

## Quantum theory of excitonic optical bistability

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We present a fully-quantum-mechanical theory of the intracavity interaction of coherent light with semiconductors. Interaction is assumed to occur via excitons. Using a master-equation approach we include such effects as exciton-lattice and exciton-exciton interactions. In the two cases of high and low exciton densities, steady-state analysis reveals bistability and hysteresis in the system. Bistability of a dispersive and absorptive nature in both exciton number and output intensity, dependent on input intensity is found. The bistability displayed in the high-density case agrees qualitatively with the experimental curves obtained by Gibbs *et al.* for GaAs. As yet the low-density bistability has not been experimentally observed.

### I. INTRODUCTION

Optical bistability has been predicted in several systems.<sup>1,2</sup> The first experimental verification involved Na vapor,<sup>3</sup> and since then it has been shown that other systems, involving nonlinear media<sup>4,5,6</sup> and three-level atoms,<sup>7</sup> should exhibit bistability.

Recently, experiments by Miller *et al.*<sup>8</sup> and Gibbs, McCall, Venkatesan, Gossard, Passner, and Weigmann<sup>9</sup> illustrated the occurrence of the phenomenon in semiconductors. The experiments of Miller *et al.* dealt with InSb, in which the mechanism for bistability was assumed to be a pseudo-two-level system existing between valence and conduction bands. Thus the theory follows as in two-level atom bistability. Gibbs *et al.*, however, conducted experiments using GaAs. In this case, interaction between the light field and excitons was seen to give rise to bistability. They show<sup>10</sup> that excitons in GaAs exhibit Bloch-like saturation.

Following the work of Hanamura<sup>11</sup> we present a microscopic theory of the intracavity interaction of coherent light and excitons. We start with a many-fermion Hamiltonian, describing the semiconductor system. By means of a modified Marumori transformation<sup>12,13</sup> we map the system to a boson space in which the Hamiltonian is written in terms of exciton operators.

To deal with infinite operator expansions in this transformation we consider two limiting cases. Firstly, the low-density case, in which the product  $\rho a_0^3$  ( $\rho$  = exciton density,  $a_0$  = Bohr radius), is small. This is not applicable to GaAs, but ade-

quately describes other systems, e.g., Cds.<sup>14</sup> Secondly, we discuss the high-density case, a more accurate description of GaAs.

We overcome the difficulties associated with a many-particle system by using a reservoir technique<sup>15</sup> to derive a master equation. That is, we assume one exciton is strongly coupled to the field and let all other exciton modes form a thermal bath to which the exciton mode of interest is weakly coupled. In this way we can also introduce radiative damping of exciton and field modes in a consistent fashion.

It is possible to identify terms describing exciton-exciton interactions (i.e. collisions), exciton-phonon, and exciton-photon interactions in the resulting master equation. We may then observe the effect of such processes on the system's behavior.

Utilizing the complex  $P$  representation,<sup>16</sup> we derive a Fokker-Planck equation from the master equation. The corresponding stochastic equations contain all information about the system's quantum fluctuations.

The solution of the steady-state deterministic equations (neglecting noise) reveals bistability. In the low-density case, exciton-exciton interaction is shown to be a necessary condition and purely absorptive bistability is possible if the exciton-exciton collisions dominate. At high densities such interactions are not necessary to observe dispersive bistability, although they are still required for absorptive bistability.

Another theory of optical bistability in semiconductors was recently presented by Goll and Haken.<sup>17</sup> They derive Heisenberg equations of motion

for exciton operators: bilinear combinations of fermion operators—in contrast to the boson operators of our theory. Damping is included phenomenologically. Their approach provides an alternative description of bistability: the equations of motion

resemble optical Bloch equations. At high exciton densities the results are similar to ours, but the low-density bistability we predict is not predicted by Goll and Haken.

## II. THE MODEL

### A. Low-density case: Hamiltonian

We consider the intracavity interaction of a coherent driving field with a semiconductor. It is assumed that the interaction between light and semiconductor occurs via excitons, thus we develop our theory in terms of these excitons. For convenience, discussion is limited to a two-band semiconductor. The light field may then excite an electron from the filled valence band to the conduction band (thereby creating a hole in the valence band) and so create an exciton.

The Hamiltonian for such a system takes the general form,<sup>18,19</sup>

$$H = \int d^3\vec{x} \psi^\dagger(\vec{x}) H_s(\vec{x}) \psi(\vec{x}) + \frac{1}{2} \int d^3\vec{x}_1 d^3\vec{x}_2 \psi^\dagger(\vec{x}_1) \psi^\dagger(\vec{x}_2) V(\vec{x}_1 - \vec{x}_2) \psi(\vec{x}_2) \psi(\vec{x}_1), \quad (2.1)$$

where  $H_s$  is the single-particle Hamiltonian and  $V$  represents the Coulomb interaction potential.

As the semiconductor is characterized only by conduction electrons and valence holes we may, in a second quantized theory, express the operators of the wave field as

$$\psi(\vec{x}) = \sum_k a_k \varphi_k(\vec{x}) + \sum_{k'} d_{k'} \varphi_{k'}(\vec{x}), \quad (2.2)$$

where the  $\varphi_k$   $\varphi_{k'}$  are single-particle wave functions (eigenvectors of  $H_s$ ),  $a_k$  is the fermion destruction operator for a conduction electron with momentum  $k$ , and  $d_{k'}$  is the fermion destruction operator for a valence hole with momentum  $k'$ .

In this way, we find the Hamiltonian for a many-fermion system in momentum space,<sup>11</sup>

$$H = \sum_{p_1} E_c(p_1) a_{p_1}^\dagger a_{p_1} - \sum_{p_2} E_v(p_2) d_{p_2}^\dagger d_{p_2} + \frac{1}{2} \sum_{\substack{p_1, p_2, q \\ p'_1, p'_2}} V(q) (a_{p_1+q}^\dagger a_{p'_1-q}^\dagger a_{p_1} a_{p_1} + d_{p_2}^\dagger d_{p_2}^\dagger d_{p_2-q} d_{p_2+q} - 2a_{p_1+q}^\dagger d_{p_2}^\dagger d_{p_2-q} a_{p_1}), \quad (2.3)$$

where

$$E_c(p_1) = \frac{\hbar^2 p_1^2}{2m_e} + E_g$$

( $m_e$  represents the effective electron mass and  $E_g$  represents the band gap energy),

$$E_v(p_2) = -\frac{\hbar^2 p_2^2}{2m_h}$$

( $m_h$  represents the effective hole mass), and represents the band gap energy),

$$v(q) = \frac{4\pi e^2}{\epsilon_0 q^2}$$

( $\epsilon_0$  represents the static dielectric constant).

The first two terms in Eq. (2.3) describe the free Hamiltonian. The third term describes interaction between electrons and holes: the first term in brackets concerns electron-electron interactions, the second term concerns hole-hole interactions, and the third term describes electron-hole interactions. We note that this Hamiltonian is equivalent to that derived by Haken.<sup>19</sup>

The Hamiltonian describing interaction between light and semiconductor has the general form

$$H_{\text{int}} = \int \psi^\dagger(\vec{x}) \vec{A} \cdot \vec{p} \psi(\vec{x}) d^3x, \quad (2.4)$$

where  $\vec{A}$  is quantized in terms of boson operators  $\hat{a}$ . Assuming it is possible to single out a particular mode of the light field (whose operator is given by  $\hat{a}$ ) we find in the rotating-wave approximation,

$$H_{\text{int}} = \hbar \sum_{p_1 p_2} a_{p_1}^\dagger d_{p_2}^\dagger \hat{a} g + \text{H.c.}, \quad (2.5)$$

where  $g$  is the coupling constant which does not depend on  $p_1$  and  $p_2$  because of the single mode approximation.

We also include a free Hamiltonian term for the field,

$$H_{0,F} = \omega_2 \hat{a}^\dagger \hat{a}$$

and our Hamiltonian must include a term describing coupling between the external driving field and the field mode,

$$H_D = i\hbar [E \exp(-i\omega_L t) \hat{a}^\dagger - \text{H.c.}], \quad (2.6)$$

Thus, the external field drives the field (cavity) mode described by  $\hat{a}$ , which in turn is coupled to the electrons and holes of the medium. So far, discussion has been in terms of electrons and holes and not excitons as such. We now wish to formulate the system in terms of excitons.

For small radius (Frenkel) excitons, it is possible to treat the fermion pairs  $a_i d_j$  as pseudospin operators.<sup>19,20</sup> This naturally leads to a theory very similar to that of a two-level atom (i.e., Bloch-like equations). However, semiconductors possess excitons of large radius (Wannier) thus we must include effects of nonlocalized electrons and holes.

