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# Dynamical Stark effect in an exciton–biexciton system

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**Abstract.** A coherently excited exciton–biexciton system under the action of a non-resonant pump field is analysed within the framework of the Hartree–Fock–Bogolyubov theory. The excitation spectra, which consist of optical absorption and gain, are determined. It is pointed out that the optical gain is greatly enhanced through the giant oscillator strength effect of biexcitonic molecules. Numerical examples for CuCl are also presented.

## 1. Introduction

Excitonic optical non-linearities are, in general, due to the anharmonicity of excitons, i.e. the deviation of excitons from non-interacting ideal bosons, and this has its roots in the internal and fermionic degree of freedom of excitons. For example, in the dynamical (AC or optical) Stark effect of excitons observed in GaAs and InGaAs quantum wells, the phase-space filling (PSF) of excitons, which causes the anharmonic exciton–photon interaction, plays an essential role [1–5]. For semiconductor materials with small exciton binding energy such as GaAs, the PSF effect is the predominant factor for non-resonant excitonic optical non-linearities [6]. Conversely, for semiconductor materials in which excitons have large binding energies, the effect of exciton–exciton Coulomb interaction becomes dominant, and when the biexcitonic molecular state is stable, the biexcitonic effect becomes a significant factor. The biexciton formation is also the manifestation of anharmonicity of excitons, and gigantic optical non-linearities due to biexcitonic molecules in CuCl are well known [7].

In this paper, we analyse the coherently excited exciton–biexciton system under the action of a non-resonant pump field within the framework of the Hartree–Fock–Bogolyubov theory. In the following section, we construct the effective boson Hamiltonian for the present system. The linear response of this system to a probe field is calculated and the absorption and gain spectra are determined in section 3. In section 4 we present some numerical results for CuCl. We take $\hbar = 1$ throughout this paper.

## 2. Effective boson Hamiltonian

The system we consider is that of excitons and biexcitons driven by a coherent classical monochromatic pump field $E_p \exp(-i\omega_p t) + \text{CC}$. The exciton and biexciton states, with translational momentum $k$ being zero, are macroscopically populated so that they can

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be regarded as condensates. We take into account the exciton–exciton interaction, the exciton–biexciton interaction, and the anharmonic exciton–photon interaction on the level of the Hartree–Fock–Bogolyubov approximation. Furthermore, we consider the system in a rotating frame so that the time dependence of the pump field is dropped and the single particle energy of an exciton (a biexciton) is measured from $\omega_p$ ($2\omega_p$). The Hamiltonian for this system is, therefore, given by

$$H = H_x^0 + H_m^0 + H_{x-x}^{(int)} + H_{x-m}^{(int)} + H_x^{(rad)} + H_{x-m}^{(rad)} \tag{1}$$

where

$$H_x^0 = \sum_k [\omega_x^0(k) - \omega_p] b_k^\dagger b_k \tag{2a}$$

$$H_m^0 = \sum_k [\omega_m^0(k) - 2\omega_p] c_k^\dagger c_k \tag{2b}$$

$$H_{x-x}^{(int)} = \tfrac{1}{2} V_1 (b_0^\dagger)^2 b_0^2 + 2V_1 b_0^\dagger b_0 \sum_k{}' b_k^\dagger b_k + \tfrac{1}{2} V_1 \sum_k{}' (b_0^2 b_k^\dagger b_{-k}^\dagger + \text{HC}) \tag{2c}$$

$$H_{x-m}^{(int)} = V_2[(b_0^\dagger)^2 c_0 + \text{HC}] + V_2 \sum_k{}' [(2b_0^\dagger b_k^\dagger c_k + c_0 b_k^\dagger b_{-k}^\dagger) + \text{HC}] \tag{2d}$$

$$H_x^{(rad)} = -\mu_x E_p \left[ b_0^\dagger - \gamma (b_0^\dagger)^2 b_0 - \gamma b_0^\dagger \sum_k{}' b_k^\dagger b_k - \tfrac{1}{2}\gamma b_0 \sum_k{}' b_k^\dagger b_{-k}^\dagger \right] - \text{HC} \tag{2e}$$

