_\alpha}{2}\right)^{1/2}(b_\alpha - b_\alpha^+) \tag{3}
$$

and they satisfy the boson commutation relations

$$
[b_\alpha, b_\beta^+] = \delta_{\alpha\beta} \qquad [b_\alpha, b_\beta] = [b_\alpha^+, b_\beta^+] = 0. \tag{4}
$$

Inside the Hamiltonian of equation (1), the linear terms represent the independent motion of the different modes, while the nonlinear terms represent the interaction between those modes. The excitation of the phonon mode may be either coherent (for example, the mode excited by the coherent electromagnetic field) or incoherent (for example, the thermal excitation); therefore, it is necessary to divide the Hamiltonian $H_{crystal}$ into coherent (reversible) and incoherent (irreversible) parts, i.e.,

$$
H_{crystal} = H_{crystal}^{coh} + H_{crystal}^{incoh}. \tag{5}
$$

The irreversible part plays the role of the reservoir of coherent phonon modes. Hereafter, for the sake of convenience, we shall consider only one kind of optical phonon mode coupled with a single photon mode, and assume that the frequency of the optical phonon mode (ω_b), which is coherently excited, is nearest to that of the cavity mode of the photon (ω_a), so that $H_{crystal}^{coh}$ can be written as

$$
\begin{aligned}
H_{crystal}^{coh} = {} & \frac{1}{2}(P^2 + \omega_b^2 Q^2) + \frac{1}{2}\mu(P^2 Q + Q P^2) + \frac{1}{2}\lambda(P^2 Q^2 + Q^2 P^2) \\
& + \frac{1}{3}\eta Q^3 + \frac{1}{4}\epsilon Q^4.
\end{aligned}
\tag{6}
$$

Substituting b and b^+ from equation (3) for P and Q, and then making the rotating-wave approximation (i.e., neglecting the products in which the numbers of appearances of b and of b^+ are unequal, e.g., bb^+b, $b^{+2}bb^+$), we can get the further simplified $H_{crystal}^{coh}$:

$$H_{crystal}^{coh} = \hbar\omega_b b^+ b + \hbar g_b''(b^{+2}b^2 + b^2 b^{+2}) \tag{7}$$

where $g_b'' = (\hbar/4)(\lambda + 3\epsilon/4\omega_b^2)$ is the phonon–phonon coupling constant (the double primes indicate that the second-order nonlinearity is considered). As a result of the rotating-wave approximation, all of the first-order nonlinear terms are eliminated. The same will be found below for the Hamiltonian of the photon–phonon interaction.

3. Nonlinear polarization of the ionic crystal

The ionic crystal is directly and strongly coupled with the electromagnetic field through its macroscopic polarization, which is defined by

$$\boldsymbol{P} = \sum_{l,r} q_r(\boldsymbol{R}_l + \boldsymbol{u}_{lr}) \qquad l = 1, 2, \ldots, N \qquad r = 1, 2, \ldots, n \tag{8}$$

where $\boldsymbol{R}_l$ stands for the lattice coordinate vector of the lth cell and we assume that every unit cell contains only one pair of positive and negative ions (i.e. $n = 2$) and the charge of ions is $q_r = \pm e$, so that

$$\boldsymbol{P} = \sum_l e(\boldsymbol{u}_{l+} - \boldsymbol{u}_{l-}) = e\sum_l \boldsymbol{u}_l = e\sum_l u_{sl}\boldsymbol{l}_s \tag{9}$$

where $\boldsymbol{u}_l$ is the vector of the relative displacement between the two ions in the unit cell and $\boldsymbol{l}_s$ is the unit vector of the cartesian coordinates. The component of the polarization vector can be expressed in terms of the optical phonon modes as follows:

$$P_s = \sum_\alpha \xi_{s,\alpha} Q_\alpha + \sum_{\alpha\beta} \xi'_{s,\alpha\beta} Q_\alpha Q_\beta + \sum_{\alpha\beta\gamma} \xi''_{s,\alpha\beta\gamma} Q_\alpha Q_\beta Q_\gamma + \cdots \tag{10}$$

where

$$\xi_{s,\alpha} = e\sum_l A_{sl,\alpha} \qquad \xi'_{s,\alpha\beta} = \frac{e}{2}\sum_l B_{sl,\alpha\beta} \qquad \xi''_{s,\alpha\beta\gamma} = \frac{e}{3}\sum_l C_{sl,\alpha\beta\gamma} \tag{11}$$

and A, B and C are transformation coefficients, which give the relations between the normal-mode (coordinate) and cartesian coordinates as

$$u_i = \sum_\alpha A_{i\alpha} Q_\alpha + \frac{1}{2}\sum_{\alpha\beta} B_{i\alpha\beta} Q_\alpha Q_\beta + \frac{1}{3}\sum_{\alpha\beta\gamma} C_{i\alpha\beta\gamma} Q_\alpha Q_\beta Q_\gamma + \cdots. \tag{12}$$

