Calculation of the nonlinear dielectric function in semiconductors

I. Abram and A. Maruani
Centre National d'Etudes des Télécommunications, 196 Rue de Paris, 92220 Bagneux, France*
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We have developed a nonperturbational treatment for the calculation of the optical susceptibility of semiconductors in the vicinity of the two-photon biexciton resonance, based on operatorial techniques. The analytical expression of the intensity-dependent dielectric function shows a two-photon polariton effect, which displays bistability when local-field considerations are included in the calculation.

The observation of strong nonlinear-optical effects in semiconductors in the vicinity of the biexciton two-photon resonance, such as up to ten-photon mixing,[1] has raised the problem of the description of the nonlinear optical susceptibility in such systems. Clearly, the near-resonance conditions and the giant oscillator strength of the exciton-biexciton transition render doubtful the validity (especially at high intensities) of the traditional perturbational formulation of nonlinear optics in terms of successive orders of the optical susceptibility. At the same time, the interpretation of biexciton spectra in semiconductors has met with many difficulties which stem essentially from the fact that at present—despite some attempts[2,3]—there is no comprehensive theoretical description for the optical response of biexciton at high light intensities. For example, the broadening of the biexciton absorption line was thought to result from collisions in the exciton-biexciton gas,[4] a hypothesis that has been discounted by recent experiments which argue in favor of power broadening due to the radiative interaction.[5]

In this Communication we outline a semiclassical nonperturbational method for the calculation of the nonlinear optical susceptibility of semiconductors in the vicinity of the two-photon biexciton resonance by treating the electromagnetic field classically and the material excitations quantum mechanically. The method relies on second quantization operator techniques which consist of simple algebraic manipulations of the material excitation operators while at the same time clearly underline the physics involved and the approximations invoked. Damping is treated phenomenologically by simply including at the end of the calculation an imaginary part in the corresponding frequency. We apply this method to the description of experiments involving one laser beam. That is, we calculate the nonlinear dispersion (intensity-dependent refractive index) of the semiconductor for the case of negligible exciton and biexciton damping. Application of the method to multibeam experiments, as, for example, four-wave mixing, is quite straightforward.

The Hamiltonian of a semiconductor interacting with an electromagnetic plane wave with real electric field $E\cos(kr-\omega t)$ near the biexciton two-photon resonance can be written as

$$H = H_0 + H_1 \ , \tag{1a}$$

$$H_0 = \sum_{k'} \omega_0(k') b_{k'}^{\dagger} b_{k'} + \sum_{K} \Omega(K) B_K^{\dagger} B_K \ , \tag{1b}$$

$$H_1 = \hat{\mu}\cdot\vec{E} = -\sqrt{N}\frac{(\mu E)}{2}[\exp(ikr - i\omega t) b_k^{\dagger} + \exp(-ikr + i\omega t) b_k] - \frac{(\phi E)}{2}[\exp(ikr - i\omega t) B_{2k}^{\dagger} b_k + \exp(-ikr + i\omega t) B_{2k} b_k^{\dagger}] \ , \tag{1c}$$

where $b_k^{\dagger}$ and b_k ($B_k^{\dagger}$ and B_k) are the creation and annihilation operators for an exciton (biexciton) of wave vector k and frequency $\omega(\Omega)$. These operators are assumed to follow Bose commutation relations. The exciton transition dipole per unit is denoted by μ, N is the number of unit cells in the crystal, while ϕ represents the exciton-biexciton transition dipole matrix element. By dropping the antiresonant terms in H_1 the rotating wave approximation has been invoked. Because of the selection rules of the radiative interaction only excitons with $k' = k$ and biexcitons with $K = 2k$ need be considered. We may thus suppress for brevity the wave-vector indices and the phase factor e^{ikr} in H_0 and H_1.