and

$$H_{x-m}^{(rad)} = -\mu_m E_p \sum_k b_k c_k^\dagger - \text{HC}. \tag{2f}$$

In equations (2), $b_k(c_k)$ is the annihilation operator for an exciton (biexciton) with energy $\omega_x^0(k)[\omega_m^0(k)]$. These operators obey usual boson commutation relations. The prime in equations (2*c*), (2*d*), and (2*e*) indicates that $k = 0$ has to be omitted in the summation over $k$. We have retained only the 1s exciton state and the lowest biexcitonic molecular state in the above Hamiltonian. This approximation is, however, justified as long as the off-resonance energy with respect to the two-photon biexcitonic resonance, $\omega_m^0(k=0) - 2\omega_p$, is of the order of, or less than, the binding energy of the lowest biexcitonic molecular state, $2\omega_x^0(k=0) - \omega_m^0(k=0) = E_B^{(m)}$. Equation (2*c*) describes the repulsive exciton–exciton interaction, and $V_1$ is given by $V_1 = 26\pi E_B^{(x)} a_x^3/(3\Omega)$, where $\Omega$ is the quantization volume of the crystal, and $a_x$ and $E_B^{(x)}$ are, respectively, the effective Bohr radius and the binding energy of the 1s exciton [8]. Equation (2*d*) represents the interaction between excitons and biexcitonic molecules. Equation (2*e*) [(2*f*)] denotes the interaction between excitons (biexcitons) and the pump field; $\mu_x$ and $\mu_m$ are, respectively, the transition dipole moment between the crystalline ground state and the exciton state and that between the exciton state and the biexcitonic molecular state. The second, third, and fourth terms in the square brackets in equation (2*e*) are the anharmonic part of the exciton-pump field interaction with $\gamma = 7\pi a_x^3/(2\Omega)$ [6]. These terms come from the Usui transform [9] on the electron–radiation field interaction:

$$-\mu_{cv} E_p \sum_k a_k^\dagger d_{-k}^\dagger - \text{HC}$$

where $a_k^\dagger(d_k^\dagger)$ is a creation operator for an electron (hole) with wavevector $k$, and $\mu_{cv}$ is the transition dipole moment between conduction and valence bands.

The momentum dependences of potentials $V_1$ and $V_2$ and the coefficient $\gamma$ are neglected in the above Hamiltonian; the exciton–exciton and exciton–biexciton interaction is approximated by the delta-function-like short range interaction. This approximation has, in principle, a deficiency in describing the internal and fermionic nature of excitons correctly, and it would become a worse approximation when the spatial extension of the lowest exciton state becomes larger. The present model can be approximately applied only for the system in which the effective Bohr radius of the lowest exciton is small and the biexcitonic molecular state is well defined, such as in CuCl.

The expectation value of condensate operators $b_0$ and $c_0$ approximately obey the following equations of motion:

$$-\mathrm{i}(\mathrm{d}\langle b_0(t)\rangle/\mathrm{d}t) \simeq \langle [H, b_0(t)]\rangle$$
$$\simeq -\omega_x\langle b_0(t)\rangle - V_1\langle b_0^\dagger(t)b_0(t)\rangle\langle b_0(t)\rangle - 2V_2\langle b_0^\dagger(t)\rangle\langle c_0(t)\rangle$$
$$+ \mu_x E_p[1 - 2\gamma\langle b_0^\dagger(t)b_0(t)\rangle] - 2\gamma\mu_x E_p\langle b_0^2(t)\rangle + \mu_m E_p\langle c_0(t)\rangle \qquad (3a)$$

and

$$-\mathrm{i}(\mathrm{d}\langle c_0(t)\rangle/\mathrm{d}t) \simeq \langle [H, c_0(t)]\rangle \simeq -\omega_m\langle c_0(t)\rangle - V_2\langle b_0^2(t)\rangle + \mu_m E_p\langle b_0(t)\rangle \qquad (3b)$$