We could, following many authors,<sup>18,21,22</sup> introduce an exciton operator:

$$C_k^\dagger = \sum_{p_1 p_2} \delta_{k, p_1 - p_2} \varphi(k) a_{p_1}^\dagger d_{p_2}^\dagger, \quad (2.7)$$

where  $\varphi$  is a wave function of the ground state of a hydrogenlike exciton. This has the commutation relation<sup>21</sup>

$$[C_k, C_k^\dagger] = 1 - O(Na_0^3),$$

where  $N$  represents the concentration of electrons and holes and  $a_0$  represents the Bohr radius. Thus, for small carrier densities we may treat the operators  $C_k$  as bosons. However, in many cases this approximation breaks down and other approaches must be employed.

Haken *et al.*<sup>22</sup> formulate a theory of Wannier excitons in terms of operators  $C$ ,  $C^\dagger$  and,

$$N = \frac{1}{2}(N_e + N_h),$$

$$N_e = \sum_l \varphi_l a_l^\dagger a_l, \quad N_h = \sum_l \varphi_l d_l^\dagger d_l$$

by deriving Heisenberg equations of motion for the operators.

However, we wish to derive a master equation for the system in order to deal with many-body effects and damping in a consistent manner. This is only possible if the Hamiltonian is expressed in terms of exciton operators which have a closed set of commutation relations. This is most easily achieved if we transform the fermion pair operators to boson operators.

## B. Transformation to boson systems

Using a modified version of the transformation due to Marumori *et al.*<sup>12</sup>

$$U = |0\rangle\langle 0| \sum_{n=0}^{\infty} \frac{1}{n!} \frac{1}{\sqrt{n!}} \left[ \sum_{\alpha\beta} b_{\alpha\beta}^\dagger d_{\beta} a_{\alpha} \right]^n |0\rangle\langle 0| \quad (2.8)$$

(where  $b_{\alpha\beta}^\dagger$  represents the exciton (boson) creation operator,  $a_{\alpha}^\dagger$  represents the conduction electron (fermion) creation operator,  $d_{\beta}^\dagger$  represents the valence hole (fermion) creation operator.  $|0\rangle$  is a fermion state,  $|0\rangle$  is a boson state), we form an equivalence between fermion pair operators and boson operators.

The transformation  $U$  is antisymmetrized in such a way as to ensure a one-to-one correspondence between boson and fermion states. The resulting Hamiltonian is also Hermitian.

The theory of the transformation  $U$  is discussed in Appendix A.

An important case of the transformation is,

$$U a_{\alpha}^\dagger d_{\beta}^\dagger U^\dagger = \left[ b_{\alpha\beta}^\dagger - \sum_{\gamma\eta} b_{\alpha\gamma}^\dagger b_{\beta\eta}^\dagger b_{\gamma\eta} \right] \times \left[ 1 + \sum_{\alpha\beta} b_{\alpha\beta}^\dagger b_{\alpha\beta} \right]^{-1/2} \hat{P}, \quad (2.9)$$

where  $\hat{P}$  is a projection operator, equal to unity in the boson subspace. We note that Eq. (2.9) gives rise to an infinite expansion.

In his theory of Wannier excitons, Hanamura<sup>11</sup> uses a transformation due to Usui,<sup>23</sup>

$$U' = |0\rangle\langle 0| \left[ \exp \left[ \sum_{\alpha} \sum_{\beta} b_{\alpha\beta}^\dagger d_{\beta} a_{\alpha} \right] \right] |0\rangle\langle 0|, \quad (2.10)$$

which generates finite operator expansions. However, there is no longer a one-to-one correspondence between boson and fermion spaces, and the transformed Hamiltonian is not Hermitian. Hanamura solves these problems by using an ordering procedure.

However, we avoid these difficulties by using the transformation  $U$  and deal with the infinite expansions by considering particular limiting cases. Consider the expansion of the right-hand side of Eq. (2.9) [see Eq. (A.16), Appendix A]:

$$b_{\alpha\beta}^\dagger \left[ 1 - (1 - \sqrt{2}) \sum_{ij} b_{\alpha_i\beta_j}^\dagger b_{\alpha_i\beta_j} + \frac{1}{2} (1 - 2\sqrt{2} + \sqrt{3}) \sum_{\substack{ijn \\ m}} b_{\alpha_i\beta_j}^\dagger b_{\alpha_n\beta_m}^\dagger b_{\alpha_i\beta_j} b_{\alpha_n\beta_m} + \dots \right] \hat{P}; \quad (2.11)$$

the terms increase in powers of  $b^\dagger b$ , or exciton intensity. Thus for low-density systems, higher-order terms will become negligible and we may expand to second order only:

$$= (2.11) \Rightarrow \left[ b_{\alpha\beta}^\dagger - (1 - \sqrt{2}) b_{\alpha\beta}^\dagger \sum_{ij} b_{\alpha_i\beta_j}^\dagger b_{\alpha_i\beta_j} \right] \hat{P}. \quad (2.12)$$

Thus, applying the transformation  $U$ , we find, in the low-density case,

$$H \rightarrow \tilde{H} \hat{P},$$

where

$$\tilde{H} = H_0 + H_{e,h} + H_{\text{int}} + H_D \quad (2.13)$$

and

$$\begin{aligned} H_0 &= \sum_{p_1 p_2} [E_c(p_1) - E_v(p_2)] b_{p_1 p_2}^\dagger b_{p_1 p_2} - \sum_{\substack{p_1 p_2 \\ q}} V(q) b_{p_1+q p_2+q}^\dagger b_{p_1 p_2} + \omega_2 \hat{a}^\dagger \hat{a}, \\ H_{e,h} &= \frac{1}{2} \sum_{\substack{p_1 p_2 q \\ p'_1 p'_2}} V(q) (b_{p'_1 - q p_2}^\dagger b_{p_1 + q p_2}^\dagger + b_{p_1 p_2 + q}^\dagger b_{p'_1 p'_2 - q}^\dagger - 2b_{p_1 + q p_2}^\dagger b_{p'_1 p'_2 + q}^\dagger) b_{p'_1 p'_2} b_{p_1 p_2} \\ &\quad - \frac{1}{2} \sum_{\substack{p_1 p_2 q \\ p'_1 p'_2}} V(q) (b_{p_1 + q p_2}^\dagger b_{p'_1 - q p_2}^\dagger + b_{p_1 p_2 - q}^\dagger b_{p'_1 p'_2 + q}^\dagger) b_{p'_1 p'_2} b_{p_1 p_2} \\ &\quad + \frac{1}{2} \sum_{\substack{p_1 p_2 q \\ p'_1 p'_2}} V(q) (b_{p_1 + q p'_2 + q}^\dagger b_{p'_1 p_2}^\dagger + b_{p_1 + q p'_2}^\dagger b_{p'_1 p_2 + q}^\dagger) b_{p'_1 p'_2} b_{p_1 p_2}, \\ H_{\text{int}} &= \hbar \left[ g \hat{a} \sum_{p_1 p_2} [b_{p_1 p_2}^\dagger - (1 - \sqrt{2}) \sum_{p'_1 p'_2} b_{p_1 p_2}^\dagger b_{p'_1 p'_2}^\dagger b_{p'_1 p'_2}] + \text{H.c.} \right], \\ H_D &= i \hbar [E \exp(-i\omega_L t) \hat{a}^\dagger - \text{H.c.}]. \end{aligned}$$

The last two terms of  $H_{e,h}$  are included to account for the effect of the Pauli exclusion principle.<sup>11</sup> They stem primarily from antisymmetrization requirements. This Hamiltonian is essentially equivalent to that of Hanamura. Following Hanamura, we utilize the transformation

$$b_{p_1 p_2} = \sum_{\nu k} \frac{1}{\sqrt{v}} \delta_{k, p_1 - p_2} f_\nu(\alpha p_1 + \beta p_2) C_{\nu, k}, \quad (2.14)$$

where  $v$  is the volume of the system,  $\gamma$  is the quantum number of the hydrogenlike state,  $\alpha = m_h / (m_e + m_h)$ ,  $\beta = 1 - \alpha$ , and  $f_\nu(\chi)$  is the wave function of the state  $\nu$  of the hydrogenlike state. In particular,  $f_0(\chi)$  (ground-state wave function of the hydrogenlike state) is  $8\sqrt{\pi a_0^3} / (1 + \chi^2 a_0^2)^2$ . The transformation (2.14) diagonalizes the excitonic harmonic part of the Hamiltonian to

$$H_0 = \sum_{\nu, k} \left[ E_g - \epsilon_{b, n} + \frac{\hbar^2 k^2}{2M} \right] C_{\nu, k}^\dagger C_{\nu, k} \equiv \sum_{\nu, k} \Omega_{\nu, k} C_{\nu, k}^\dagger C_{\nu, k}, \quad (2.15)$$

where  $k$  is center of mass momentum,  $\epsilon_{b, n}$  is the binding energy of  $n$ th excitonic state,  $M = m_e + m_h$ , and  $E_g$  is the band gap energy. The Hamiltonian then becomes