The polarization induced on the system by the radiation field is given by

$$P=\left\langle\psi^{+}\left|\frac{\hat{\mu}}{V}\right|\psi^{+}\right\rangle$$
$$=\left\langle\psi^{+}\left|\sqrt{N}\frac{\mu}{V}(b^{\dagger}+b)+\frac{\phi}{V}(B^{\dagger}b+Bb^{\dagger})\right|\psi^{+}\right\rangle$$
$$\equiv P_1+P_2 \ , \tag{2}$$

where V is the crystal volume. For the sake of clarity, the overall polarization is separated into two parts, P_1 the direct contribution of the ground state to exciton transition, and P_2 the exciton to biexciton term. $|\psi^{+}\rangle$ is the perturbed wave function at large positive times, when the radiative interaction is turned-on adiabatically.[6] The adiabatically accessible state is related to the initial (unperturbed) state of the system $|\psi_0\rangle$ by the unitary transformation

$$|\psi^{+}\rangle = U|\psi_0\rangle \tag{3}$$

such that $U^{-1}(H_0+H_1)U$ diagonalizes the Hamiltonian.

The nonlinear polarization is therefore obtained as

$$P=\left\langle\psi_0\left|U^{-1}\frac{\hat{\mu}}{V}U\right|\psi_0\right\rangle \ . \tag{4}$$

The response of the system at the frequency ω is easiest calculated if we place ourselves in a rotating frame at frequency ω for the excitons and 2ω for the biexcitons. In that frame of reference the Hamiltonian is

$$H'=\delta b^{\dagger}b+\Delta B^{\dagger}B-\sqrt{N}\frac{(\mu E)}{2}(b^{\dagger}+b)$$
$$-\frac{(\phi E)}{2}(B^{\dagger}b+Bb^{\dagger}) \ , \tag{5}$$

where $\delta=\omega_0(k)-\omega$ and $\Delta=\Omega(2k)-2\omega$. The unitary transformation which diagonalizes H' can be expressed as three successive partial transformations: two translations, one for the exciton and one for the biexciton coordinates, and a rotation mixing the two types of coordinates. While the details of the calculations will be developed elsewhere,[7] we just give here the expression for the induced polarization calculated according to Eqs. (2) and (4). If the exciton-biexciton system is in its ground state before the radiative interaction the only nonzero terms in the polarization are

$$P_1=\frac{N}{V}\frac{\Delta\mu^2E}{\Delta\delta-(\phi E/2)^2} \ , \tag{6a}$$

$$P_2=\frac{N}{V}\frac{\Delta\mu^2\phi^2E^3}{4[\Delta\delta-(\phi E/2)^2]^2} \ . \tag{6b}$$

If E is identified with the incident electric field the overall nonlinear susceptibility is given by

$$\chi=\frac{P_1+P_2}{E}=\frac{N}{V}\frac{\mu^2\Delta^2\delta}{[\Delta\delta-(\phi E/2)^2]^2} \ . \tag{7}$$

If we disregard all other polarization sources in the system, the dielectric function is given in cgs units by

$$\epsilon=1+4\pi\chi=1+\frac{\omega_{LT}\Delta^2\delta}{[\Delta\delta-(\phi E/2)^2]^2} \ , \tag{8}$$

where $\omega_{LT}=4\pi N\mu^2/V$ is the longitudinal transverse splitting. If relaxation mechanisms are included, the detuning parameters Δ and δ are complex, so that the dielectric function is also complex. It is easily shown that the expansion of ϵ in powers of the electric field yields the classic dispersion relation for the exciton polariton, the standard expression for the third-order susceptibility $\chi^{(3)}$, and so on.

It is useful at this stage to compare Eq. (8) with the expression obtained by März *et al.*[2] through Green's-function techniques. Within the rotating wave approximation, and taking $\epsilon_\infty=1$ and $\omega\simeq\Omega/2$ their expression can be written in our notation as

$$\epsilon\simeq 1+\frac{\omega_{LT}\Delta}{\Delta\delta-n_p(\phi e)^2} \ , \tag{9}$$

where n_p is the polariton density and e the unit electric field. It is readily seen that if the direct contribution of the biexciton transition to the polarization P_2 [Eq. (6)] were neglected in the calculation of the dielectric function, our Eq. (8) would reduce to Eq. (9) for $n_p=E^2/4$.