where $\omega_x = \omega_x^0(k=0) - \omega_p$ and $\omega_m = \omega_m^0(k=0) - 2\omega_p$. Since the rotating wave frame is adopted and the time evolution of the amplitude of the pump field is neglected, $b_0(t)$ and $c_0(t)$ should be exactly independent of $t$; $\mathrm{d}b_0(t)/\mathrm{d}t = \mathrm{d}c_0(t)/\mathrm{d}t = 0$. Here, we assume that these condensate operators are real $c$-numbers so that we can replace $b_0$ and $c_0$ with their corresponding expectation values; $b_0 \to \langle b_0\rangle = \zeta_{0x}\,(>0)$ and $c_0 \to \langle c_0\rangle = \zeta_{0m}$. These $c$-numbers determine a coherent virtual exciton–biexciton ground state, i.e. the ground state of the correlated exciton–biexciton system dressed with pump photons. In leading order of pump field intensity, $I_p = cE_p^2/(4\pi)$, $\zeta_{0x}$ and $\zeta_{0m}$ are, respectively, given by

$$\zeta_{0x} \simeq \mu_x E_p/\omega_x \qquad (4a)$$

and

$$\zeta_{0m} \simeq (\mu_x\mu_m E_p^2/\omega_x\omega_m)[1 - ((\mu_x/\mu_m)V_2/\omega_x)]. \qquad (4b)$$

Replacing the condensate operators $b_0$ and $c_0$ by the corresponding $c$-numbers thus obtained, the total Hamiltonian is cast into the following form:

$$H_{\mathrm{eff}} = \sum_k{}' [\omega_x(k) + 2(V_1\zeta_{0x} + \gamma\mu_x E_p)\zeta_{0x}]b_k^\dagger b_k + \sum_k{}' \omega_m(k)c_k^\dagger c_k$$
$$+ (1/2)[(V_1\zeta_{0x} + \gamma\mu_x E_p)\zeta_{0x} + 2V_2\zeta_{0m}]\sum_k{}' (b_k^\dagger b_{-k}^\dagger + \mathrm{HC})$$
$$- (\mu_m E_p - 2V_2\zeta_{0x})\sum_k{}' (c_k^\dagger b_k + \mathrm{HC}) \qquad (5)$$

where $\omega_x(k) = \omega_x^0(k) - \omega_p$ and $\omega_m(k) = \omega_m^0(k) - 2\omega_p$, and we have dropped the $c$-number terms in equation (5).

## 3. Linear response to a probe field

The changes of the properties of the exciton–biexciton system under the action of the coherent pump field can be determined by measuring the linear response to a weak

probe field. To distinguish between the polarization induced by the pump field with $k = 0$ and that induced by the probe field, we take the wavevector of the probe field to be finite but close to zero. A weak external probe field with $k \simeq 0$ but $k \neq 0$ induces a polarization:

$$P_k = \mu_x(b_k - \gamma \sum_{k_1,k_2}'' b^\dagger_{k_1+k_2-k} b_{k_1} b_{k_2}) + \mu_m \sum_{k'} c_{k+k'} b^\dagger_{k'} \tag{6}$$

where the double prime on the summation means the restriction due to ordering between $k_1$ and $k_2$ [9]. Since $b_{k=0}$ ($b^\dagger_{k=0}$) and $c_{k=0}$ ($c^\dagger_{k=0}$) are macroscopic $c$-numbers, we approximate $P_k$ as

$$P_k \simeq \mu_x(1 - \gamma\zeta_{0x}^2)b_k + (\mu_m \zeta_{0m} - \gamma\mu_x \zeta_{0x}^2)b^\dagger_{-k} + \mu_m \zeta_{0x} c_k \equiv \bar{P}_k. \tag{7}$$

