Optical bistability of a dense exciton-biexciton system: CuCl

C. C. Sung
Department of Physics, The University of Alabama in Huntsville, Huntsville, Alabama 35899

C. M. Bowden
Research Directorate, U.S. Army Missile Laboratory, U.S. Army Missile Command, Redstone Arsenal, Alabama 35898
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The steady-state solution of the Langevin equation for a dense exciton-biexciton system is investigated. The corrections to the previous work, the contribution of the field—exciton-biexciton interaction, and the local-field correction are shown to modify the threshold of optical bistability significantly.

I. INTRODUCTION

Optical bistability (OB) is the nonlinear response of a material to incident laser light which results in bistability and reversible hysteresis of the output as a function of the incident intensity. Absorptive OB (the nonlinear response of a saturable absorber) was first predicted by Szöke *et al.*[1] Later, McCall[2] showed by numerical integration of the Maxwell-Bloch equations with boundary conditions for a Fabry-Perot cavity, that under suitable conditions such a system can exhibit differential gain and transistor action. This work led to the experiments of Gibbs, McCall, and Venkatesan[3] in Na vapor in which the main predictions of McCall[2] were realized. The analysis of the data, however, indicated that the observed OB was essentially of a dispersive nature and this was subsequently explained by a simple phenomenological cubic model.

The results of the earlier work stimulated much theoretical and experimental activity.[4] Of particular note is the contribution to the fundamental understanding of absorptive OB by Bonifacio and Lugiato[5,6] who showed that OB can be regarded as a first-order phase transition far from thermodynamic equilibrium. The case for mixed absorptive and dispersive OB has also been worked out by them[6] and the general conditions for OB in collections of two-level atoms have been predicted.[7–9]

Because of rather obvious potential practical applications, interest in OB later turned to its observation in semiconductors. The first demonstrated OB in a semiconductor was conducted by Gibbs *et al.*[10] in GaAs at low temperature in which the OB is primarily dispersive and the nonlinear index arises from light-induced changes in excitonic absorption. Subsequently, and almost coincident with the first GaAs experiment[10] was the work of Miller, Smith, and Johnston[11] who observed OB in InSb at 5 K due to a direct band-gap resonance mechanism. Quite recently, Gibbs *et al.* have observed excitonic OB at room temperature in a GaAs-GaAlAs superlattice etalon.[12]

Theoretically, Koch and Haug[13] (KH) have studied OB in the vicinity of the biexciton resonance, and indications are that the intensity-dependent resonance may indeed be the mechanism for the observed OB in GaAs of Ref. 10. Furthermore, the results of the work of KH relate quantitatively to the two-photon resonance Raman scattering experiments in CuCl.[14] Thus, CuCl holds particular interest as a candidate for OB, and indeed KH predicted OB in CuCl as a function of frequency of the incident beam using only the intrinsic reflectivity in a CuCl resonator of 1-μm thickness (i.e., without extraneous reflecting surfaces such as mirrors). This is what we shall refer to as truly intrinsic OB, i.e., due entirely to the nonlinear interaction between the incident light and the material itself.

Recently, it was claimed[15] that incorporation of the complete field—exciton-biexciton (FEB) interaction and the local-field correction[16] (LFC) in a semiclassical model for CuCl predicts OB for a thin slab of material of dimension less than a wavelength of the incident field, tuned near the two-photon biexciton resonance. Apparently, the results of Ref. 15 are intended to be a correction to the nonlinear dielectric function ϵ in KH. Unfortunately, the results reported in Ref. 15 for ϵ cannot be correct for the main reason that their complex dielectric function ϵ contains a double pole that does not satisfy the Kramers-Kronig relation.[17] The problem originated from an incorrect treatment of the FEB as we will show later.

