BISTABLE REFLECTIVITY IN BULKY SEMICONDUCTORS DUE TO TWO PHOTON-BIEXCITON TRANSITIONS

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A new mechanism has been proposed for optical bistability in a bulk system of interacting photons and biexcitons. Bistable behaviour can occur for very low pumping intensities if the input light frequency is clearly less than the two-photon resonance of the biexciton.

I. Introduction

The two-photon resonance of the biexciton in direct-band-gap semiconductor like CuCl, CuBr, ... is an important source of large nonlinearities which may be employed to obtain bistability [1−11]. Koch and Haug [1] showed that in the vicinity of the biexciton resonance the complex non-perturbational dielectric function ε depends on the intracrystal or internal photon concentration n and optical bistability can be generated with the use of a Fabry-Perot resonator or cavity. (Note that a platelet of material can form a cavity with its natural end faces serving as mirrors.) The presence of the cavity plays a significant role. If there were not any cavity or if one had a bulk crystal (in a bulk material, as opposed to a thin one, the reflection at the right end face is very weak and one can neglect the backward wave, i.e., a bulk sample cannot be treated as a mirrorless cavity), no bistability might occur though ε remains still n-dependent. Similar situations were reported in [2] where for deriving ε not a Green function method as in [1] but the density matrix formalism was applied. Abram and Maruani [3] based their treatment on operator technique, which yields an analytic expression for ε directly dependent on the input intensity. Instead of the feedback provided by the cavity, the authors of [3] suggested to take into account the local field effect (LFE) that caused the refractive index to be a multi-valued function of the incident intensity, implying a potentially bistable optical behaviour. Unfortunately the results of [3] cannot be correct because of the presence of a double pole in the expression for ε (the well-known Kramers-Kronig relation requests ε to possess simple poles only [12]). In [4] Sung and Bowden attempted to obtain from Langevin equations the function which contains no double poles. Then, however, it becomes apparent that the dispersion relation of Sung and Bowden looks very strange. Now ε has got a strange pole at the frequency corresponding to induced emission or absorption even when neither excitons nor biexcitons exist in the material [4, 6, 7]. The LEF should also be treated to generate bistability of refractivity.

Further, Dror Sarid, Peyghambarian and Gibbs [5] have shown that the inclusion of the LFE in calculating ε for the exciton-biexciton system does not yield bistability on reflection as proposed in [3, 4]. The LFE really provides a feedback in the system and makes ε to be a tri-valued function of intensity, but among the three solutions to ε two do diverge at zero input intensity (physically invalid) and therefore the bistability due to the LFE is purely mathematical but not physical. In [6] Haug has developed a nonequilibrium many-body theory of optical nonlinearities by applying the nonequilibrium Green function technique and arrived at the same results as in [1–4]. Recently, Henneberger in his review [8] has added to the study also the equations of motion for the exciton and biexciton populations with taking into account the longitudinal lifetime of the quasiparticles. However, the above-said strange pole cannot be removed yet and the resonator or/and the LFE have to be included for the appearance of bistability. So far, one can see that in spite of various approaches the results are almost the same but regretfully unacceptable. There must be some basic inconsistent assumption common to all the works cited above. To our knowledge the origin of such inconsistencies may be sought in the starting Hamiltonian of the system. Since we are interested in the giant two-photon-biexciton transition in which excitons appear only as virtual intermediate states, it seems quite reasonable to work with an effective Hamiltonian [9, 11, 13] describing just the relevant transition and not containing exciton operators explicitly. We expect that the use of such an effective Hamiltonian would exclude the strange pole. To get rid of the double pole we shall involve into the study the internal photons [9, 11] which are driven by an externally classical laser field and responsible for the coupling with biexciton in the semiconductor. By the way, Khadzi, Moskalenko et al. [9] have pointed out that the internal photon number could behave in a bistable manner depending on two resonant pump fields simultaneously acting on the system. In our previous paper [11], we have predicted the possibility of the occurrence of bistability even when if the system were pumped by a single field only.

In this paper we intend to derive the complex dielectric function ε for a coherent photon-biexciton system in a bulky semiconductor under the action of an externally driving laser field and to show that reflectivity can be bistable even when the resonator as well as the LFE are not being taken into account.