The Fourier transform of the susceptibility seen by a probe (test) field is given by

$$\chi_t(\omega) = -\lim_{k\to 0} \int_{-\infty}^{\infty} dt \exp[i(\omega + i0_+)t] \langle\langle \bar{P}_k(t); \bar{P}^\dagger_k \rangle\rangle \tag{8}$$

with

$$\langle\langle \bar{P}_k(t); \bar{P}^\dagger_k \rangle\rangle = -i\theta(t)\langle[\bar{P}_k(t), \bar{P}^\dagger_k]\rangle. \tag{9}$$

The frequency $\omega$ is measured from $\omega_p$. In equation (9), $\langle \ldots \rangle$ means the expectation value with respect to the coherent virtual exciton–biexciton ground state. Explicit calculation yields

$$\chi_t(\omega) = -Z(\omega)/D(\omega^2) \tag{10}$$

where

$$\begin{aligned} Z(\omega) = {} & \mu_x^2(1 - \gamma\zeta_{0x}^2)^2(\omega - \omega_m)[(\omega + \bar{\omega}_x)(\omega + \omega_m) - \alpha^2] \\ & + \mu_m^2 \zeta_{0x}^2[(\omega^2 - \bar{\omega}_x^2 + \beta^2)(\omega + \omega_m) - \alpha^2(\omega - \bar{\omega}_x)] \\ & - (\mu_m \zeta_{0m} - \gamma\mu_x \zeta_{0x}^2)^2(\omega + \omega_m)[(\omega - \bar{\omega}_x)(\omega - \omega_m) - \alpha^2] \\ & - 2\mu_x(1 - \gamma\zeta_{0x}^2)\mu_m \zeta_{0x}\alpha[(\omega + \bar{\omega}_x)(\omega + \omega_m) - \alpha^2] \\ & - 2\mu_x(1 - \gamma\zeta_{0x}^2)(\mu_m \zeta_{0m} - \gamma\mu_x \zeta_{0x}^2)\beta(\omega^2 - \omega_m^2) \\ & + 2\mu_m(\mu_m \zeta_{0m} - \gamma\mu_x \zeta_{0x}^2)\zeta_{0x}\alpha\beta(\omega + \omega_m) \end{aligned} \tag{11}$$

and

$$D(\omega^2) = \omega^4 - (\bar{\omega}_x^2 + \omega_m^2 + 2\alpha^2 - \beta^2)\omega^2 + (\bar{\omega}_x\omega_m - \alpha^2)^2 - \beta^2\omega_m^2 \tag{12}$$

with

$$\alpha = \mu_m E_p - 2V_2\zeta_{0x} \tag{13a}$$

$$\beta = (V_1\zeta_{0x} + \gamma\mu_x E_p)\zeta_{0x} + 2V_2\zeta_{0m} \tag{13b}$$

and

$$\bar{\omega}_x = \omega_x + 2(V_1\zeta_{0x} + \gamma\mu_x E_p)\zeta_{0x}. \tag{14}$$

When $E_p = 0$, the solution of $D(\omega^2) = 0$ is, apparently, $\omega^2 = \omega_x^2$ or $\omega_m^2$. Noting this fact, we take $\omega_1$ ($\omega_2$) as the solution which approaches to $\omega_x$ ($\omega_m$) as $E_p$ tends to zero.

These solutions, i.e. the poles of $\chi_t(\omega)$ give the excitation spectra. Of course, $\chi_t(\omega)$ at $E_p = 0$ is reduced to the susceptibility for the direct excitonic transition [8]:

$$\lim_{E_p \to 0} \chi_t(\omega) = \frac{\mu_x^2}{\omega_x - \omega - i0_+}. \tag{15}$$

Now we define the dimensionless reduced oscillator strength:

$$f_i^{\pm} = -\frac{1}{\mu_x^2} \lim_{\omega \to \pm\omega_i} (\omega \mp \omega_i)\chi_t(\omega) \qquad i = 1, 2. \tag{16}$$