$$\begin{aligned} \tilde{H} = & \sum_{\nu, k} \Omega_{\nu, k} C_{\nu, k}^\dagger C_{\nu, k} + \omega_2 \hat{a}^\dagger \hat{a} + \sum_{\substack{\mu, k, k', q \\ \nu, \nu', \mu'}} M_1 C_{\mu, k+q}^\dagger C_{\mu', k'-q}^\dagger C_{\nu, k} C_{\nu', k'} \\ & - \sum_{\substack{p_2, k, k', \nu, \nu' \\ p_2', \mu, \mu', q}} M_2 C_{\mu, k+p_2-p_2'+q}^\dagger C_{\mu', k'+p_2'-p_2-q}^\dagger C_{\nu, k} C_{\nu', k'} \\ & + i\hbar[E \exp(-i\omega_L t) \hat{a} - \text{H.c.}] + \hbar[g\hat{a}(r_1 c_k^\dagger - r_2 c_k^\dagger c_k^\dagger c_k) + \text{H.c.}], \end{aligned} \quad (2.16)$$

where

$$\begin{aligned} M_1 = & \frac{1}{2v^2} V(q) [f_\mu^*(\alpha(p_2+k+q) + \beta p_2) f_{\mu'}^*(\alpha(k'+p_2'-q) + \beta p_2') \\ & + f_\mu^*(\alpha(k'+p_2) + \beta(p_2+q)) f_{\mu'}^*(\alpha(k+p_2')\beta(p_2'-q)) \\ & - 2f_\mu^*(\alpha(k+p_2+q) + \beta p_2) f_{\mu'}^*(\alpha(k'+p_2') + \beta(p_2'+q))] \\ & \times f_\nu(\alpha(k+p_2) + \beta p_2) f_{\nu'}(\alpha(k'+p_2') + \beta p_2'), \\ M_2 = & \frac{1}{2v^2} V(q) [f_\mu^*(\alpha(k+p_2) + \beta(p_2'-q)) f_{\mu'}^*(\alpha(k'+p_2') + \beta(p_2+q)) \\ & - f_\mu^*(\alpha(k+p_2+q) + \beta p_2') f_{\mu'}^*(\alpha(k'+p_2'-q) + \beta p_2) \\ & - 2f_\mu^*(\alpha(k+p_2+q) + \beta p_2') f_{\mu'}^*(\alpha(k'+p_2') + \beta(p_2+q))] \\ & \times f_\nu(\alpha(k+p_2) + \beta p_2) f_{\nu'}(\alpha(k'+p_2') + \beta p_2'), \end{aligned}$$

where summation is implied over all indices and

$$r_1 = \frac{1}{\sqrt{v}} \sum_{p_1} f(\alpha p_1 + \beta(p_1 - k)), \quad r_2 = \frac{1 - \sqrt{2}}{v\sqrt{v}} \sum_{p_1} |f(\alpha p_1 + \beta(p_1 - k))|^2 f^*(\alpha p_1 + \beta(p_1 - k)).$$

In deriving  $H_{\text{int}}$  we have assumed it is possible to single out a particular exciton mode  $k$ . That is, we assume all other modes are so weakly coupled to the field that we may ignore them. We have also dropped the summation over the quantum number  $\nu$ , assuming we are dealing with one level of the exciton only (ground state). The terms  $M_1, M_2$  describe interaction between all exciton modes. This gives rise to damping which can be attributed to collisions between different excitons.

As well as this, we assume there is damping of excitons via coupling to the crystal lattice, i.e., phonons. We treat these phonon modes as a reservoir in thermal equilibrium weakly coupled to the exciton mode we are considering. The Hamiltonian describing this process takes the form

$$H_{\text{ex-damp}} = \sum_i \chi_i C_i \tau_i^\dagger + \text{H.c.}, \quad (2.17)$$

where  $\tau_i$  represents the reservoir operator.

This is similar to the exciton-phonon coupling model considered by Toyazawa. In his extensive papers,<sup>24,25</sup> he derives the Hamiltonian

$$H_{\text{ex-phon}} = \sum_{\mu, k, k'} w(k, k', \mu) (D_{\mu, k-k'} + D_{\mu, -k+k'}^\dagger) \times C_{\lambda, k}^\dagger C_{\lambda', k'}, \quad (2.18)$$

where  $D_{\mu, \nu}^\dagger$  represents the boson creation operator (phonon),  $\mu, \nu$  refer to mode and wave number of phonon,  $C_{\lambda k}^\dagger$  represents the creation operator (boson) for exciton,  $\lambda$  represents the internal quantum number, and  $k$  represents the wave number. The two Hamiltonians will be equal if we make the

identification

$$D_{\mu, k'-k} C_{\lambda k}^\dagger \rightarrow \tau_i.$$

Thus the reservoir mode is a combination of exciton and phonon modes, and damping of a particular exciton occurs via another exciton mode and not solely through the lattice.

To include damping of the field mode in our theory we assume modes in the cavity form a reservoir to which the field mode is coupled:

$$H_{\text{field damp}} = \hat{a}^\dagger \sum_j \chi_4 \tau_j^4 + \text{H.c.}, \quad (2.19)$$

where  $\tau_j^4$  represents the reservoir mode operators.

### C. Master Equation

The Hamiltonian derived in the previous section is that of a many-body system, so in order to perform any quantitative calculations we need to reduce its complexity.

This is accomplished by assuming one exciton mode only is of interest, and is strongly coupled to the field. All other exciton modes then form a thermal reservoir, weakly coupled to the mode of interest. We can then use the quantum theory of damping<sup>15</sup> to derive a Markovian master equation.

We thus write the Hamiltonian (in a frame rotating at frequency  $\omega_L$ ) as

$$\begin{aligned} \tilde{H} = & \hbar \delta_1 C^\dagger C + \hbar \delta_2 \hat{a}^\dagger \hat{a} - M_2 C^\dagger C^\dagger C C + \hbar g (r_1 \hat{a}^\dagger C - r_2 \hat{a}^\dagger C^\dagger C C) + i \hbar E \hat{a} \\ & + C^\dagger \sum_{k_1, k_2, k_3} \chi_1 C_{k_1}^\dagger C_{k_2} C_{k_3} + C^\dagger C^\dagger \sum_{k_1, k_2} \chi_2 C_{k_1} C_{k_2} + C^\dagger C \sum_{k_1, k_2} \chi_3 C_{k_1}^\dagger C_{k_2} + \hat{a}^\dagger \sum_j \chi_4 \tau_j^4 \\ & + C^\dagger \sum_k \chi_5 \tau_k^5 + \text{H.c.}, \end{aligned} \quad (2.20)$$

where  $\delta_1, \delta_2$  are detuning terms,  $\delta_1 = \Omega - \omega_L$ ,  $\delta_2 = \omega_2 - \omega_L$ ,

$$\chi_1 = M_1 - M_2, \quad \chi_2 = M_1(k_1 = k_2 - 2q) - M_2(k_1 = k_2 + 2p'_2 - 2p_2 - 2q),$$

$$\chi_3 = M_1(k_1 + q = k_2) - M_2(p_2 - p'_2 + q = 0) - M_2(k_2 = k_1 + p_2 - p'_2 + q),$$

and  $\tau_j^4$  represents the boson reservoir operator of the form (2.19),  $\chi_4$  represents the coupling between field mode and reservoir,  $\tau_k^5$  represents the exciton-phonon reservoir operator as in (2.17), and  $\chi_5$  represents the coupling between exciton mode and exciton-phonon reservoir.

We have ignored the summation over the internal quantum numbers, assuming it is possible to consider the exciton ground-state only. The last five terms of Eq. (2.20) describe coupling of field and exciton modes to reservoirs.

Applying standard methods of the quantum theory of damping<sup>15</sup> we then find the master equation—the equation of motion for the density operator  $\rho$ :

$$\begin{aligned} \frac{\partial \rho}{\partial t} = & -i \delta_1 [C^\dagger C, \rho] - i \delta_2 [\hat{a}^\dagger \hat{a}, \rho] + \frac{i M_2}{\hbar} [C^\dagger C^\dagger C C, \rho] - i g [(r_1 \hat{a}^\dagger C - r_2 \hat{a}^\dagger C^\dagger C C) + (r_1 C^\dagger \hat{a} - r_2 C^\dagger C^\dagger C \hat{a}), \rho] \\ & + [E \hat{a}^\dagger - E^* \hat{a}, \rho] + \left. \frac{\partial \rho}{\partial t} \right|_{\text{ex-ex}} + \left. \frac{\partial \rho}{\partial t} \right|_{\text{ex-phon}} + \left. \frac{\partial \rho}{\partial t} \right|_{\text{field damp}}, \end{aligned} \quad (2.21)$$

where

$$\begin{aligned} \left. \frac{\partial \rho}{\partial t} \right|_{\text{ex-ex}} &= K_{1,a}([C\rho, C^\dagger] + [C, \rho C^\dagger]) + K_{1,b}([C^\dagger \rho, C] + [C^\dagger, \rho C]) \\ &+ K_{2,a}([CC\rho, C^\dagger C^\dagger] + [CC, \rho C^\dagger C^\dagger]) + K_{2,b}(C^\dagger C^\dagger \rho, CC) + [C^\dagger C^\dagger, \rho CC] \\ &+ K_3([C^\dagger C\rho, C^\dagger C] + [C^\dagger C, \rho C^\dagger C]), \end{aligned}$$