We shall utilize the system of units with $\hbar = c = 1$, where $\hbar$ and c are the Planck constant and the velocity of light.

II. Intensity-dependent complex dielectric function

Consider a bulk semiconductor onto the left end surface of which a monochromatic light beam with wave vector $\boldsymbol{q}$ and frequency Ω_q is incident. Suppose the beam propagating inside the crystal is characterized by its wave vector $\boldsymbol{k}$ and frequency ω_k. Thanks to Maxwell-Frenkel boundary conditions Ω_q must be equal to ω_k but $\boldsymbol{q}$ and $\boldsymbol{k}$ are different. Let us treat the external electric field $E_q(t)$ classically, i.e.

$$E_{\boldsymbol{q}}(t) = E_{\boldsymbol{q}}^{(-)}(t) + E_{\boldsymbol{q}}^{(+)}(t) \tag{1}$$

$$E_{\boldsymbol{q}}^{(\pm)}(t) = E_{\boldsymbol{q}}^{(\pm)}\,\mathrm{e}^{\pm \mathrm{i}\Omega_q t} \tag{2}$$

with $E_{\boldsymbol{q}}^{(\pm)}$ being its amplitudes and the intracrystal field to be secondly quantized with the photon creation (annihilation) operator labelled by $c_{\boldsymbol{k}}^{+}(c_{\boldsymbol{k}})$. The external field, then, drives the internal photons which may interact with each other to generate biexcitons in the crystal. The effective Hamiltonian for the coherent photon-biexciton system driven by the external pumping field (1) can be constructed in the form [9, 11, 13, 14]:

$$H(t) = H_{\mathrm{ext}}(t) + H_{\mathrm{int}} \tag{3}$$

$$H_{\mathrm{ext}}(t) = -(\tfrac{1}{2}V\omega_k)^{1/2}\,E_{\boldsymbol{q}}^{(-)}\mathrm{e}^{-\mathrm{i}\Omega_q t}c_{\boldsymbol{k}}^{+} + \mathrm{h.c.} \tag{4}$$

$$H_{\mathrm{int}} = (\omega_k + A_k)\,c_{\boldsymbol{k}}^{+}c_{\boldsymbol{k}} + \omega_{m2k}b_{2\boldsymbol{k}}^{+}b_{2\boldsymbol{k}} + \frac{1}{\sqrt{V}}\,g_k(b_{2\boldsymbol{k}}^{+}c_{\boldsymbol{k}}c_{\boldsymbol{k}} + c_{\boldsymbol{k}}^{+}c_{\boldsymbol{k}}^{+}b_{2\boldsymbol{k}}) \tag{5}$$

where $b_{2\boldsymbol{k}}$ and $b_{2\boldsymbol{k}}^{+}$ are bosonic operators for biexcitons with momentum $2\boldsymbol{k}$ and energy ω_{m2k}; $A_k = \omega_0^2/4\varepsilon_\infty\omega_k$ stems from the A^2-type term of the light-matter interaction Hamiltonian representing the couple of a photon with a valence electron [14]; ω_0 is the plasma frequency of unexcited crystal ε_∞ serves for the background dielectric constant covering the contribution of all those transitions other than that involved in (5); g_k denotes the effective biexciton-two-photon interaction constant [9, 11, 13] and, finally, V stands for the volume of the sample. Because of the boundary conditions and the intracrystal selection rules only the $\boldsymbol{q}$-component of the external field, photons with the wave vector $\boldsymbol{k}$ and biexciton with the momentum $2\boldsymbol{k}$ need be considered. Thus, from now on we may drop for brevity the indices $\boldsymbol{q}$, $\boldsymbol{k}$ and $2\boldsymbol{k}$ in all further formulae. Note that $H_{\mathrm{ext}}(t)$ in (4), on the other hand, can be expressed in a semiclassical picture in terms of polarization operators $P^{(\pm)}(t)$ as below:

$$H_{\mathrm{ext}}(t) = -[E^{(-)}(t)\,P^{(+)}(t) + E^{(+)}(t)\,P^{(-)}(t)]\,. \tag{6}$$