So expression (10) can be considered as a normal-mode transformation of the macroscopic polarization, which may be induced by the electromagnetic field. On the other hand, it tells us that the polarization, in general, is nonlinear in its normal modes; the coherently induced polarization P_s is simply written as

$$P_s = \xi_s Q + \xi_s' Q^2 + \xi_s'' Q^3. \tag{13}$$

4. The total Hamiltonian of the ionic crystal coupled with an electromagnetic field

It is further assumed that the wavelength of the incident electromagnetic field is much greater than the thickness of the crystal film, so that the long-wave approximation may be made. The electric field $\boldsymbol{E}$ in the film (cavity) will be approximately homogeneous, and the electromagnetic field is polarized in the direction parallel to the surface of the film. Then, the coherent Hamiltonian of photon–phonon interaction in the dipole approximation can be expressed in the following form:

$$H^{coh}_{photon-phonon} = -\boldsymbol{P}\cdot\boldsymbol{E} = -(\xi Q + \xi' Q^2 + \xi'' Q^3)E \tag{14}$$

where the direction of the $\boldsymbol{E}$-vector coincides with one of the coordinate axes and the component index 's' is omitted. In equation (14),

$$E = \frac{1}{\mathrm{i}}\left(\frac{\hbar\omega_a}{2}\right)^{1/2}(a - a^+). \tag{15}$$

and

$$Q = \left(\frac{\hbar}{2\omega_b}\right)^{1/2}(b + b^+). \tag{16}$$

As we stated above, the frequency of the excited phonon ω_b is very near to that of the photon ω_a, so $H^{coh}_{photon-phonon}$ can also be treated by using the rotating-wave approximation (RWA). After some algebraic calculation, we obtain

$$H^{coh}_{photon-phonon} = \mathrm{i}\hbar(g_{ab}ab^+ + g''_{ab}ab^{+2}b) + \text{HC} \tag{17}$$

where g_{ab} and g''_{ab} are the photon–phonon coupling constant (double primes stand for the second-order coupling), which are given by

$$\begin{aligned} g_{ab} &= \frac{1}{2}\sqrt{\omega_a/\omega_b}(\xi + \xi''(3\hbar/2\omega_b)) \\ g''_{ab} &= \frac{3\hbar}{4}\xi''\omega_a^{1/2}/\omega_b^{3/2}. \end{aligned} \tag{18}$$

Finally, combining equation (17) with equation (7) and taking the driving term of the incident electromagnetic field into account, we obtain the total coherent Hamiltonian of the system as

$$H^{coh}_{total} = \hbar\Delta_a a^+a + \hbar\Delta_b b^+b + \mathrm{i}\hbar(g_{ab}ab^+ + g''_{ab}ab^{+2}b^2 - \mathrm{i}\, g''_b b^{+2}b^2 + ga^+f) + \text{HC} \tag{19}$$

where f and ω are the amplitude and the frequency of the incident light, respectively, $\Delta_a = \omega_a - \omega$ and $\Delta_b = \omega_b - \omega$ are the detuning of the photon and phonon, respectively, and g is the relevant coupling constant for the coupling of the incident light with the system [16].