Although interesting in itself, mirrorless (intrinsic) OB is also intriguing from the potential application to small, fast-switching devices. In this situation, one may have the possibility for elements of thickness smaller than a resonance wavelength and switching rates perhaps limited by the inverse of the transit time of light through the element. The possibility of achieving intrinsic OB without mirrors was first suggested by Bowden and Sung[18] and by Bowden[19] based upon a fully quantum-mechanical treatment of interatomic correlation in the presence of an externally applied laser field. A close classical analog to the results of the fully quantum-mechanical model is the application of the local field correction[16] to the macroscopic Maxwell field in a semiclassical Maxwell-Bloch model.[20]

The purpose of this paper is to reexamine the effect of the FEB as a cause of OB together with the LFC. In Sec. II the Hamiltonian[21,22] for the exciton and exciton-biexciton model is presented and discussed, and the Langevin equations are obtained from which the complex dielectric function ϵ is derived. Next, we review the

local-field correction pertinent to this problem in Sec. III. Then, in Sec. IV, the correction to the main conclusion of KH is discussed, and a detailed comparison is presented. Section V is used to summarize our results and review the implication for observation of intrinsic OB in CuCl.

II. FIELD DEPENDENCE OF THE DIELECTRIC FUNCTION $\epsilon(k,\omega)$

The Hamiltonian given in Refs. 13, 15, and 21 for CuCl consists of two parts: the free excitons and biexcitons H_0, and the interaction of excitons and biexcitons with the external laser field H'. H_0 and H' in the rotating frame of the external field frequency ω are, respectively,

$$H_0=(\omega_x-\omega)b^\dagger b+(\omega_m-2\omega)B^\dagger B\ , \tag{1}$$

$$H'=ig_1E^+b^\dagger+ig_2E^+B^\dagger b+\text{H.c.} \tag{2}$$

Here, ω_x and ω_m are the energy levels of excitons and biexcitons and $b^\dagger$ (b) and $B^\dagger$ (B) are their collective creation (annihilation) operators, and units are chosen such that $\hbar=1$. The external field amplitude E^+ (E^-) associated with $e^{-i\omega t}$ ($e^{i\omega t}$) is treated as a c number. $g_{1,2}$ is the coupling constant of the external field with the induced dipole moment,

$$g_1=-(N/2V)^{1/2}\vec{e}\cdot\langle\vec{d}\rangle_1\ , \tag{3a}$$

$$g_2=-(N/2)^{1/2}\vec{e}\cdot\langle\vec{d}\rangle_2\ , \tag{3b}$$

where N and V are the number of unit cells and the volume, respectively, and $\vec{e}$ is the polarization vector of the external field. The dipole moment matrix elements $d_{1,2}$ are usually inferred from the experimental data,[13] since it is difficult to obtain the quantitative wave functions of the excitons and biexcitons. Notice also that the factor $\omega^{1/2}$ in the definition of $g_{1,2}$ is included in $E^\pm$. A short review of the approximations involved in our Hamiltonian is in order here. First, the spatial dependence of the matrix elements is neglected throughout the paper. As a result of the approximation the ensemble averages $\langle B^\dagger B\rangle$ and $\langle b^\dagger b\rangle$ are the number density of biexcitons and excitons, respectively. In principle, one must include the external field and diagonalize the exciton-field interaction in H' to introduce polaritons. The polaritons of different branches would then replace the E^+ and b in the interaction involving the biexcitons. Our assumption that $E^\pm$ are c numbers makes a complete treatment of polaritons impossible. However, a full account only introduces a shift of ω_x and ω_m as demonstrated in Ref. 21, and should not affect the nonlinear part of the dielectric function ϵ. Also since the actual values ω_x and ω_m are determined experimentally, whatever effects the self-consistent treatment introduces should be included in the empirical parameters. Obviously, our model is consistent with the earlier works on the OB in CuCl.[13,15,21]

We set up the Langevin equations for the b and B by neglecting the fluctuations but introducing relaxation widths γ_x and γ_m

$$-i\left\langle\frac{\partial b}{\partial t}\right\rangle=-\delta\langle b\rangle-ig_1E^++ig_2E^-\langle B\rangle+i\langle b\rangle\gamma_x\ , \tag{4}$$

$$-i\left\langle\frac{\partial B}{\partial t}\right\rangle=-\Delta\langle B\rangle-ig_2E^+\langle b\rangle+i\langle B\rangle\gamma_m\ , \tag{5}$$

where $\delta=\omega_x-\omega$ and $\Delta=\omega_m-2\omega$. The dielectric function $\epsilon^\pm$ for $E^\pm$ is defined by

$$\epsilon^\pm=1+4\pi\langle P^\pm\rangle/E^\pm\ , \tag{6}$$

where in the rotating-wave approximation, the polarization $P^\pm$ is obtained by writing $H'\equiv-P^+E^--P^-E^+$ and

$$\langle P^+\rangle=ig_1\langle b\rangle+ig_2\langle b^\dagger B\rangle\ . \tag{7}$$

The incident wave is $\sim E^+$ throughout the paper, and ϵ^+ is to be calculated.