By using these quantities, $\chi_t(\omega)$ can be approximately expressed as

$$\chi_t(\omega) \sim \mu_x^2 \sum_{i=1}^{2} \left( \frac{f_i^+}{\omega_i - \omega - i0_+} - \frac{f_i^-}{\omega_i + \omega + i0_+} \right). \tag{17}$$

It is apparent from this form that $\omega = \pm\omega_i$ gives the photon energy for optical absorption (gain) as $f_i^{\pm}$ is positive (negative). In leading order of pump field intensity, it is shown that $f_i^+ > 0$ and $f_i^- < 0$ $(i = 1, 2)$. However, $f_2^-$ is at least in the order of $E_p^6$ and its magnitude is much smaller than that of $f_1^-$ except when $\omega_x \sim \omega_m$. We therefore do not discuss the optical gain given by the solution $\omega = -\omega_2$ in this section. For a weak pump field, we can write $\omega_i$ and $f_i^{\pm}$ $(i = 1, 2)$ in leading order of the pump field intensity as follows:

(i) $\omega_x \neq \omega_m$

$$\omega_1 \simeq \omega_x + 2(V_1 + \gamma\omega_x)\zeta_{0x}^2 + (1/(\omega_x - \omega_m))(\mu_m E_p - 2V_2\zeta_{0x})^2 \tag{18a}$$

$$\omega_2 \simeq \omega_m - (1/(\omega_x - \omega_m))(\mu_m E_p - 2V_2\zeta_{0x})^2 \tag{18b}$$

$$f_1^+ \simeq 1 - 2\gamma\zeta_{0x}^2 - (1/(\omega_x - \omega_m)^2) \times [(2 - (\omega_m/\omega_x))\mu_m E_p - 2V_2\zeta_{0x}]^2 + (\mu_m E_p/\omega_x)^2 \tag{19a}$$

$$f_2^+ \simeq (1/(\omega_x - \omega_m)^2)[(2 - (\omega_m/\omega_x))\mu_m E_p - 2V_2\zeta_{0x}]^2 \tag{19b}$$

and

$$f_1^- \simeq -[((\mu_m/\mu_x) - (V_2/\omega_x))\zeta_{0m} - (V_1 + 3\gamma\omega_x)\zeta_{0x}^2/2\omega_x]^2 \tag{19c}$$

(ii) $\omega_x = \omega_m \equiv \omega_0$

$$\omega_1 \simeq \omega_0 + |\mu_m E_p - 2V_2\zeta_{0x}| \tag{20a}$$

$$\omega_2 \simeq \omega_0 - |\mu_m E_p - 2V_2\zeta_{0x}| \tag{20b}$$

$$f_1^+ \simeq (1/2) + (1/2)[(V_1 + \gamma\omega_0)\zeta_{0x}^2/|\mu_m E_p - 2V_2\zeta_{0x}|] - (\mu_m/\mu_x)\zeta_{0x}\,\mathrm{sign}(\mu_m E_p - 2V_2\zeta_{0x}) \tag{21a}$$

$$f_2^+ \simeq (1/2) - (1/2)[(V_1 + \gamma\omega_0)\zeta_{0x}^2/|\mu_m E_p - 2V_2\zeta_{0x}|] + (\mu_m/\mu_x)\zeta_{0x}\,\mathrm{sign}(\mu_m E_p - 2V_2\zeta_{0x}) \tag{21b}$$

and

$$f_1^- \simeq f_2^- \simeq -(1/2)[((\mu_m/\mu_x) - (V_2/\omega_0))\zeta_{0m} - (V_1 + 3\gamma\omega_0)\zeta_{0x}^2/2\omega_0]^2. \tag{21c}$$

The second term on the right-hand side of equation (18*a*), i.e. the excitonic Stark shift

in the absence of biexcitonic effects, coincides exactly with that obtained by Schmitt-Rink *et al* [10, 11]. It is also the same as the corresponding expression derived by Combescot and Combescot [12, 13] and that derived by Balslev and Hanamura [14], which are, respectively, written as

$$\delta\omega_x^{(CC)} = (\lambda^2/\omega_x)(\alpha_1 + \beta) \tag{22}$$

and

$$\delta\omega_x^{(BH)} = M_0^2 E_p^2((\gamma/\omega_x) + (\lambda_u/\omega_x^2)). \tag{23}$$