The incoherent part of the Hamiltonian—the interaction between the photon and the optical phonon with the respective reservoirs—will simply be written as

$$H^{incoh} = a^+\Gamma_a + b^+\Gamma_b + \text{HC} \tag{20}$$

where Γ_a and Γ_b are the reservoir operators. Applying standard techniques of the quantum theory of damping [18] we find the following master equation for the reduced density operator of the system:

$$\frac{\partial\rho}{\partial t} = \frac{1}{\mathrm{i}\hbar}[H^{coh}_{total},\rho] + \sum_{i=1}^{2}\gamma_i(2e_i\rho e_i^+ - e_i^+e_i\rho - \rho e_i^+e_i) + \sum_{i=1}^{2}2\gamma_i n_i^{th}[[e_i,\rho],e_i^+] \tag{21}$$

where $(e_1, e_1^+) = (a, a^+)$ and $(e_2, e_2^+) = (b, b^+)$. Also, γ_1 $(=\gamma_a)$ and γ_2 $(=\gamma_b)$ are the damping rates of the photon and phonon, respectively, and n_1^{th} (n_2^{th}) represents the mean thermal photon (phonon) number of the reservoir.

5. Semiclassical equations of motion and IAOB

Let

$$\begin{aligned}\langle a\rangle &= \alpha\exp(\mathrm{i}\omega t) = |\alpha|\exp(\mathrm{i}(\omega t+\phi_a))\\ \langle b\rangle &= \beta\exp(\mathrm{i}\omega t) = |\beta|\exp(\mathrm{i}(\omega t+\phi_b))\end{aligned} \tag{22}$$

where ϕ_a (ϕ_b) is the phase of the photon (phonon) relative to the incident light. The semiclassical equation of motion, corresponding to equations (19) and (21), should be

$$\begin{aligned}\dot{\alpha} &= -(\gamma_a+\mathrm{i}\Delta_a)\alpha + gf - g_{ab}\beta - g''_{ab}\beta|\beta|^2\\ \dot{\beta} &= -(\gamma_b+\mathrm{i}\Delta_b)\beta - 4g''_b\beta|\beta|^2 + g_{ab}\alpha + g''_{ab}(2\alpha|\beta|^2-\alpha^*\beta^2)\end{aligned} \tag{23}$$

together with the complex conjugate equations.

Set $\dot{\alpha} = \dot{\beta} = 0$ for the steady state, and for simplicity let $\Delta_a = \Delta_b = 0$, which corresponds to the resonant situation; we can now obtain the stationary solution of equation (23).

Define

$$n = |\beta|^2 \qquad x = |\alpha|^2 \qquad y = g^2f^2/\gamma_a^2 \tag{24}$$

where x and y stand for the output cavity-field intensity and input driving-field intensity, respectively, and n is the number of coherent phonons (excitation intensity).

From equation (23) we have

$$gf = \gamma_a\alpha + g_{ab}(1+v_1n)\beta \tag{25}$$

$$\gamma_b n = g_{ab}(1+v_1n)\sqrt{xn}\cos(\phi_a-\phi_b) \tag{26}$$

$$4g''_b n^2 = g_{ab}(1+3v_1n)\sqrt{xn}\sin(\phi_a-\phi_b) \tag{27}$$

after a simple calculation, we can obtain the output x versus incident y characteristics of steady states in the parametric form (the number of coherent phonons n is taken as a controlling parametric quantity)

$$x = \frac{k_1 n}{(1+v_1n)^2}\left(1+\frac{v_2^2n^2(1+v_1n)^2}{(1+3v_1n)^2}\right) \tag{28}$$

$$y - x = k_2(1+v_1n)^2n + 2\sqrt{k_1k_2}n. \tag{29}$$

In the above equations,

$$k_1 = \gamma_b^2/g_{ab}^2 \qquad k_2 = g_{ab}^2/\gamma_a^2 \tag{30}$$

$$v_1 = g''_{ab}/g_{ab} \qquad v_2 = 4g''_b/\gamma_b. \tag{31}$$

Equations (28) and (29) are similar to the equations (30) and (31) in reference [14], which could lead to an IAOB, as was expected. We would like to call x versus n in equation (28) the 'characteristic curve' and $(y-x)$ versus n in equation (29) the 'efficiency curve'. From equation (27) we can see that the interaction between phonons (presented as $g''_b n^2$ in the left-hand side of equation (27)), which is neglected in reference [14], will cause a phase detuning between the photon (α) and phonon (β) even in the resonant situations. And it will also wash out the characteristics of the IAOB, which can be seen clearly from equations (28) and (29).

When the coupling constant associated with the nonlinear photon–phonon interaction v_1 is considerably greater than that of the phonon–phonon interaction v_2, the steady-state equations (28) and (29) give the typical x versus y curves of IAOB on choosing suitable parameters. Our numerical calculations are demonstrated in figures 1 to 3.