From (6) and (4) it follows that $P^{(+)}(t)$ may be determined by the functional derivative of $H_{\mathrm{ext}}(t)$ with respect to the external field

$$P^{(\pm)}(t) = -\frac{\delta H_{\mathrm{ext}}(t)}{\delta E^{(\mp)}(t)} = (\tfrac{1}{2}V\omega)^{1/2} \times \begin{cases} c^{+} \\ c \end{cases} \tag{7}$$

According to definition, the macroscopic nonlinear susceptibilities $\chi^{(\pm)}(t)$ are of the forms

$$\chi^{(\pm)}(t) = \frac{1}{V}\frac{\langle P^{(\pm)}(t)\rangle}{E^{(\pm)}(t)} = \frac{1}{E^{(\pm)}(t)}\left(\frac{\omega}{2V}\right)^{1/2} \times \begin{cases} \langle c^{+}\rangle \\ \langle c\rangle \end{cases} \tag{8}$$

Now the complex dielectric functions $\varepsilon^{(\pm)}$ for $E^{(\pm)}(t)$ are determined by (in the rationalized unit system)

$$\varepsilon^{(\pm)} = \varepsilon_\infty + \chi^{(\pm)}\,. \tag{9}$$

In this paper the incoming beam is assumed to be $\sim E^{(-)}$ and hence we should calculate $\varepsilon^{(-)}$ which for brevity will be written as ε

$$\varepsilon \equiv \varepsilon^{(-)} = \varepsilon_{\infty} + \left(\frac{\omega}{2V}\right)^{1/2} \frac{\langle c \rangle}{E^{(-)}(t)} \,. \tag{10}$$

In order to obtain the desirable expression for ε we have to set up appropriate Heisenberg equations of motion and then solve them for $\langle c \rangle$. Using the effective Hamiltonian (3) we obtain the following system of equations [11]:

$$\left(\frac{d}{dt} + \gamma_{\perp} + i(\omega + A)\right) c + \frac{2ig}{\sqrt{V}} c^{+} b = i(\tfrac{1}{2}V\omega)^{1/2} E^{(-)}(t) \,, \tag{11}$$

$$\left(\frac{d}{dt} + \Gamma_{\perp} + i\omega_m\right) b + \frac{ig}{\sqrt{V}} cc = 0 \,, \tag{12}$$

where $\gamma_{\perp}$ and $\Gamma_{\perp}$ are the inversions of the transverse phase relaxation times for the photon and the biexciton, resp., which are introduced phenomenologically. Multiplying both sides of (11), from the right, by c and then taking the expectation values of (11) and (12) over the state of the system under the action of the pump field, we arrive at

$$\left(\frac{d}{dt} + \gamma_{\perp} + i(\omega + A)\right) \langle cc \rangle + \frac{2ig}{\sqrt{V}} \langle c^{+} cb \rangle = i(\tfrac{1}{2}V\omega)^{1/2} E^{(-)}(t) \langle c \rangle \tag{13}$$

$$\frac{ig}{\sqrt{V}} \langle cc \rangle + \left(\frac{d}{dt} + \Gamma_{\perp} + i\omega_m\right) \langle b \rangle = 0 \,. \tag{14}$$

For solving (13) and (14) let us confine ourselves to the simplest approximations such as

$$\langle cc \rangle = \langle c \rangle \langle c \rangle \tag{15}$$

$$\langle c^{+} cb \rangle = \langle c^{+} c \rangle \langle b \rangle = Vn \langle b \rangle \tag{16}$$

with $n = (1/V) \langle c^{+} c \rangle$ being the internal photon distribution function which is in this case nonequilibrium one but not Bose-Einstein's. For the stationary regime we look for particular solutions in the forms:

$$\langle b \rangle = B\, e^{-2i\Omega t} \tag{17}$$

$$\langle c \rangle = C\, e^{-i\Omega t} \tag{18}$$

with B, C = const. Inserting (15–18) into (13, 14) it is easy to get:

$$C = \frac{i(2i\delta + \Gamma_{\perp}) E^{(-)} (\tfrac{1}{2}V\Omega)^{1/2}}{(iA + \gamma_{\perp})(2i\delta + \Gamma_{\perp}) + 2ng^2} \tag{19}$$

$$B = \frac{gCE^{(-)}(\tfrac{1}{2}\Omega)^{1/2}}{(iA + \gamma_{\perp})(2i\delta + \Gamma_{\perp}) + 2ng^2} \tag{20}$$

where $\delta = \frac{1}{2}\omega_m - \Omega$. Thus, for ε we have

$$\varepsilon = \varepsilon_\infty + \frac{(\frac{1}{2}\mathrm{i}\Gamma_\perp - \delta)\,\Omega}{(\mathrm{i}A + \gamma_\perp)(2\mathrm{i}\delta + \Gamma_\perp) + 2ng^2}\,. \tag{21}$$