Here, $\alpha_1 = 7$, $\beta = (52E_B^{(x)}/3)/\omega_x$, and $\lambda = \mu_{cv}E_p$ for equation (22) and $\gamma = 7$, $\lambda_u = 52E_B^{(x)}/3$, and $M_0 = \mu_{cv}$ for equation (23) so that

$$\delta\omega_x^{(CC)} = \delta\omega_x^{(BH)} = ((\mu_{cv}E_p)^2/\omega_x)(7 + (52/3)(E_B^{(x)}/\omega_x)). \tag{24}$$

This is same as our result for the excitonic Stark shift without the contribution owing to stable biexcitonic molecules.

The term $(\mu_m E_p - 2V_2\zeta_{0x})^2/(\omega_x - \omega_m)$ on the right-hand side of equations (18) is the manifestation of biexcitonic effects and it yields a negative contribution for excitonic shift when $\omega_x < \omega_m$ but $\omega_x \sim \omega_m$; it gives rise to a red shift of an exciton resonance for $\Delta = \omega_x^0(k = 0) - \omega_p > E_B^{(m)}$. The excitonic Stark red shift has been recently observed in CuCl [15].

The solution $\omega = \omega_2$ gives the induced absorption; the transition from a virtually created exciton to a biexcitonic molecule by the absorption of a probe photon. The process creating the virtual exciton state requires one pump photon so that the reduced oscillator strength $f_2^+$ is at least linear in the pump field intensity. When $\omega_x \sim \omega_m$, the exciton and biexciton states are strongly coupled due to the pump field, forming new eigenstates $\omega_1$ and $\omega_2$, in both of which the excitonic component has the same magnitude as the biexcitonic one; only their phases are opposite. Therefore, the oscillator strength of excitonic absorption and that of induced absorption are equalized; $f_1^+ \sim f_2^+ \sim 1/2$ (see equations (21*a*) and (21*b*)). In other words, we have twin absorption peaks with almost the same magnitude for $\omega_x \sim \omega_m$. The energy splitting between these two absorption peaks is $\Delta\omega = 2|\mu_m E_p - 2V_2\zeta_{0x}|$, and this splitting, without the term $V_2\zeta_{0x}$, has been found previously in the effects related to virtual biexciton formation [16] as well as in references [12, 13]. It should be noted that this splitting is proportional to $E_p$ not to the pump field intensity $I_p$ ($\propto E_p^2$).

The solution $\omega = -\omega_1$ gives the optical gain because $f_1^-$ is negative. This solution is rewritten as $\omega^0 + \omega_x^0 = 2\omega_p$, where $\omega_x^0 = \omega_1 + \omega_p$ is the renormalized exciton energy, and $\omega^0 = \omega + \omega_p$. This process is interpreted as follows; a probe photon is absorbed and a renormalized exciton with energy $\omega_x^0$ is created, and at the same time two pump photons are destroyed. As a result, a photon with energy $2\omega_p - \omega_x^0$ is emitted. The coherent virtual exciton–biexciton ground state is accompanied with virtual photon annihilation and creation processes. Such processes contain the annihilation of two pump photons and the creation of one photon with energy $2\omega_p - \omega_1$, which could be stimulated by a probe field, leading to an optical gain. This gain process requires two pump photons so that $f_1^-$ is quadratic in $I_p$. As discussed in the last paragraph, the magnitude of the excitonic component of the eigenstate $\omega_2$ turns out to be the same as that of eigenstate $\omega_1$ when $\omega_x = \omega_m$. As a result, $f_1^-$ and $f_2^-$ are equalized and both $f_1^-$ and $f_2^-$ become a half of the oscillator strength given by equation (19*c*).