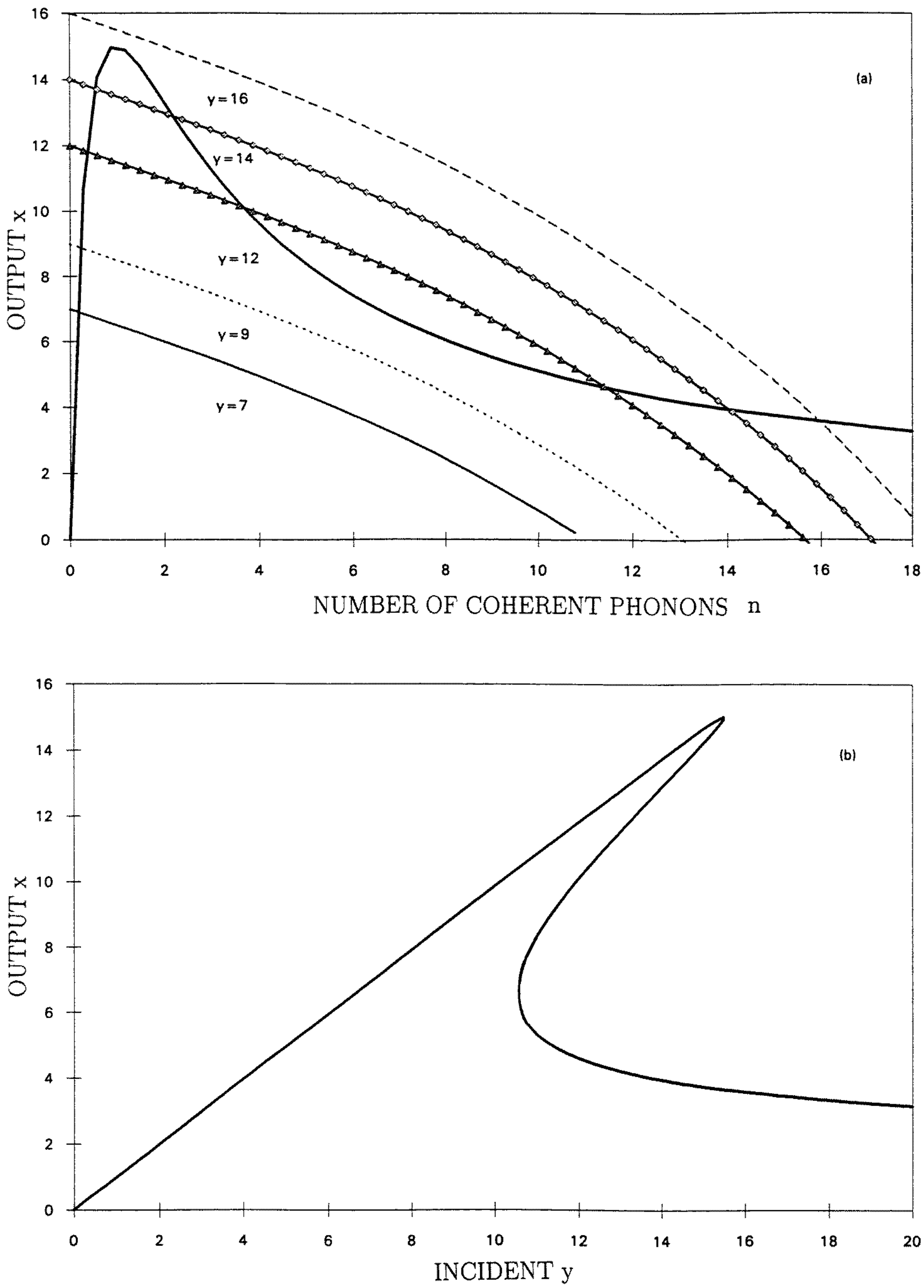

Figure 1. (a) The steady-state output x versus n 'characteristic curve' and the 'effective curve' with the parameters $k_1 = 60, k_2 = 0.001, v_1 = 1, v_2 = 0.05$ for different inputs y. (b) The output x versus input y curve determined from (a).