The major difference of our work and others in the calculation of $\epsilon^\pm$ is that either it is assumed that[13]

$$\langle b^\dagger B\rangle=0 \tag{8}$$

or in Eq. (7) (Ref. 15)

$$\langle b^\dagger B\rangle=\langle b^\dagger\rangle\langle B\rangle\ , \tag{9}$$

which introduces complex poles in both upper and lower planes and violates the Kramers-Kronig relations. Instead, we take another Langevin equation with damping constant γ,

$$-i\left\langle\frac{\partial}{\partial t}(b^\dagger B)\right\rangle=(\delta-\Delta+i\gamma)\langle b^\dagger B\rangle-ig_1E^-\langle B\rangle-ig_2E^+(\langle b^\dagger b\rangle-\langle B^\dagger B\rangle)\ . \tag{10}$$

The populations $\langle b^\dagger b\rangle$ and $\langle B^\dagger B\rangle$ can be determined by taking another set of Langevin equations as done in Sec. III. In the thermal equilibrium approach, which is used for a comparison later, we take

$$\langle b^\dagger b\rangle\approx\langle b^\dagger b\rangle_0,\ \ \langle B^\dagger B\rangle\approx\langle B^\dagger B\rangle_0\ , \tag{11}$$

where $\langle\ \rangle_0$ is the equilibrium value.

The steady-state solution of Eqs. (10), (4), and (5) are given by

$$\langle b\rangle=\frac{-ig_1E^+\Delta'}{\delta\Delta'-|g_2|^2|E|^2}\ , \tag{12}$$

$$\langle B^\dagger\rangle=\frac{-g_1g_2(E^+)^2}{\delta\Delta'-|g_2|^2|E|^2}\ , \tag{13}$$

where $\Delta'=\Delta-i\gamma_m$, $|E|^2=E^+E^-$, and γ_x is taken to be zero to avoid unnecessary enlargement of the parameter space. In general, $\gamma_x<\gamma_m<\gamma$, but even for the case $\gamma_x\approx\gamma_m$, γ_x does not seriously affect the results, since $\delta\gg\Delta$. We therefore take $\gamma_x=0$ throughout in order to proceed with the analysis unencumbered,

$$\langle b^\dagger B\rangle=\frac{-ig_1^2}{\delta-\Delta+i\gamma}\,\frac{g_2|E|^2E^+}{\delta\Delta'-|g_2|^2|E|^2}-ig_2E^+S_0\ , \tag{14}$$

$$\epsilon^+=1+4\pi\left[\frac{g_1^2\Delta'}{\delta\Delta'-|g_2|^2|E|^2}+\frac{g_1^2g_2^2|E|^2}{(\delta\Delta'-|g_2|^2|E|^2)(\delta-\Delta+i\gamma)}+g_2^2S_0\right], \tag{15}$$

$$S_0=\frac{\langle B^\dagger B\rangle_0-\langle b^\dagger b\rangle_0}{\delta-\Delta+i\gamma}\ , \tag{16}$$

and $\langle\ \rangle_0$ is the equilibrium expectation value.