From (21) follows that there is only a simple peak at $\delta = 0$, i.e. at $\Omega = \frac{1}{2}\omega_m$ in ε. The unphysically quadratic and strange poles disappear, which displays an advantage of our approach. Equation (21) exhibits a one-to-one correspondence between ε and n. Nevertheless, regarding the light intensity dependence, ε might become a tri-valued function capable to cause a bistable behaviour in reflection spectrums. This results from the specific dependence of n on the light intensity I

$$I = E^{(+)}E^{(-)} \equiv |E^{(+)}|^2 \equiv |E^{(-)}|^2\,. \tag{22}$$

Using again $H(t)$ in (3) we write down the Heisenberg equation of motion for $\langle c^+c\rangle$

$$\left(\frac{\mathrm{d}}{\mathrm{d}t} + \gamma_\parallel\right)\langle c^+c\rangle = \frac{2\mathrm{i}g}{\sqrt{V}}\left(\langle ccb^+\rangle - \langle c^+c^+b\rangle\right)$$

$$+ \mathrm{i}(\tfrac{1}{2}V\omega)^{1/2}\left(E^{(-)}(t)\langle c^+\rangle - E^{(+)}(t)\langle c\rangle\right) \tag{23}$$

with the empirical photon longitudinal lifetime $\sigma_\parallel = \gamma_\parallel^{-1}$. For the steady state and within the framework of the approximations (15, 16), we may solve (23) for $\langle c^+c\rangle = Vn$ and obtain:

$$V\gamma_\parallel n = -\frac{4g}{\sqrt{V}}\,Im(C^2B^*) - 2(V\omega)^{1/2}\,Im(E^{(-)}c^*) \tag{24}$$

Substituting C and B in (24) by those in (19, 20) we come to

$$n = \frac{\gamma_\perp\gamma_\parallel^{-1}\Omega(4\delta^2 + \Gamma_\perp^{\,2})\,I}{(\gamma_\perp\Gamma_\perp - 2A\delta + 2ng^2)^2 + (A\Gamma_\perp + 2\gamma_\perp\delta)^2}\,. \tag{25}$$

For convenience, we cast (25) into another, more compact form as below

$$J = [(n + \alpha)^2 + \beta^2]\,n \tag{26}$$

where

$$J = \tfrac{1}{4}\gamma_\perp\gamma_\parallel^{-1}g^{-4}\Omega(4\delta^2 + \Gamma_\perp^2)\,I \tag{27}$$

$$\alpha = \frac{1}{2\,g^2}(\gamma_\perp\Gamma_\perp - 2A\delta) \tag{28}$$

$$\beta = \frac{1}{2g^2}(A\Gamma_\perp + 2\gamma_\perp\delta)\,. \tag{29}$$

As already shown in [11], the conditions for n to be a tri-valued function of J are put on parameters α and β, namely they are either

$$\beta > 0 \quad \text{and} \quad \alpha + \beta\sqrt{3} < 0 \tag{30}$$

or

$$\beta < 0 \quad \text{and} \quad \alpha - \beta\sqrt{3} < 0\,. \tag{31}$$

When (30) or (31) holds, there exist two critical values of intensity $\dot{J}_{\swarrow}$ and $\dot{J}_{\nearrow} > \dot{J}_{\swarrow}$ between which one value of $\dot{J}$ corresponds to three values of n (among them one is unstable [15]). Concerning the intensity dependence of ε, as it is obvious from (21) and (26), ε does depends on $\dot{J}$ parametrically by means of n. Consequently, we have

$$\frac{\partial \dot{J}}{\partial \varepsilon} = \frac{\partial \dot{J}}{\partial n} \cdot \frac{\partial n}{\partial \varepsilon} = \frac{\partial \dot{J}/\partial n}{\partial \varepsilon/\partial n} . \tag{32}$$