Figure 1 shows the steady-state output x versus n 'characteristic curve' and the 'efficiency curve' for the parameters $k_1 = 60, k_2 = 0.001, v_1 = 1$, and $v_2 = 0.05$. The

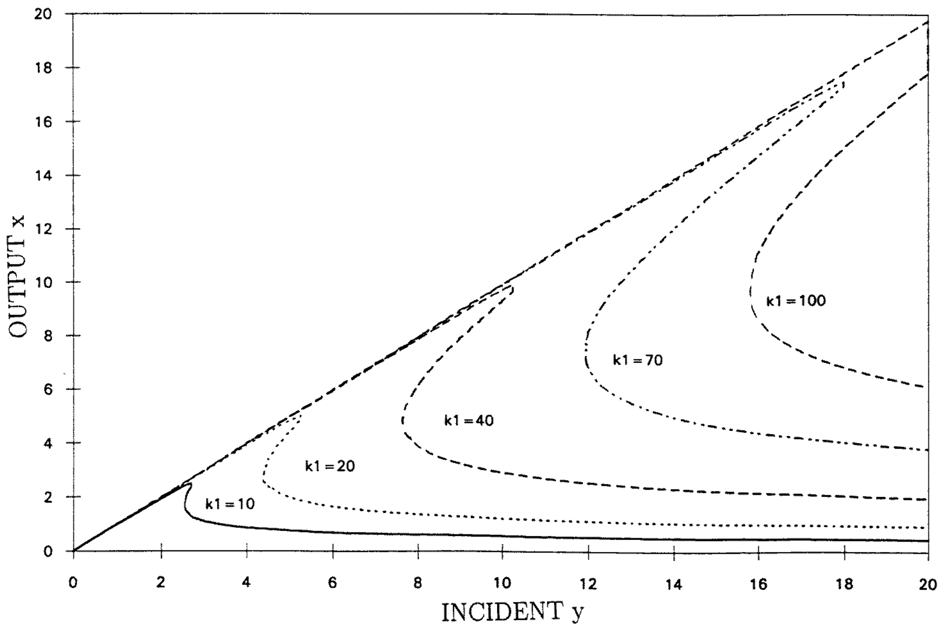

Figure 2. The output x versus input y characteristics with different values of $k_1 = 100, 70, 40, 20, 10$; the other parameters are the same as in figure 1.

characteristics of the IAOB are obviously illustrated in the figure. To study the effect of the parameters, the output–incident characteristics for different values of k_1 and v_2 are plotted in figure 2 and figure 3, from which we can see that if the coupling constants and the damping rate of the photons keep constant, the behaviour of the IAOB will become more obvious with the increasing of the damping rate of the phonons (where $k_1 = \gamma_b^2/g_{ab}^2$). But the phonon–phonon interaction will reduce the behaviour of the IAOB. As we can see from figure 3, with the increase of v_2 (standing for the strength of the phonon interaction), the IAOB will be weakened; when v_2 is more than 0.5, IAOB is hardly found. From our numerical calculation we also found that if the damping rate of photons is very small, no IAOB will occur, because the 'characteristic curve' is far from the 'IAOB characteristic curve' presented in reference [14].

6. Discussion

The coupled equations of motion (equation (23)) are the main results of our model for IAOB. It is interesting that our fundamental results, i.e., H_{total}^{coh}, expressed by equation (19), are completely the same as the coherent part of the Hamiltonian of the photon–exciton coupled system (in the low-density case), derived in the paper about the theory of excitonic optical bistability by Steyn-Ross and Gardiner [16]. This fact shows that the optical nonlinearities due to the interaction of photons with various elementary excitations of bosonic type such as phonons and excitons may be the origin of the IAOB. Therefore, both this paper and Steyn-Ross and Gardiners work [16], in some sense, support the Hamiltonian of the IAOB model obtained in a phenomenological way in reference [14].

However, the IAOB presented here would hardly be expected from the work of Steyn-