A comparison of Eq. (15) with the earlier works shows that in KH, the last two terms are neglected, which is justified for large width γ or small lifetime of exciton-biexciton pair. It is to be noted that γ may have significant contributions from collisions of the exciton-biexciton pairs with other particles, as well as radiative damping. In Ref. 15, S_0 is dropped; in addition, the term proportional to $g_2^2|E|^2$ from $\langle b^\dagger B\rangle$ has a denominator $(\delta\Delta-g_2^2|E|^2)^2$ which originated from the unjustified factorization approximation given in Eq. (9). In principle, one should have another set of equations of motion to determine the expectation values in S_0 instead of taking the equilibrium value. This procedure is not taken to avoid more parameters.[23]

III. LOCAL-FIELD CORRECTION

We will briefly review the major effect of the local-field correction (LFC) before applying it to our problem. The correction relates the macroscopic field E, which appears in Maxwell's equations, to the local-field E' which induces the polarization P,

$$\langle\vec{P}\rangle=\rho\alpha\vec{E}'\ , \tag{17}$$

where ρ is the density of the molecules and α is the molecular polarizability. The origin of the LFC is discussed in detail by Van Kronendonk and Sipe[16] where it appears as removal of the self-field of an atom, at a specified position, from the macroscopic field E to give the local-field E' which is responsible for the induced dipole moment of the atom as given in Eq. (17). The correction they derive is

$$\vec{E}'=\vec{E}+(4\pi/3+S')\langle\vec{P}\rangle\ , \tag{18}$$

where S' is a term which depends upon the relative location of surrounding atoms; $S'=0$ for cubic or spherical symmetry. In what follows, we shall take $S'=0$ in (18) and use the LFC without apology.

From Eqs. (17) and (18) we have

$$\vec{E}'=\vec{E}(1-\tfrac{4}{3}\pi\rho\alpha)^{-1}\ , \tag{19}$$

$$\langle\vec{P}\rangle=\vec{E}\rho\alpha(1-\tfrac{4}{3}\pi\rho\alpha)^{-1}\ , \tag{20}$$

and

$$\epsilon=1+4\pi\rho\alpha(1-\tfrac{4}{3}\pi\rho\alpha)^{-1}\ . \tag{21}$$

The LFC, therefore, is carried out by computing $\rho\alpha$ at first and introducing $(1-\frac{4}{3}\pi\rho\alpha)^{-1}$ in various places. In practice, if there are many different electronic states localized at the same position, and if only one particular state is of interest, say, the state with the lowest eigenfrequency $(\rho_0\alpha_0)$, then ϵ can be written as

$$\epsilon=1+\frac{4\pi\sum_i\rho_i\alpha_i}{1-\frac{4}{3}\pi\sum_i\rho_i\alpha_i}\equiv\epsilon_\infty+\frac{4\pi\rho_0\alpha_0}{1-\frac{4}{3}\pi\sum_i\rho_i\alpha_i}\ . \tag{22}$$

In general, ϵ_∞ is known ($\epsilon_\infty=5$ for CuCl), then Eq. (22) is equivalent to

$$\epsilon=\epsilon_\infty+z_0 4\pi\rho_0\alpha_0\ , \tag{23}$$

where we have used

$$z_0=\frac{\epsilon_\infty+2}{3}\frac{1}{1-\frac{4}{3}\pi\rho_0\alpha_0}=\left[1-\tfrac{4}{3}\pi\sum_i\rho_i\alpha_i\right]^{-1}\ , \tag{24}$$

and the factor z_0 is the general LFC term for $\epsilon_\infty>1$.

Before the LFC is applied to the calculation of ϵ for CuCl we must emphasize the assumptions made in the formalism. First, the excitons and biexcitons must be localized (Frenkel excitons), for obvious reasons. Second, the constituents that contribute to ϵ_∞ are assumed to be localized in the same position as those for α_0. How to verify these assumptions for excitons and biexcitions in CuCl is difficult. Our discussion can be summarized by modification of Eq. (15) according to the LFC to give

$$\epsilon^+=\epsilon_\infty+4\pi z_0\left[\frac{g_1^2\Delta'}{\delta\Delta'-g_2^2|E_c|^2}+\frac{g_1^2g_2^2|E|^2}{(\delta\Delta'-g_2^2|E_c|^2)(\delta-\Delta+i\gamma)}+g_2^2S_0\right]\ , \tag{25}$$

where

$$E_c=E \tag{26}$$

or

$$E_c=z_0E \tag{27}$$

depending on whether or not the LFC is applied to the term $E^-B^\dagger b$.