where  $K_{1,a} = \pi g(\omega_0) |\chi_1(\omega_0)|^2 \bar{n}_{1,a}(1 + \bar{n}_{1,b})(1 + \bar{n}_{1,c})$ ,  $g(\omega_0)$  represents the density of states of reservoir modes, and  $\omega_0$  represents the exciton resonance frequency.  $\bar{n}_{1a}$ ,  $\bar{n}_{1b}$ ,  $\bar{n}_{1c}$  are thermal populations of various reservoirs ( $\bar{n}_{1a} \neq \bar{n}_{1b} \neq \bar{n}_{1c}$ ); in general,  $\bar{n} = [\exp(\hbar\omega/k_B T) - 1]^{-1}$ . Also,

$$\begin{aligned} K_{1,b} &= \pi g(\omega_0) |\chi_1(\omega_0)|^2 \bar{n}_{1,b} \bar{n}_{1,c}, \quad K_{2,a} = \pi g(\omega_0) |\chi_2(\omega_0)|^2 (1 + \bar{n}_{2,a})(1 + \bar{n}_{2,b}), \\ K_{2,b} &= \pi g(\omega_0) |\chi_2(\omega_0)|^2 \bar{n}_{2,a} \bar{n}_{2,b}, \quad K_3 = \pi g(\omega_0) |\chi_3(\omega_0)|^2 (\bar{n}_{3,a} + \bar{n}_{3,b} + 2\bar{n}_{3,a} \bar{n}_{3,b}). \end{aligned}$$

(A detailed derivation of these expressions can be found in Ref. 15.) Thus,  $K_1$ ,  $K_2$ ,  $K_3$  describe the strength of exciton-exciton interactions (collisions), and

$$\left. \frac{\partial \rho}{\partial t} \right|_{\text{ex-phon}} = K_5 \{ (1 + \bar{n}_{\text{ex}}) ([C\rho, C^\dagger] + [C, \rho C^\dagger]) + \bar{n}_{\text{ex}} ([C^\dagger \rho, C] + [C^\dagger, \rho C]) \}, \quad K_5 = \pi g'(\omega_0) |\chi_5|^2$$

and

$$\left. \frac{\partial \rho}{\partial t} \right|_{\text{field damp}} = K_4 \{ (1 + \bar{n}) ([\hat{a}\rho, \hat{a}^\dagger] + [\hat{a}, \rho \hat{a}^\dagger]) + \bar{n} ([\hat{a}^\dagger \rho, \hat{a}] + [\hat{a}^\dagger, \rho \hat{a}]) \}, \quad K_4 = \pi g(\omega_0) |\chi_4|^2.$$

#### D. Fokker-Planck and Langevin equations

We can evaluate the system's quantum fluctuations explicitly by transforming the operator master equation into a  $c$ -number equation, the Fokker-Planck equation. This is derived via the generalized  $P$  representation.<sup>16</sup> We write:

$$\rho = \int P(\alpha_1, \beta_1, \alpha_2, \beta_2) \frac{|\alpha_1, \alpha_2\rangle \langle \beta_1^*, \beta_2^*|}{\langle \beta_1^*, \beta_2^* | \alpha_1, \alpha_2 \rangle} d\alpha_1 d\beta_1 d\alpha_2 d\beta_2 \quad (C \rightarrow \alpha_1, \hat{a} \rightarrow \alpha_2, C^\dagger \rightarrow \beta_1, \hat{a}^\dagger \rightarrow \beta_2).$$

Using this transformation, we find the Fokker-Planck equation

$$\begin{aligned} \frac{\partial P}{\partial t} &= \left[ \frac{\partial}{\partial \alpha_1} [\gamma_1 \alpha_1 + \chi \alpha_1 \alpha_1 \beta_1 + i g_1 \alpha_2 - i g_2 (\beta_2 \alpha_1^2 + 2 \alpha_2 \alpha_1 \beta_1)] \right. \\ &+ \frac{\partial}{\partial \alpha_2} (\gamma_2 \alpha_2 - E + i g_1 \alpha_1 - i g_2 \alpha_1 \alpha_1 \beta_1) + \frac{1}{2} \frac{\partial^2}{\partial \alpha_1^2} (K_1 \alpha_1^2 + 2 i g_2 \alpha_1 \alpha_2) \\ &\left. + \frac{1}{2} \frac{\partial^2}{\partial \alpha_1 \partial \beta_1} (K_2 \alpha_1 \beta_1 + \eta) + \frac{\partial^2}{\partial \alpha_1 \partial \alpha_2} (i g_2 \alpha_1^2) + \frac{1}{2} \frac{\partial^2}{\partial \alpha_2 \partial \beta_2} 2 K_4 \bar{n} + \text{c.c.} \right] P, \end{aligned} \quad (2.22)$$

where c.c. means  $\alpha_1 \rightarrow \beta_1$ ,  $\alpha_2 \rightarrow \beta_2$ ,  $i \rightarrow -i$ , and

$$\begin{aligned} \gamma_1 &= i\delta_1 - 4K_{2,b} + K_3 + K_{1,a} - K_{1,b} + K_5 = \gamma_a + i\gamma_b, \quad g_1 = gr_1, \quad g_2 = r_2 g, \\ \chi &= 2[K_{2,a} - K_{2,b} - (i/\hbar)M_2] = \chi_a + i\chi_b, \\ K_1 &= 2[(i/\hbar)M_2 - K_{2,a} - K_{2,b} - K_3], \quad K_2 = 8K_{2,b} + 2K_3, \\ \eta &= 8K_{2,b} + 2K_{1,b} + 2K_5 \bar{n}_{\text{ex}}, \quad \gamma_2 = K_4 + i\delta_2 = \gamma_c + i\gamma_d. \end{aligned}$$

In deriving this equation we have assumed it is possible to neglect derivatives higher than second order. As noted in Ref. 16, Eq. (2.22) is not a Fokker-Planck equation for a real positive  $P$ . However, there exists, nevertheless, a corresponding Langevin equation:

$$\begin{pmatrix} \dot{\alpha}_1 \\ \dot{\beta}_1 \\ \dot{\alpha}_2 \\ \dot{\beta}_2 \end{pmatrix} = \begin{pmatrix} -\gamma_1\alpha_1 - \chi\alpha_1\alpha_1\beta_1 - ig_1\alpha_2 + ig_2(\beta_2\alpha_1^2 + 2\alpha_2\alpha_1\beta_1) \\ -\gamma_1^*\beta_1 - \chi^*\beta_1\alpha_1\beta_1 + ig_1\beta_2 - ig_2(\alpha_2\beta_1^2 + 2\beta_2\alpha_1\beta_1) \\ -\gamma_2\alpha_2 + E - ig_1\alpha_1 + ig_2\alpha_1\alpha_1\beta_1 \\ -\gamma_2^*\beta_2 + E^* + ig_1\beta_1 - ig_2\beta_1\alpha_1\beta_1 \end{pmatrix} + \begin{pmatrix} K_1\alpha_1^2 + 2ig_2\alpha_1\alpha_2 & K_2\alpha_1\beta_1 + \eta & ig_2\alpha_1^2 & 0 \\ K_2\alpha_1\beta_1 + \eta & K_1^*\beta_1^2 - 2ig_2\beta_1\beta_2 & 0 & -ig_2\beta_1^2 \\ ig_1\alpha_1^2 & 0 & 0 & 2K_4\bar{n} \\ 0 & -ig_2\beta_1^2 & 2K_4\bar{n} & 0 \end{pmatrix}^{1/2} \begin{pmatrix} \xi_1(t) \\ \xi_2(t) \\ \xi_3(t) \\ \xi_4(t) \end{pmatrix}, \quad (2.23)$$

where the  $\xi_i(t)$  comprise a Gaussian stochastic process:

$$\langle \xi_i(t)\xi_j(t') \rangle = \delta_{ij}\delta(t-t').$$

### III. STEADY-STATE BEHAVIOR

#### A. Low-density case

The Langevin equation [Eq. (2.23)] corresponds to a low-density system. As a first approximation, we can neglect noise and investigate the steady-state properties of such a system by considering the deterministic equations ( $\beta_1 \rightarrow \alpha_1^*$ ,  $\beta_2 \rightarrow \alpha_2^*$ ):

$$\dot{\alpha}_1 = -\gamma_1\alpha_1 - \chi\alpha_1|\alpha_1|^2 - ig_1\alpha_2 + ig_2(\alpha_2^*\alpha_1^2 + 2\alpha_2|\alpha_1|^2), \quad (3.1a)$$

$$\dot{\alpha}_1^* = -\gamma_1^*\alpha_1^* - \chi^*\alpha_1^*|\alpha_1|^2 + ig_1\alpha_2^* - ig_2(\alpha_2\alpha_1^{*2} + 2\alpha_2^*|\alpha_1|^2), \quad (3.1b)$$

$$\dot{\alpha}_2 = -\gamma_2\alpha_2 + E - ig_1\alpha_1 + ig_2\alpha_1|\alpha_1|^2, \quad (3.1c)$$

$$\dot{\alpha}_2^* = -\gamma_2^*\alpha_2^* + E^* + ig_1\alpha_1^* - ig_2\alpha_1^*|\alpha_1|^2. \quad (3.1d)$$