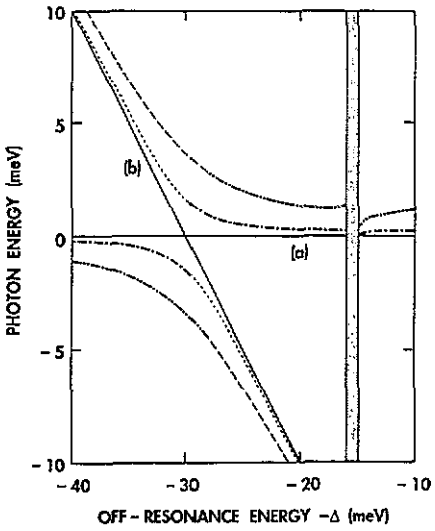


**Figure 1.** Photon energies for optical absorption near the exciton resonance as a function of the off-resonance energy of the pump field. The photon energies for excitonic absorption are denoted by chain curves (pump field intensity $I_p = 1$ MW cm$^{-2}$) and by doubly dotted chain curves ($I_p = 5$ MW cm$^{-2}$), and those for biexcitonic induced absorption are denoted by thin dotted curves ($I_p = 1$ MW cm$^{-2}$) and by thin broken curves ($I_p = 5$ MW cm$^{-2}$). The exciton levels without the pump field (line (a)) are taken to be zero. Line (b) represents the photon energy which would yield the biexcitonic induced absorption for $I_p \to 0$.

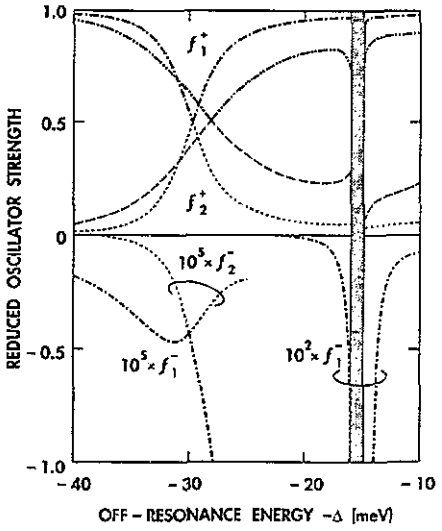


**Figure 2.** Dimensionless reduced oscillator strengths $f_1^{\mp}$ and $f_2^{\mp}$ as a function of the off-resonance energy of the pump field. The reduced oscillator strengths of excitonic absorption ($f_1^{+}$) are denoted by chain curves (pump field intensity $I_p = 1$ MW cm$^{-2}$) and by doubly dotted chain curves ($I_p = 5$ MW cm$^{-2}$), and those of biexcitonic induced absorption ($f_2^{+}$) are denoted by thin dotted curves ($I_p = 1$ MW cm$^{-2}$) and by thin broken curves ($I_p = 5$ MW cm$^{-2}$). The lower part of this figure shows reduced oscillator strengths of optical gain ($f_1^{-}$ and $f_2^{-}$) for $I_p = 1$ MW cm$^{-2}$. The results for $f_2^{-}$ at $\Delta \sim 15$ meV are not shown.

## 4. Numerical examples and discussion

In this section, we present the numerical examples for $\omega_i$ and $f_i^{\mp}$ ($i = 1, 2$). We use the following material parameters: $E_B^{(x)} = 200$ meV, $E_B^{(m)} = 30$ meV, and $a_x = 7$ Å, which are appropriate for CuCl. The exciton transition dipole moment is estimated from the transverse–longitudinal splitting of the exciton, $\Delta_{LT}$ through the relation: $\Delta_{LT} = 4\mu_{cv}^2/(\varepsilon_0 a_x^3)$. We choose $\Delta_{LT} = 5.5$ meV and the static dielectric constant $\varepsilon_0 = 5.1$. The transition dipole moment $\mu_m$ is evaluated, assuming $\mu_m = 8(\pi a_m^3/\Omega)^{1/2}\mu_x$, where $a_m$ is the effective Bohr radius of the biexcitonic molecular state and we choose $a_m = 14$ Å for CuCl. The analytical expression for $V_2$ is, however, not known, so we adopt an approximate relation: $V_2 = 0.5(\mu_x/\mu_m)V_1$, which has been found by the Monte-Carlo calculation [14].