We should emphasize that the parameters g_1^2, g_2^2, etc., are inferred from the experimental data on the Raman scattering without the LFC. A consistent treatment of this effect in all cases is essential for any quantitatively reliable conclusion. In view of this remark, the following calculation on the LFC shows the importance of the effect but should not be taken as exhibiting a quantitatively reliable representation of ϵ^+.

Also, it is probable that in the Raman scattering experiments, the intensity is sufficiently low that the inferred values g_1^2 and g_2^2 are field independent. Thus, we take the parameters as though they stem from first principles and therefore introduce z_0.

IV. APPLICATION TO CuCl

In order to exhibit the essential features of the effects of the FEB interaction and the LFC on intrinsic (absence of cavity mirrors) OB in CuCl, the transmittance τ is computed using the results of the previous sections. In the mean-field approximation,[24] which is valid for small

volumes and sufficiently low absorption, the transmittance τ is given by[13,25]

$$\tau=\frac{I_t}{I_0}=\left|\frac{4(\epsilon^+)^{1/2}}{[1+(\epsilon^+)^{1/2}]^2e^{\delta''-i\delta'}-[(\epsilon^+)^{1/2}-1]^2e^{-\delta''+i\delta'}}\right|^2. \tag{28}$$

In this expression ϵ^+ is the complex dielectric function given by (25) and

$$\delta=\delta'+i\delta''=dn\omega/c \tag{29}$$

is the phase shift through the medium of the field at normal incidence. Here, the width of the medium is d, ω is the frequency of the incident field, c is the velocity of light in vacuum, and n is the complex index of refraction in the medium. The quantities I_t and I_0 are the intensities of the transmitted and normally incident fields, respectively. The reflection coefficient R associated with the material boundary is determined using the complex index of refraction $n=(\epsilon^+)^{1/2}$,

$$R=\left|\frac{1-n}{1+n}\right|^2. \tag{30}$$

Consistent with the mean-field approach of KH, the transmitted intensity I_t is expressed in terms of the polariton number as N_p,[26]

$$I_t=(1-R)N_p\hbar\omega V_E\ , \tag{31}$$

where the polariton number $N_p=|E|^2/\omega$ and V_E is the polariton energy velocity.[27]

A. Contribution of field—exciton-biexciton interaction

The contribution to intrinsic OB in CuCl of the correction term to the FEB contribution, the second term in the large parentheses in Eq. (25), which is neglected by KH, is shown in Fig. 1. In this case we have neglected the LFC, i.e., set $z_0=1$. Curve a is the uncorrected results and corresponds to identical conditions treated by KH for the incident field frequency chosen in value above frequency threshold for OB. Qualitatively, our result is consistent with theirs. Quantitatively, there is a large discrepancy. Also, we chose the effective width of the biexciton to conform to the value for ω at threshold for intrinsic OB exhibited in the results of KH. This gives a value for γ_m nearly an order of magnitude smaller than that used by KH. The values of all the other material parameters are the same as those used by KH, with the further exception of γ. Curve b of Fig. 1 shows intrinsic OB with a threshold of $I_0\approx 12$ MW/cm^2 in contrast to the threshold at approximately 0.14 MW/cm^2 of KH.

The result of adding the correction to the FEB interaction in Eq. (25) is shown by curve b in Fig. 1 for the effective width γ of the exciton-biexciton coupling approximately 2 orders of magnitude larger than γ_m. Even for such a relatively large width γ the correction is nonnegligible, exhibiting successively higher-order bistable regions characteristic of dispersive OB in a Fabry-Perot cavity filled with a nonlinear medium with low absorption. Thus, inclusion of the FEB correction term in (25) strongly modifies the real part of the complex phase shift of the field in the medium. For a still larger width γ, about 3