Steady-state behavior is found by setting  $\dot{\alpha}_1 = \dot{\alpha}_2 = \dot{\alpha}_1^* = \dot{\alpha}_2^* = 0$  in Eqs. (3.1); algebraic manipulation yields

$$I = n_1 \frac{|\gamma_2^*[(\gamma_b + \chi_b n_1)(g_2 n_1 - g_1) + i(\gamma_a + \chi_a n_1)(3g_2 n_1 - g_1)] + i(g_2 n_1 - g_1)^2(3g_2 n_1 - g_1)|^2}{|(3g_2 n_1 - g_1)(g_2 n_1 - g_1)|^2}, \quad (3.2)$$

$$n_2 = n_1 \left[ \frac{(\gamma_b + \chi_b n_1)^2}{(3g_2 n_1 - g_1)^2} + \frac{(\gamma_a + \chi_a n_1)^2}{(g_2 n_1 - g_1)^2} \right], \quad (3.3)$$

where  $I = |E|_{\text{ss}}^2$ , the steady-state input field intensity,  $n_1 = (|\alpha_1|^2)_{\text{ss}}$ , the steady-state exciton intensity, and  $n_2 = (|\alpha_2|^2)_{\text{ss}}$ , the steady-state output field intensity.

To determine the possibility of observing bistability in this low-exciton-density system, it is necessary to perform a stability analysis. However, the highly nonlinear nature of this four-dimensional system renders such an analytical calculation intractable.

As a first approximation, we consider the very-low-density limit:

$$g_2 n_1 - g_1 \rightarrow -g_1, \quad 3g_2 n_1 - g_1 \rightarrow -g_1$$

[i.e., we set  $g_2 = 0$  in Eqs. (3.1)].

We then find

$$I = \frac{n_1}{g_1^2} (an_1^2 + bn_1 + c), \quad (3.4)$$

$$n_2 = \frac{n_1}{g_1^2} [(\gamma_b + \chi_b n_1)^2 + (\gamma_a + \chi_a n_1)^2], \quad (3.5)$$



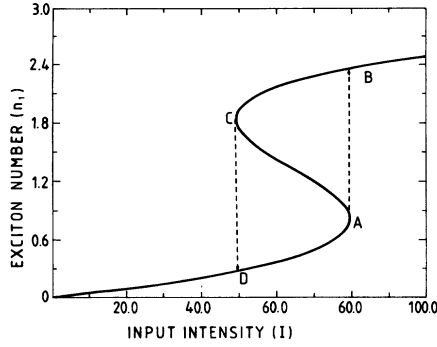


FIG. 1. Variation of exciton number with input intensity in the low-density limit ( $\gamma_a = \gamma_c = \gamma_d = g_1 = 1$ ,  $\gamma_b = -10$ ,  $g_2 = 0.01$ ,  $\chi_a = 1$ ,  $\chi_b = 5$ ).

where

$$\begin{aligned} a &= (\gamma_c \chi_b + \gamma_d \chi_a)^2 + (\gamma_c \chi_a - \gamma_d \chi_b)^2, \\ b &= 2[(\gamma_c \gamma_b + \gamma_d \gamma_a)(\gamma_c \chi_b + \gamma_d \chi_a) \\ &\quad + (g_1^2 + \gamma_c \gamma_a - \gamma_d \gamma_b)(\gamma_c \chi_a - \gamma_d \chi_b)], \\ c &= (\gamma_c \gamma_b + \gamma_d \gamma_a)^2 + (g_1^2 + \gamma_c \gamma_a - \gamma_d \gamma_b)^2. \end{aligned}$$

Linearized stability analysis (Appendix C) shows that such a system will exhibit bistability in exciton number ( $n_1$ ), dependent on input intensity ( $I$ ). As output intensity depends parametrically on  $I$  through  $n_1$ , we expect a corresponding bistability will be observed in output intensity  $n_2$ , dependent on input intensity  $I$ .

The conditions for bistability are determined by requiring the equation  $\partial I / \partial n_1 = 0$  to have real positive solutions. This implies

$$2b < 0, \quad 4b^2 - 12ac > 0$$

for bistability to occur. This will obviously be satisfied for a large range of parameters. We note that the bistability depends solely on exciton-exciton interaction.

Let us consider the absorptive limit,  $\gamma_b = \gamma_d = 0$  (no detuning). In this case,  $b = 2(g_1^2 + \gamma_c \gamma_a) \gamma_c \chi_a$  which is negative only if  $\gamma_a$  is negative, as all other

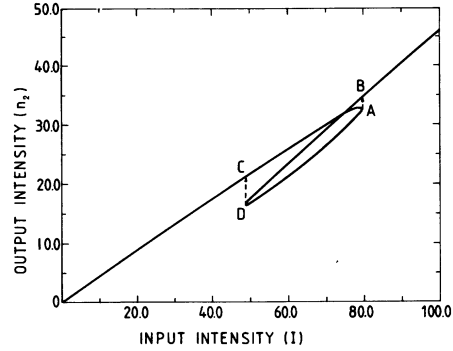


FIG. 2. Variation of output intensity with input intensity in the low-density limit. (Data as given for Fig. 1.)

parameters are positive. This requires either very large values of the thermal occupation numbers of the reservoir modes, or strong exciton-exciton interactions. Thus purely absorptive bistability is possible when exciton-exciton collisions become dominant.

If we now include a nonzero value of  $g_2$  into the system, we do not expect the bistable behavior to change significantly—as long as  $g_2 \ll 1$  and  $n_1$  remains relatively small.

Figures 1 and 2 show plots of  $n_1$  vs  $I$  and  $n_2$  vs  $I$  for small  $g_2$ . (The values of  $n_1$  at the points A, B, C, D in Fig. 1 correspond to the values of  $n_1$  at the points A, B, C, D in Fig. 2.) Dotted arrows indicate expected bistability. That is, from Appendix C we find that the middle branch of Fig. 1 will be unstable as  $\partial I / \partial n_1 < 0$ , and the top and bottom branches will be stable, as  $\partial I / \partial n_1 > 0$ . Bistable transitions occur at the points A and C on Fig. 1 as  $\partial I / \partial n_1 = 0$  at these points. (A similar argument can be applied to the behavior indicated in Fig. 2.)

We see then that bistability is possible in a low-exciton-density system. However, in the experiments involving GaAs, exciton densities are too high to justify the neglect of higher-order terms in the expansion, Eq. (2.9).

Thus we need to consider the high-density case separately.

## B. High-density case

The explicit form of the interaction Hamiltonian obtained from Eq. (2.5) and using the bosonization transformation (2.9) is

$$H_{\text{int}} = \hbar g \hat{a} (\epsilon_1 C^\dagger - \epsilon_2 C^\dagger C^\dagger C) (1 + \epsilon_3 C^\dagger C)^{-1/2} + \text{H.c.}, \quad (3.6)$$

where

$$\epsilon_1 = \frac{1}{\sqrt{v}} \sum_{p_1} f(\alpha p_1 + \beta(p_1 - k)), \quad \epsilon_2 = \frac{1}{v\sqrt{v}} \sum_{p_1} |f(\alpha p_1 + \beta(p_1 - k))|^2 f^*(\alpha(p_1 + \beta(p_1 - k))),$$

$$\epsilon_3 = \frac{1}{v} \sum_{p_1} |f(\alpha p_1 + \beta(p_1 - k))|^2.$$

We now set

$$C = A + \lambda, \quad C^\dagger = A^\dagger + \lambda^*, \quad (3.7)$$

where  $A$  is a boson operator, with  $\langle A \rangle = 0$ , and  $\lambda$  is constant defined such that  $\lambda = \langle c \rangle$ . Thus the large density limit corresponds to large  $\lambda$ , and we can expand perturbatively about the deterministic solution.

Substituting Eq. (3.7) in Eq. (3.6), we expand the denominator to first order and keep terms of order  $\lambda^2$  and  $\lambda$  only:

$$H_{\text{int}} = -\frac{\hbar}{2} g \hat{\epsilon}_2 (\epsilon_3 |\lambda|^2)^{-1/2} \times \lambda^* [(3A^\dagger + 2\lambda^*)\lambda + A\lambda^*] + \text{H.c.} \quad (3.8)$$

This Hamiltonian contributes the following deterministic terms ( $\langle \rangle$  denotes average value):

$$\langle \dot{\hat{a}} \rangle = i\kappa \langle (|\lambda|^2)^{-1/2} \lambda [(3A + 2\lambda)\lambda^* + A^\dagger \lambda] \rangle, \quad (3.9)$$

$$\langle \dot{\hat{C}} \rangle = i\kappa \langle 3\hat{a} (|\lambda|^2)^{-1/2} |\lambda|^2 + \hat{a}^\dagger (|\lambda|^2)^{-1/2} \lambda^2 \rangle, \quad (3.10)$$

where  $\kappa = g\epsilon_2\epsilon_3^{-1/2}/2$ .