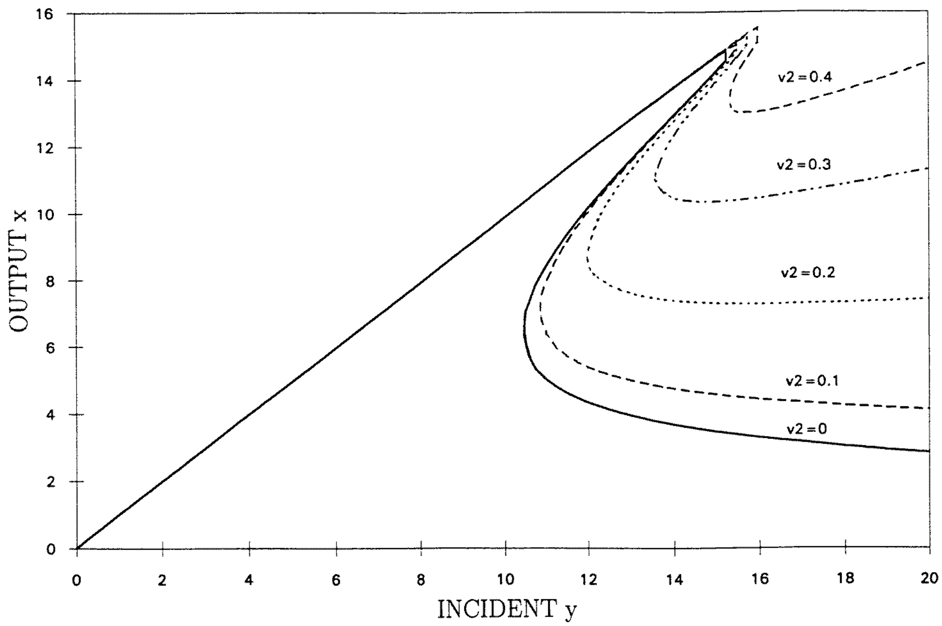

Figure 3. The output x versus input y characteristics with different values of $v_2 = 0.4, 0.3, 0.2, 0.1, 0$; the other parameters are the same as in figure 1.

Ross and Gardiner [16]. In their analysis of the steady-state behaviour they neglected the photon–exciton coupling in reality and emphasized the exciton–exciton interaction, so they believed that 'the bistability depends solely on exciton–exciton interaction', which could be seen clearly in the third part of their paper with regard to the effect of the parameters. This is just the opposite of our present result. In our opinion, the origin of the IAOB lies in the nonlinear interaction between the photon and the elementary excitations, including the exciton; the phonon–phonon coupling is not the cause of the IAOB, but is what disturbs or weakens the IAOB, as can be seen clearly from figure 3 and the corresponding discussion in the text.

Acknowledgments

This work was supported by the National Natural Science Foundation of China; K W Yu acknowledges support from the Research Grants Council of the Hong Kong Government under numbers CUHK 78/93E and 461/95P.

References

[1] Fu H and Hagen H 1989 *Phys. Rev. Lett.* **60** 2641
[2] Gong S Q, Pan S H and Yang G Z 1993 *Phys. Rev.* A **47** 2205
[3] Gong S Q, Pan S H and Yang G Z 1992 *Phys. Rev.* A **45** 6655
[4] Wang Z C, Guo W and Zheng S 1992 *Phys. Rev.* A **46** 7635
[5] Grand Z 1992 *Phys. Rev.* A **46** 5860
[6] Li X S, Lin D L, George T F and Sun X 1990 *Phys. Rev.* B **41** 3280
[7] Lin D L, Li X, Liu Z D and George T F 1991 *Phys. Lett.* **152A** 229; 1991 *Phys. Lett.* **159A** 369

[8] Joshi A and Lawande S V 1992 *Phys. Rev.* A **46** 5903
[9] Reid M D and Yurke B 1992 *Phys. Rev.* A **46** 413
[10] Li X S, Lin D L, George T F and Sun X 1990 *Phys. Rev.* B **42** 2977
[11] Grangier P, Roch J F, Roger J, Lugiato L A, Pessina E M, Scandroglio G and Galatola P 1992 *Phys. Rev.* A **46** 2735
[12] Ou F, Wei B H, Yu K W and Liu C H 1996 *J. Phys.: Condens. Matter.* **8** 2957
[13] Ou F 1990 *Phys. Rev.* A **41** 3021
[14] Ou F 1992 *Z. Phys.* B **88** 117
[15] Gibbs H M 1985 *Optical Bistability: Controlling Light with Light* (Orlando, FL: Academic)
[16] Steyn-Ross M L and Gardiner C W 1983 *Phys. Rev.* A **27** 310
[17] Wallis R F 1977 Phonons and polaritons *Interaction of Radiation with Condensed Matter* vol 17 (Vienna: IAEA) p 163
[18] Louisell H W 1973 *Quantum Statistical Properties of Radiation* (New York: Wiley)