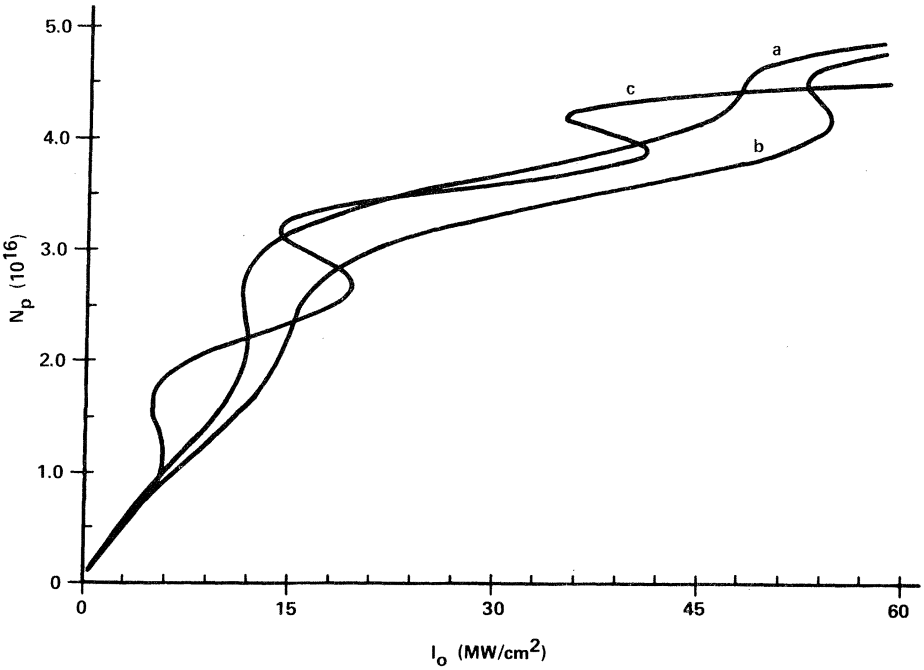

FIG. 1. Polariton number N_p vs intensity of the incident field I_0 for the incident field frequency $\omega=3.18585$ eV, and without local-field correction. Curve a: neglect of exciton-biexciton interaction correction (see text). Curve b: the same conditions as for curve a except that $\gamma_m/\gamma=2.9\times10^{-2}$. Curve c: the same conditions as for curve a except that $\gamma_m/\gamma=2.9\times10^{-3}$. The parameters are $4\pi g_1^2=2.75\times10^{-2}$ eV; $|M|^2=1.572\times10^{-22}$ eV^{2}cm^3 ($N_P|M|^2=g_2^2|E|^2$); $\gamma_m=0.4\times10^{-4}$ eV; $\omega_x=3.2027$ eV; and $\omega_m=6.3725$ eV.