Equation (32) speaks that once (30) or (31) is met, ε as well, as n would behave in a bistable manner within the same intensity interval $[\dot{J}_{\swarrow}, \dot{J}_{\nearrow}]$ disregarding the particular form of function $\varepsilon = \varepsilon(n)$. A noteworthy thing here is that to obey (30) or (31) α must anyway be negative (easily to be verified). This might be provided by the presence of the second term in (28) which is proportional to $A\delta$. If either A or δ were zero, then α would always be positive and bistability could never happen. Thus, the driving field frequency must be tuned below (see later) the exact two-photon resonance of the biexciton and, more than that, the interaction of the internal photon with the crystal valence electron ($\sim A$) must also taken into account to expect the bistability.

Before proceeding further, we try to represent (30, 31) directly in terms of material characteristics. It is not difficult to prove that (31) should not be satisfied for the case under investigation and (30) would be equivalent to

$$A > \gamma_\perp \sqrt{3} \tag{33}$$

and

$$\delta > \delta_c = \frac{\Gamma_\perp}{2} \frac{\gamma_\perp + A\sqrt{3}}{A - \gamma_\perp \sqrt{3}} . \tag{34}$$

Introducing a few normalized dimensionless quantities such as

$$x = \frac{\delta}{\delta_c} , \quad y = \frac{A}{\gamma_\perp \sqrt{3}} , \quad z = \frac{\Omega}{2\gamma_\perp} , \tag{35}$$

$$\tilde{n} = \frac{2g^2}{\gamma_\perp \Gamma_\perp} n , \quad \tilde{I} = \frac{2\Omega g^2}{\gamma_\parallel \gamma_\perp^2 \Gamma_\perp} I , \tag{36}$$

we can rewrite (33), (34), (26) and (21) into the following forms:

$$x > 1 , \quad y > 1 , \tag{37}$$

$$\tilde{I} = \frac{\left\{\left[\tilde{n} + 1 - \dfrac{(1 + 3y)\, xy}{(y - 1)}\right]^2 + \left[y\sqrt{(3)} + \dfrac{(1 + 3y)\, x}{\sqrt{(3)}\,(y - 1)}\right]^2\right\} \tilde{n}}{1 + \dfrac{(1 + 3y)^2\, x^2}{3(y - 1)^2}} , \tag{38}$$

$$\varepsilon = \varepsilon_\infty + \frac{z\left[\mathrm{i} - \dfrac{(1+3y)\,x}{\sqrt{(3)}\,(y-1)}\right]}{1 + \tilde{n} - \dfrac{(1+3y)\,xy}{(y-1)} + \mathrm{i}\left[y\sqrt{(3)} + \dfrac{(1+3y)\,x}{\sqrt{(3)}\,(y-1)}\right]}. \tag{39}$$

To carry out the necessary numerical evaluation, we remark that the quantities involved by (35) are not independent. For example, Z is governed not only by x and y but also by some material characteristics such as ω_m, $\Gamma_\perp$ and A, i.e.

$$Z = \left[\omega_m - \frac{\Gamma_\perp}{\sqrt{3}}\frac{(1+3y)x}{(y-1)}\right]\frac{y\sqrt{3}}{4A}. \tag{40}$$

III. Numerical calculations of the reflection coefficient

In this section we are going to calculate the reflection coefficient for CuCl the parameters of which are $\omega_x = 3{\cdot}2027$ eV (ω_x – the exciton energy), $\omega_m = 6{\cdot}3725$ eV, $\varepsilon_\infty = 5$; $\Gamma_\perp = 0{\cdot}2$ meV, $\gamma_\perp = 0{\cdot}1$ meV and $\Delta_{LT} = 5{\cdot}5$ meV (Δ_{LT} – the measurable longitudinal-transverse splitting). These data give $y = 15{\cdot}94$ and $\delta_c = 0{\cdot}188$ meV implying that (33) is automatically fulfilled and the incident laser frequency Ω must be tuned at least 0·188 m eV lower than the exact two-photon resonance of the bi-exciton to observe optical bistability. The reflection coefficient R experienced by the light wave propagating in the direction perpendicular to the surface of the sample can be determined by the complex dielectric function $\varepsilon = \varepsilon_1 + \mathrm{i}\varepsilon_2$ in (39):