Figure 1 presents a comparison of the intensity-dependent dispersion relation calculated (a) according to Eq. (8) and (b) according to Eq. (9) with the substitution $n_p\rightarrow E^2/4$. The parameters used are those of CuCl: $\omega_0=3.204$ eV, $\Omega=6.372$ eV, $\omega_{LT}=5.6$ meV, and $\phi=1.65\times10^{-17}$ esu, with no damping. The dispersion relation in the vicinity of the two-

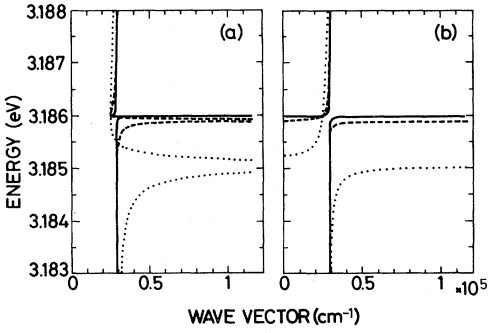

FIG. 1. Polariton dispersion relation near the two-photon biexciton resonance in CuCl (a) using the total polarizability, (b) neglecting the direct biexciton contribution as in Ref. 6. Curves correspond to intensities of 1 MW/cm^2 (——), 10 MW/cm^2 (– – –), and 100 MW/cm^2 (· · · ·).

photon biexciton resonance (equals the two-photon polariton effect) is qualitatively different in the two calculations. The scarcity of experimental data at present,[2] however, does not permit us to validate one calculation over the other through a quantitative fit.

Now, the field seen by the exciton-biexciton system is not equal to the incident field but contains also the contributions of all polarizable entities in the crystal.[8] If the electrons are not delocalized with respect to the holes in the excitons and the biexcitons (a condition which is approximately satisfied in CuCl) the local field is related to the incident field according to

$$E_{\rm loc}=\frac{\epsilon+2}{3}E_{\rm inc} \ . \tag{10}$$

The dielectric function is then given by the implicit equation

$$\epsilon=1+\frac{\omega_{\rm LT}\Delta^2\delta[(\epsilon+2)/3]}{\{\Delta\delta-(\phi E/2)^2[(\epsilon+2)/3]^2\}^2} \ . \tag{11}$$

Figure 2 presents a numerical solution of Eq. (11) in the vicinity of the two-photon biexciton resonance in CuCl as a function of the incident intensity. As seen in Fig. 2 for some frequencies the refractive index of the semiconductor is not a single-valued function of the intensity. That is, the local-field effect introduces a feedback which may produce an intrinsic bistability in the optical response of the semiconductor. Since near the biexciton resonance the light intensities involved in bistable behavior are relatively low, bistability should be readily observable in the reflectivity or transmittance of CuCl. In addition, since no optical cavity is necessary to provide a feedback mechanism this bistability should display a very fast response.

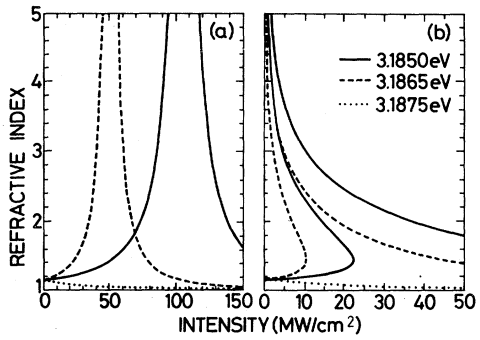

FIG. 2. Calculated refractive index of CuCl as a function of incident intensity, in the vicinity of the two-photon biexciton resonance (3.186 eV): (a) without local-field considerations and (b) including local-field effects.

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