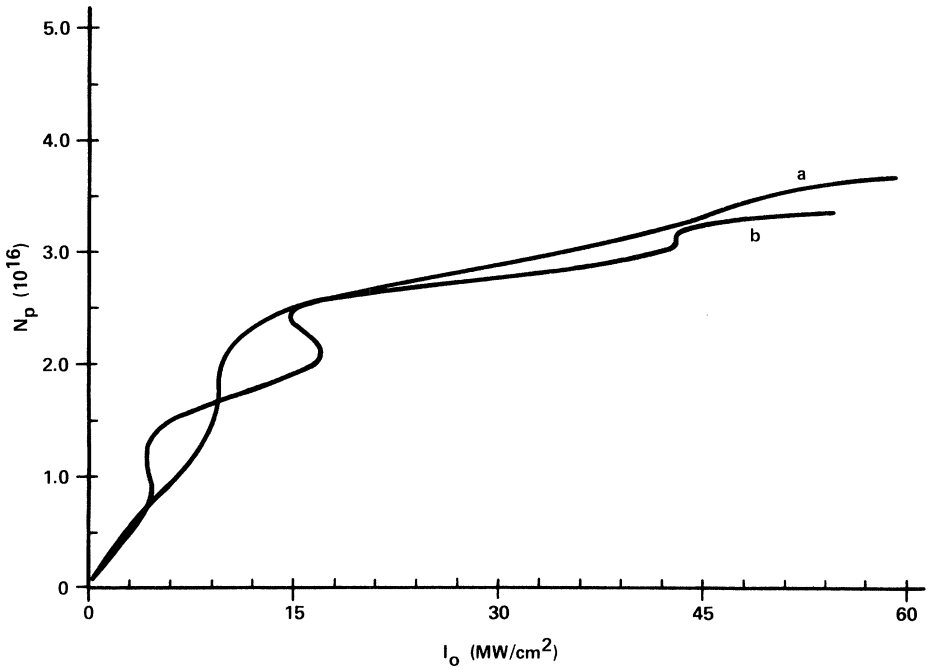

FIG. 2. Polariton number N_p vs intensity of the incident field I_0 for the incident field frequency $\omega=3.18594$ eV, and without local-field correction. Curve *a*: neglect of the FEB correction (see text). Curve *b*: inclusion of exciton-biexciton interaction correction with $\gamma_m/\gamma=2.9\times10^{-2}$.

orders of magnitude larger than γ_m, the effect of the correction is significant as shown by curve *c* in Fig. 1. Overall, the effect of the correction to the FEB contribution is to shift the threshold for OB to lower input intensity I_0, to enhance the difference between the switch-up and switch-down input intensities and to diminish the change in output intensity at switching.

When the incident field frequency ω is chosen to correspond to a value below threshold under the conditions treated by KH, the result is curve *a* of Fig. 2. In this case, there is no intrinsic OB. If, however, the FEB correction [the second term in the large parentheses in (25)] is incorporated in the same way as for Fig. 1, curve *b* results. The main point here is that inclusion of the correction term shifts the threshold frequency for OB to higher values.

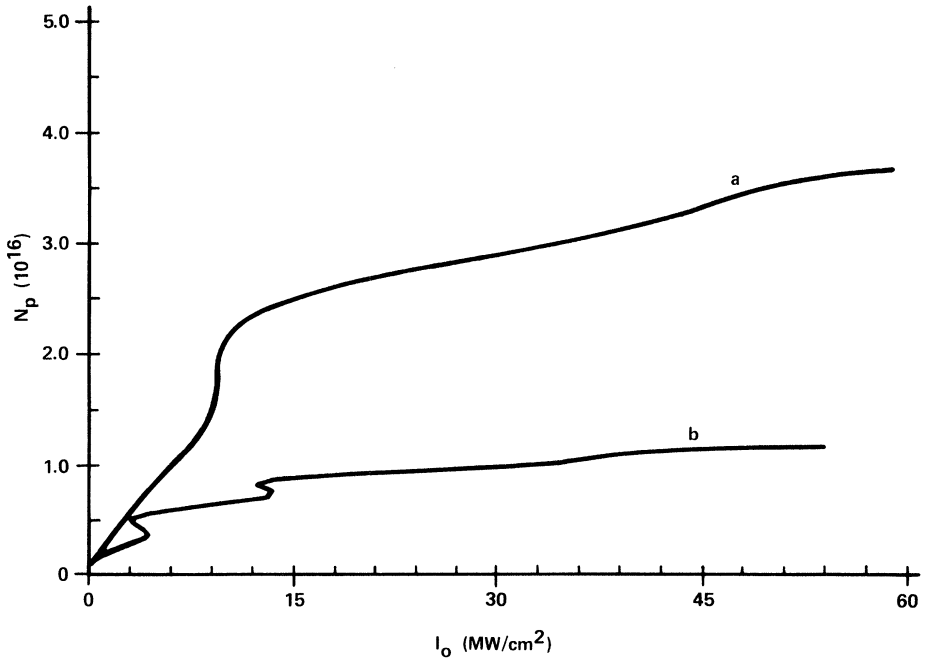

FIG. 3. Polariton number N_p vs intensity of the incident field I_0 for the incident field frequency $\omega=3.18594$ eV, and without the FEB correction. Curve *a*: without the LFC. Curve *b*: LFC applied to the exciton sites. All other parameters are the same as for Fig. 1.