Utilizing the definitions  $\langle A \rangle = 0$ ,  $\langle C \rangle = \lambda = \alpha$ , and  $\langle \hat{a} \rangle = \alpha_2$ , we combine Eqs. (3.9) and (3.10) with the deterministic equations corresponding to the rest of the Hamiltonian [Eqs. (3.1) with  $g_1 = g_2 = 0$ ] to find

$$\dot{\alpha}_1 = -\gamma_1 \alpha_1 - \chi \alpha_1 |\alpha_1|^2 + i\kappa (|\alpha_1|^2)^{-1/2} (3\alpha_2 |\alpha_1|^2 + \alpha_2^* \alpha_1^2), \quad (3.11)$$

$$\dot{\alpha}_2 = -\gamma_2 \alpha_2 + E + 2i\kappa \alpha_1 (|\alpha_1|^2)^{-1/2}, \quad (3.12)$$

and complex conjugate equations. These can be solved in the steady state to yield:

$$I = \frac{(a_1 + n_1 a_2)^2 + (a_3 + a_4 n_1)^2}{4\kappa^2}, \quad (3.13)$$

$$n_2 = \frac{1}{4\kappa^2} \left[ \frac{(\gamma_b + \chi_b n_1)^2}{4} + (\gamma_a + \chi_a n_1)^2 \right], \quad (3.14)$$

where

$$a_1 = \frac{\gamma_c \gamma_b}{2} + \gamma_a \gamma_a, \quad a_2 = \frac{\gamma_c \chi_b}{2} + \gamma_a \chi_a$$

$$a_3 = \gamma_c \gamma_a - \frac{\gamma_d \gamma_b}{2}, \quad a_4 = \gamma_c \chi_a - \frac{\gamma_d \chi_b}{2} + 4\kappa^2.$$

### C. Steady-state curves

Figure 3 shows the variation of exciton number  $n_1$  with input intensity  $I$ . The graph is a combination of two curves; the upper part representing the high-density case [Eq. (3.13)], and the lower part describing the low-density case [Eq. (3.2)]. It is necessary to include both theories, as neither is applicable over the whole range of the exciton number  $n_1$ .

In these two distinct regions, the respective theories accurately predict the system's behavior. However, in the intermediate region (indicated by broken lines) we are unsure of the applicability of either theory. We expect the actual behavior of the system to follow Fig. 3, except in the intermediate region, where we assume a smooth transition between high- and low-density behavior will occur.

We expect that the point G in Fig. 3 will correspond to a point of critical instability and a bistable transition will occur as indicated.

As we are unsure of the precise behavior of the system in the region around the point E in Fig. 3, the stability analysis necessary to determine the actual nature of the hysteresis was not performed. Such investigations are proceeding. Nevertheless, we do expect a transition to occur in the neighborhood of the point E, and that bistability will arise as indicated in Fig. 3.

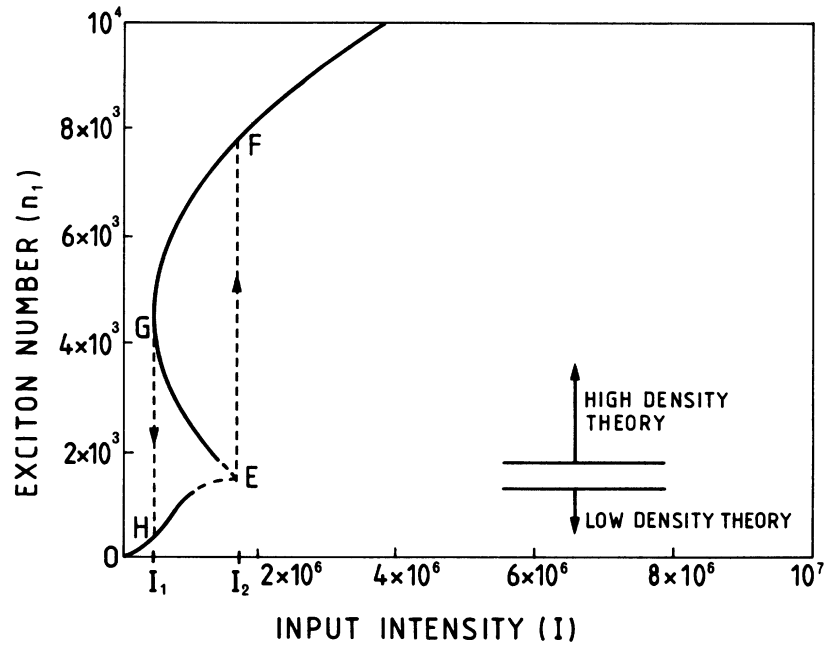


FIG. 3. Exciton number vs input intensity, including the effect of high density of excitons. Broken lines indicate regions in which the theories break down ( $\gamma_a=0.5$ ,  $\gamma_b=-5$ ,  $\gamma_c=1$ ,  $\gamma_d=-5$ ,  $\kappa=0.0037$ ,  $g_1=34$ ,  $g_2=0.0073$ ,  $\chi_a=10^{-4}$ ,  $\chi_b=10^{-3}$ ).

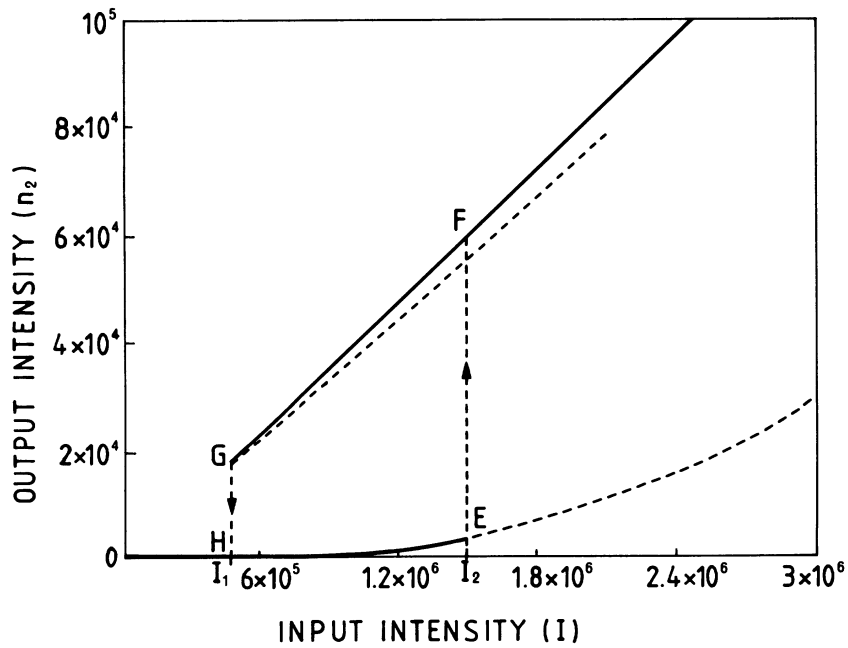


FIG. 4. Output intensity vs input intensity including effects of high exciton density. Broken lines correspond to regions in which the theories break down. (Data as given for Fig. 3.)

Figure 4 shows the corresponding variation of output intensity  $n_2$  with input intensity. (The values of  $n_1$  at the points E, F, G, H in Fig. 3 correspond to the values of  $n_1$  at the points E, F, G, H in Fig. 4.) The upper and lower curves were obtained from the high- and low-density theories, respectively. Again, broken lines indicate regions in which neither theory is valid. As  $I$  is increased, exciton number  $n_1$  varies as in Fig. 3, until at  $I_2$  a transition occurs, indicated by  $\bar{E}\bar{F}$ . We thus expect similar unstable behavior in output intensity  $n_2$  at  $I_2$ : this is indicated by the arrow  $\bar{E}\bar{F}$  in Fig. 4. Similarly, the transition  $\bar{G}\bar{H}$  at  $I_2$  occurs in Fig. 4. Again, the possibility of observing bistability required  $\partial I/\partial n_1 = 0$  to have real, positive solutions. This results in the condition

$$-(a_1 a_2 + a_3 a_4) > 0.$$

In contrast to the low-density case, a nonzero  $\chi$  is no longer necessary for bistability. However, absorptive bistability still requires  $\gamma_a < 0$ , i.e., dominant exciton-exciton interaction.

Thus, bistability and hysteresis in output intensity dependent on input intensity is also displayed. We note that behavior follows the experimental curves of Gibbs *et al.*,<sup>9</sup> showing output intensity vs input intensity for GaAs.