$$R = \left|\frac{1-\sqrt{\varepsilon}}{1+\sqrt{\varepsilon}}\right|^2 = \frac{1 + (\varepsilon_1^2+\varepsilon_2^2)^{1/2} - (2(\varepsilon_1 + (\varepsilon_1^2+\varepsilon_2^2)^{1/2}))^{1/2}}{1 + (\varepsilon_1^2+\varepsilon_2^2)^{1/2} + (2(\varepsilon_1 + (\varepsilon_1^2+\varepsilon_2^2)^{1/2}))^{1/2}}. \tag{41}$$

The interaction constant g in (5) is defined as [16]

$$g^2 = \left|\frac{\mu_{0x}}{\Omega - \omega}\right|^2 |\mu_{xm}|^2 \tag{42}$$

where μ_{0x} and μ_{xm} are the matrix elements of the optical ground state-to-exciton and exciton-to-biexciton transitions, resp. According to [14, 17] $\mu_{0x}^2 = \omega_x^2 \Delta_{LT}/2\Omega$ and $\mu_{xm}^2 = 3{\cdot}14 \times 10^{-23}\,(\mathrm{eV})^2\,(\mathrm{cm})^3$. In the calculation process we can take the value of g^2 near the point with $\Omega = \frac{1}{2}\omega_m$ that is $g^2 \approx 1{\cdot}04 \times 10^{-21}\,(\mathrm{eV})^2\,(\mathrm{cm})^3$. Using (38), (39) and (41) we may plot $\tilde{n}$, ε_1, ε_2 and R as functions of $\tilde{I}$ for various x. All these quantities will be bistable for $x > 1$. For example, we represent in the figure the intensity-dependence of R for four values of $x = 0{\cdot}5$; $1{\cdot}0$; $2{\cdot}0$ and $5{\cdot}0$. As seen from the figure for 0·5 the reflection coefficient grows monotonously with increasing incident intensity while for $x = 2{\cdot}0$ or $5{\cdot}0$ there exists some interval of $\tilde{I}$ within which R behaves in a bistable way. The $x = 1$ case is the critical one. The intensity corresponding to the inflection point of the curve $R = R(\tilde{I})$ for $x = 1$ is the threshold of the input intensity higher than which the hysteresis effect of R (when $x > 1$) starts to appear. Directly inspecting the figure and using (36) with $\gamma_{\parallel} \approx 1$ meV we could estimate the threshold of I by the following formula:

$$I_c \approx \frac{9 \times 10^3 \gamma_{\parallel} \gamma_{\perp} \Gamma_{\perp}}{(\omega_m - 2\delta_c)\, g^2} \approx 13 \frac{\mathrm{W}}{(\mathrm{cm})^2}. \tag{43}$$

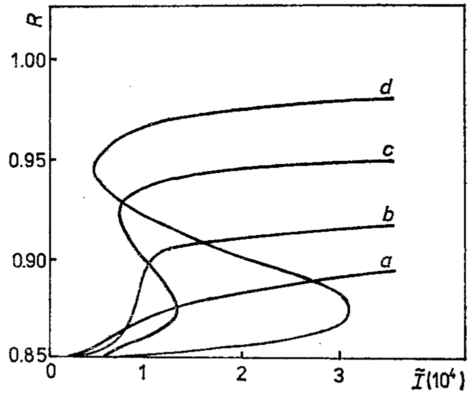

Fig. 1. Reflection coefficient R vs the normalized input laser intensity $\tilde{I}$ for $x = 0{\cdot}5$ (curve *a*); 1·0 (*b*); 2·0 (*c*) and 5·0 (*d*).

Note that the above-estimated value of I_c is much less than those of [1, 4] and is about the same order as that for the optical bistability of excitons in a Fabry-Perot resonator [18] is.

IV. Conclusion

We have investigated the bistable behaviour of the reflectivity in the bulky semiconductor by using the effective two photon-biexciton Hamiltonian. The double and strange poles do not appear in our treatment. The mechanism of bistability is the renormalization of the internal photons due to their interaction with the valence electrons. The threshold of input intensity for the occurence of bistability is estimated for CuCl which seems to be very low.

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