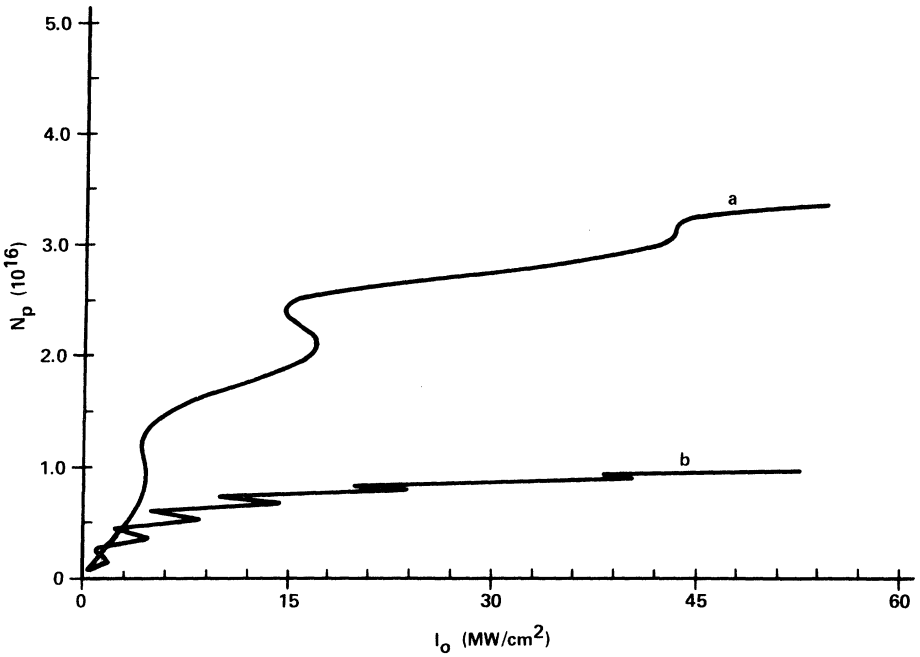

FIG. 4. Polariton number N_p vs intensity of the incident field I_0 for the incident field frequency $\omega=3.18594$ eV, and with the FEB correction included (see text). Curve *a*: without LFC. Curve *b*: LFC applied to the exciton sites. All other parameters are the same as for Fig. 1.

B. Local-field correction

The LFC in (25) and (28) is applied to the excitons only, to demonstrate the importance of the effect. Application of the LFC to the FEB interaction, Eq. (27), as well, would drastically change the results. However, whereas the exciton sites can be thought of as well localized in CuCl, the biexcitons are not really local entities, and it is not clear what a LFC means with respect to them. The field that drives the FEB contribution is then more like the macroscopic field in the crystal. The results for intrinsic OB are shown in Fig. 3. Here we have neglected the FEB [second term in parentheses in (25)]. The conditions treated by KH below frequency threshold are represented in curve *a*, which is identical to curve *a* of Fig. 2. Curve *b* shows the effect of the LFC under otherwise identical conditions that resulted in curve *a*.

If the FEB correction is incorporated in the calculation, the result is depicted in Fig. 4. Without any LFC, the result is curve *a*. This is the same as curve *b* of Fig. 2, and is included for comparison with curve *b*, Fig. 4, which is the result of including the LFC for excitons. All other conditions are identical with those associated with curve *a*. Comparing the curve *b* of Figs. 3 and 4, it is seen that the LFC in this case also causes hysteresis cycles at higher input fields due to the combined contribution with exciton-biexciton interaction correction in our calculations.

V. CONCLUSIONS

We have studied OB in the CuCl system where the properties of the biexcitons are well demonstrated.[14] Our treatment takes into account the FEB interaction and resolves the difficulties of some earlier works. Despite the small magnitude, the effect on the threshold is substantial as shown in this paper. We also have used the available parameters to investigate the LFC, which also turns out to be important for CuCl where excitons are presumably very much localized.

Although our calculation is very general and the approximations involved should not invalidate a quantitative prediction of possible experimental data, this cannot be done without clarification of several key issues. First, there is some debate as to the field dependence and magnitude of γ_m.[28] Consideration of this issue will certainly change our conclusion. The calculated OB being sensitive to the parameters is subject to the uncertainty of the analysis of the experimental data on the Raman scattering.[29] The second and also a related issue is that the LFC should be included in the analysis of the experimental data consistently to determine the relevant parameters. Finally, the calculation for a cavity of larger dimension for which the mean-field approximation is not justified calls for a different treatment. We plan to do some numerical work on this subject in the future.

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