#### IV. DISCUSSION

We have presented a theory of light-semiconductor interaction which adequately includes effects such as exciton-exciton interactions and radiative damping. We were able to model the system in the two limiting cases of low and high exciton densities. Steady-state analysis showed the system exhibits bistability in both exciton number and output intensity, dependent on input intensity. Recent experiments concerned only the latter bistability.

At low intensities, dispersive and absorptive bistability were shown to depend critically on exciton-exciton interaction. Thus, such interactions provide the nonlinearity necessary to produce an intensity dependent refractive index. We also see that these exciton-exciton interactions generate the cooperative effects necessary for bistability.

At high densities, in the dispersive limit, bistability was shown to occur without the presence of exciton-exciton interactions. The nonlinearity in the system causing bistability was due to interaction between light field and medium. We see from the expression for  $H_{\text{int}}$  that this involves the in-

teraction of a photon with two or three excitons—producing cooperativity in the system. This agrees in essence with the suggested bistability mechanism proposed by Gibbs *et al.*:<sup>9</sup> light absorbed just below the exciton resonance frequency produces carriers, thus changing the absorptivity and polarization of the medium, leading to an intensity dependent refractive index.

Purely absorptive bistability is also predicted at high densities and again is seen to require strong exciton-exciton interactions. So far, only dispersive bistability has been experimentally observed.

At present, excitonic bistability of output intensity dependent on input intensity has been observed in GaAs only in the high-density limit. Our work, however, shows that bistability can occur in other materials having lower exciton density.

Finally, we point out that, although our formulation includes all fluctuations, the results presented are only for the limit of small fluctuations. The evaluation of the effects of fluctuation terms on our results is in progress.

#### APPENDIX A: BOSONIZATION TRANSFORMATIONS

This appendix is a summary of the principles and techniques of bosonization transformations in many-fermion systems. The detailed treatment of systems of identical fermions upon which this appendix is based is given by Janssen *et al.*<sup>13</sup> The method itself in this form was first set out by Marumori *et al.*<sup>12</sup> The major difference in our treatment arises from our generalization of the method to deal with electron-hole systems, i.e., systems of two different kinds. The principal formulas are unaltered, but there are changes in technical details. The results can all be proved using methods almost identical to those used by Janssen *et al.*

The operator

$$U = |0\rangle\langle 0| \sum_{n=0}^{\infty} \frac{1}{n!} \frac{1}{\sqrt{n!}} \left[ \sum_{\alpha\beta} b_{\alpha\beta}^\dagger d_{\beta} a_{\alpha} \right]^n |0\rangle\langle 0| \quad (\text{A.1})$$

maps directly from a fermion space to a suitably antisymmetrized boson subspace. That is, we assume the semiconductor system, described in terms of electron and hole operators, is characterized by the anti-symmetrized basis vectors:

$$|m\rangle = a_{\alpha_1}^\dagger d_{\beta_1}^\dagger \cdots a_{\alpha_n}^\dagger d_{\beta_n}^\dagger |0\rangle, \quad (\text{A2})$$

where  $|0\rangle$  is the fermion ground state and  $a_{\alpha_i}$  and  $d_{\beta_i}$  are electron and hole fermion operators, respectively, for the single-particle states  $\alpha_i, \beta_i$ .

The operator  $U$  [Eq. (A1)] maps the state (A2) to a correctly antisymmetrized boson state,

$$|m\rangle = \frac{1}{\sqrt{n!}} \sum_p (-1)^P P b_{\alpha_1 \beta_1}^\dagger \cdots b_{\alpha_n \beta_n}^\dagger |0\rangle, \quad (\text{A3})$$

where  $|0\rangle$  is the boson ground state and  $b_{\alpha\beta}$  is a boson operator;  $P$  is a permutation operator, generating all permutations of indices  $(\alpha_1, \dots, \alpha_n)$  with the indices  $(\beta_1, \dots, \beta_n)$  fixed (or vice versa).

The set of boson states given by Eq. (A3) is in one-to-one correspondence with the fermion states described by Eq. (A2), and the operator  $U$  maps as

$$\hat{P} = \sum_m |m\rangle \langle m| = \sum_{n=0}^{\infty} \sum_{\alpha_1 \cdots \alpha_n} \sum_{\beta_1 \cdots \beta_n} \frac{1}{(n!)^3} B_{\alpha_1 \beta_1}^\dagger \cdots B_{\alpha_n \beta_n}^\dagger |0\rangle \langle 0| B_{\alpha_n \beta_n} \cdots B_{\alpha_1 \beta_1}. \quad (\text{A8})$$

From Eqs. (A4) and (A5) we see that the transformation preserves normalization of states:

$$\begin{aligned} \langle m' | U^\dagger U | m \rangle &= \langle 0 | \langle m' | U^\dagger U | m \rangle | 0 \rangle \\ &= \langle 0 | \langle m' | m \rangle | 0 \rangle = \delta_{mm'} \end{aligned} \quad (\text{A9})$$

and

$$\begin{aligned} \langle m' | U U^\dagger | m \rangle &= \langle 0 | \langle m' | U U^\dagger | m \rangle | 0 \rangle \\ &= \langle 0 | \langle m' | m \rangle | 0 \rangle = \delta_{mm'} \end{aligned}$$

Equations (A9) indicate that  $U^\dagger U$  behaves as unity in the fermion space, and  $U U^\dagger$  as unity in the boson subspace. Also, from Eqs. (A1) and (A8) one may easily show

$$U U^\dagger = \hat{P}. \quad (\text{A10})$$

Thus the projection operator acts like unity in the boson subspace.

The matrix element of an arbitrary operator  $T$ , between fermion states is thus given by

$$\begin{aligned} \langle m | T | m' \rangle &= \langle m | U^\dagger T U | m' \rangle \\ &= \langle m | \mathcal{T} | m' \rangle, \end{aligned} \quad (\text{A11})$$

where  $\mathcal{T} = U T U^\dagger$  is the boson image of  $T$ . Thus the matrix element of any operator is unchanged under the action of the operator  $U$ .

follows:

$$U |m\rangle |0\rangle = |m\rangle |0\rangle, \quad (\text{A4})$$

$$U^\dagger |m\rangle |0\rangle = |m\rangle |0\rangle. \quad (\text{A5})$$

By introducing the operator

$$B_{\alpha\beta}^\dagger = b_{\alpha\beta}^\dagger - \sum_{\gamma\delta} b_{\alpha\gamma}^\dagger b_{\delta\beta}^\dagger b_{\delta\gamma}, \quad (\text{A6})$$

we can write Eq. (A3) in the alternative form:

$$|m\rangle = \frac{1}{\sqrt{n!}} B_{\alpha_1 \beta_1}^\dagger \cdots B_{\alpha_n \beta_n}^\dagger |0\rangle, \quad (\text{A7})$$

i.e., the  $B_{\alpha_i \beta_i}$  generate the desired permutations when acting on the boson ground state (for further discussion, see Ref. 26).

We also define the projection operator  $\hat{P}$  which projects out the nonantisymmetrized components from each boson state,

Pairs of fermion operators are transformed as follows:

$$U a_\mu^\dagger a_\nu U^\dagger = \sum_\rho b_{\mu\rho}^\dagger b_{\nu\rho} \hat{P}, \quad (\text{A12})$$

$$U d_\mu^\dagger d_\nu U^\dagger = \sum_{\rho'} b_{\rho'\mu}^\dagger b_{\rho'\nu} \hat{P}, \quad (\text{A13})$$

$$\begin{aligned} U a_\mu^\dagger d_\nu^\dagger U^\dagger &= B_{\mu\nu}^\dagger \frac{1}{(1+\hat{N})^{1/2}} \hat{P} \\ &= \hat{P} b_{\mu\nu}^\dagger (1+\hat{N})^{1/2}, \end{aligned} \quad (\text{A14})$$

where  $\rho'$  and  $\rho$  indicate summation over electron and hole states, respectively, and

$$\hat{N} = \sum_{\alpha\beta} b_{\alpha\beta}^\dagger b_{\alpha\beta}. \quad (\text{A15})$$

We note that the transformation of the electron-hole pair, Eq. (A14), involves the function

$$(1+\hat{N})^{1/2}.$$

This expression is expanded using the formula,

$$f(\hat{N}) = \sum_{k=0}^{\infty} : \hat{N}^k : \sum_{r=0}^k \frac{f(r) (-1)^{k-r}}{r!(k-r)!}, \quad (\text{A16})$$

where  $::$  denotes the normally ordered product. Equation (A16) is proved in Appendix B.

Transformations of fermion pairs such as

$$Ua^\dagger a^\dagger U^\dagger, Ua^\dagger dU^\dagger$$

are not required in our problem, since we can use the fermion anticommutation relations to relate any term to the expressions  $a_\mu^\dagger a_\nu$ ,  $d_\mu^\dagger d_\nu$ ,  $a_\mu^\dagger d_\nu^\dagger$ , or  $a_\mu d_\nu$ . This occurs because the Hamiltonians used can only produce electrons and holes in pairs.

We note that the transformation  $U$  is in essence a modification of the transformation developed by Marumori *et al.*,<sup>12</sup>

$$U_M = |0\rangle\langle 0| \sum_{n=0}^{\infty} \frac{1}{(2n)!!} \frac{1}{\sqrt{(2n-1)!!}} \times \left[ \sum_{\alpha\beta} b_{\alpha\beta}^\dagger a_\beta a_\alpha \right]^n |0\rangle\langle 0|. \quad (\text{A17})$$

The operator  $U_M$  was used to transform electron-electron pairs and thus is not directly applicable to our semiconductor system.

To bosonize a similar excitonic system, Hanamura<sup>11</sup> used a transformation due to Usui,<sup>23</sup>

$$U_1 = |0\rangle\langle 0| \exp \left[ \sum_{\alpha\beta} b_{\alpha\beta}^\dagger d_\beta a_\alpha \right] |0\rangle\langle 0|. \quad (\text{A18})$$

However, we did not use the transformation

$$\exp(\lambda \sum_i b_i^\dagger b_i) \phi(\vec{\alpha}) = \exp(\lambda \vec{\alpha} \cdot \nabla_{\vec{\alpha}}) \phi(\vec{\alpha}) = \phi(e^\lambda \vec{\alpha}) = \phi[\vec{\alpha} + (e^\lambda - 1)\vec{\alpha}] \quad (\text{B5})$$

$$= \sum_{k=0}^{\infty} \frac{(e^\lambda - 1)^k}{k!} \sum_{\{\vec{r}\}} \left[ \frac{(\alpha_1)^{r_1} (\alpha_2)^{r_2} \cdots (\partial/\partial \alpha_1)^{r_1} (\partial/\partial \alpha_2)^{r_2} \cdots}{r_1! r_2! r_3! \cdots} \delta \left[ \sum r_i, k \right] k! \phi(\vec{\alpha}) \right] \quad (\text{B6})$$

and reverting to the notation  $b_i, b_i^\dagger$ ,

$$\exp(\lambda \hat{N}) = \sum_{k=0}^{\infty} \frac{(e^\lambda - 1)^k}{k!} : \hat{N}^k : \quad (\text{B7})$$

$$= \sum_{k=0}^{\infty} : \hat{N}^k : \sum_{r=0}^k \frac{e^{\lambda r} (-1)^{k-r}}{r!(k-r)!}. \quad (\text{B8})$$

We can write most functions  $f(z)$  in the form

$$f(z) = \int d\lambda e^{\lambda z} F(\lambda) \quad (\text{B9})$$

for some  $F(\lambda)$  and some contour in the complex

described by operator  $U_1$  as it does not preserve normalization of states and there is not a one-to-one correspondence between fermion and boson states.

## APPENDIX B: PROOF OF EQ. (A16)

We wish to show that if  $b_i$  ( $i=1, \dots, n$ ) are independent boson destruction operators, satisfying

$$[b_i, b_j^\dagger] = \delta_{ij} \quad (\text{B1})$$

with

$$\hat{N} = \sum_i b_i^\dagger b_i \quad (\text{B2})$$

and if  $f(z)$  is expressible as a power series in  $z$ , then

$$f(\hat{N}) = \sum_{k=0}^{\infty} : N^k : \sum_{r=0}^k \frac{f(r) (-1)^{k-r}}{r!(k-r)!}, \quad (\text{B3})$$

where  $::$  indicates the normally ordered product.

*Proof.* Represent the creation and destruction operators by

$$b_i \rightarrow \frac{\partial}{\partial \alpha_i}, \quad b_i^\dagger \rightarrow \alpha_i, \quad (\text{B4})$$

and note that for any function  $\phi(\vec{\alpha})$ ,

plane. Doing so we deduce Eq. (B3).

## APPENDIX C

### 1. Stability analysis of the very-low-density system

In such a low-density system ( $g_2=0$ ) we linearize the deterministic Eqs. (3.1) by substituting

$$\alpha_1 = \alpha_0 + \delta\alpha, \quad \alpha_2 = \beta_0 + \delta\beta \quad (\text{C1})$$

(where  $\alpha_0, \beta_0$  are the deterministic means of  $\alpha_1, \alpha_2$ , respectively;  $\delta\alpha, \delta\beta$  are small deviations from

these means), and retaining only constant terms and terms linear in  $\delta\alpha$ ,  $\delta\beta$  in Eqs. (3.1). In this way we find the linearized equations

$$\begin{aligned}\delta\dot{\alpha} &= -(\gamma_1 + 2\chi|\alpha_0|^2)\delta\alpha - \chi\alpha_0^2\delta\alpha^* - ig_1\delta\beta, \\ \delta\dot{\alpha}^* &= -(\gamma_1^* + 2\chi^*|\alpha_0|^2)\delta\alpha^* - \chi^*\alpha_0^{*2}\delta\alpha \\ &\quad + ig_1\delta\beta^*,\end{aligned}\quad (C2)$$

$$\delta\dot{\beta} = -\gamma_2\delta\beta - ig_1\delta\alpha,$$

$$\delta\dot{\beta}^* = -\gamma_2^*\delta\beta^* + ig_1\delta\alpha^*.$$

Equations (C2) yield the dispersion equation

$$m_0\lambda^4 + m_1\lambda^3 + m_2\lambda^2 + m_3\lambda + m_4 = 0, \quad (C3)$$

where

$$\begin{aligned}m_0 &= 1, \quad m_1 = \text{Re}\gamma_2 - \text{Re}(-\gamma_1 - 2\chi|\alpha_0|^2), \\ m_2 &= 2g_1^2 - |\chi|^2(|\alpha_0|^2)^2 + |\gamma_2|^2 + |\gamma_1 + 2\chi|\alpha_0|^2|^2 - \text{Re}(-\gamma_1 - 2\chi|\alpha_0|^2)\text{Re}\gamma_2, \\ m_3 &= g_1^2 m_1 - |\chi|^2(|\alpha_0|^2)^2 \text{Re}\gamma_2 - |\gamma_2|^2 \text{Re}(-\gamma_1 - 2\chi|\alpha_0|^2) + |\gamma_1 + 2\chi|\alpha_0|^2|^2 \text{Re}\gamma_2, \\ m_4 &= \frac{\partial I}{\partial n_1},\end{aligned}\quad (C4)$$

obtained from Eq. (3.4).

## 2. Stability Conditions

The Hurwitz stability criteria for this system are thus,

$$\begin{aligned}(1) \quad & m_0 > 0, \\ (2) \quad & m_1 > 0, \\ (3) \quad & m_1 m_2 - m_3 > 0, \\ (4) \quad & m_1(m_1 m_2 - m_3) - m_1^2 \frac{\partial I}{\partial n_1} > 0,\end{aligned}\quad (C5)$$

and

$$(5) \quad \frac{\partial I}{\partial n_1} \left[ m_3(m_1 m_2 - m_3) - m_1^2 \frac{\partial I}{\partial n_1} \right] > 0.$$

The system exists in a stable state only when conditions (1)–(5) are satisfied. Given that the parameters can be chosen in such a way that all these conditions are met, conditions (4) and (5) imply the system is only stable when

$$\frac{\partial I}{\partial n_1} > 0$$

and critically unstable when

$$\frac{dI}{dn_1} = 0.$$

In fact, in the realistic limit of large excitonic

damping,

$$\begin{aligned}m_0 &= 1 > 0, \\ m_1 &\sim +\text{Re}(\gamma_1) > 0, \\ m_2 &\sim [\text{Re}(\gamma_1)]^2, \\ m_3 &\sim [\text{Re}(\gamma_1)]^2 \text{Re}(\gamma_2), \\ \frac{\partial I}{\partial n_i} &\equiv m_4 \sim \frac{1}{g_1^2}(\gamma_d^2 + \gamma_c^2)[\text{Re}(\gamma_1)]^2,\end{aligned}\quad (C6)$$

and in this limit we also find

$$m_1 m_2 - m_3 \sim [\text{Re}(\gamma_1)]^3 > 0,$$

and

$$m_1(m_1 m_2 - m_3) - m_1^2 \frac{\partial I}{\partial n_1} \sim [\text{Re}(\gamma_1)]^5 > 0,$$

so that conditions (1), (2), (3), and (4) are satisfied.

On a curve of  $I$  vs  $n_1$  the value of  $\partial I/\partial n_1$  is equal to the gradient of the curve and  $\partial I/\partial n_1 = 0$  corresponds to a turning point. Thus, on such a curve, the system will be unstable on branches of negative slope, stable on branches of positive slope and transitions will occur at the turning points. Clearly, this is the same behavior as usually found in bistable systems, e.g. the nonlinear polarizability model.<sup>6</sup>

We thus conclude that this low-density system will exhibit bistability in exciton number  $n_1$ , dependent on input intensity  $I$ .

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