The photon energies giving optical absorption near the exciton resonance, which are measured in the original frame and not in the rotating frame, are shown in figure 1. The exciton level without the pump field (line (a)) is taken to be zero; the energy in this figure shows $\omega_i + \omega_p - E_x^0$ ($i = 1, 2$) where $E_x^0$ denotes the exciton level without the pump field and is also measured in the original frame. Line (b) represents $\omega = -E_B^{(m)} - (-\Delta)$, the photon energy which would yield the biexcitonic induced absorption ($f_2^{+}$) for $I_p \to 0$. The results for $-16$ meV $< -\Delta < -15$ meV are not depicted

because the spectra for $\Delta \sim 15$ meV will be strongly influenced by the real excitation of biexcitonic molecules.

The level repulsion between the upper branch and the lower branch of optical absorption takes place at $\Delta \sim 30$ meV ($\omega_x \sim \omega_m$) and the excitonic absorption is given by the lower (upper) branch for $\Delta > 30$ meV ($\Delta < 30$ meV); the character of upper and lower branches of optical absorption is interchanged at $\Delta \sim 30$ meV. This is also clearly shown in figure 2, where the dimensionless reduced oscillator strengths, as a function of the off-resonance energy of the pump field, are plotted. The branch giving the excitonic absorption (biexcitonic induced absorption) for $\Delta > 30$ meV turns out to give the biexcitonic induced absorption (excitonic absorption) for $\Delta < 30$ meV. The excitonic Stark shift is slightly enhanced near $\Delta = 30$ meV, although the corresponding reduced oscillator strength is reduced to 1/2. It should be noted that the excitonic Stark shift does not diverge even when $\omega_x = \omega_m$, in contradiction to the results of [14].

The optical gain ($f_1^-$) is greatly enhanced for $\Delta \simeq 15$ meV. This is because the population of virtual excitons and that of virtual biexcitons are strongly inverted for $\omega_m \sim 0$ ($\Delta \sim 15$ meV) as is evident from equation (4*b*). The results for the reduced oscillator strength $f_2^-$ at $\Delta \sim 15$ meV are not shown in figure 2 because $f_2^-$ is much smaller than $f_1^-$. However, $f_1^-$ and $f_2^-$ become comparable at $\Delta \sim 30$ meV, where the characters of two branches of optical gain are interchanged.

The oscillator strength of optical gain is much smaller than that of predominant optical absorption. Nevertheless, it is already greatly enhanced by the biexcitonic effects. Without the biexcitonic effects, the excitation spectrum consists of one excitonic absorption and one optical gain. The oscillator strength for this optical gain is calcuated as

$$f_0^- = -\tfrac{1}{4}((V_1/\omega_x) + 3\gamma)^2(\mu_x E_p/\omega_x)^4. \tag{25}$$

The ratio $f_1^-/f_0^-$ for $\omega_x = \omega_m = \omega_0$ (=30 meV) in the case of CuCl is approximately evaluated as $f_1^-/f_0^- \sim 76$. This enhancement, which also depends on the off-resonance energy, originates from the simple fact that the spatial extension of the biexcitonic molecule is larger than that of the electron–hole relative motion in the exciton state. This situation is much the same as the so-called giant two-photon absorption due to biexcitonic molecules observed in CuCl [17]. That is, the giant oscillator strength effect of the biexcitonic molecules [18] leads to the enhanced optical gain.

As mentioned before, the present model cannot be applied for a system in which the lowest exciton state has a large spatial extension and the biexcitonic molecular state is not well defined, such as in GaAs. For such a case, all exciton and biexciton states which include their unbound states, should be incorporated, while the momentum dependence of potentials $V_1$ and $V_2$ and the coefficient $\gamma$ should be included in the present theory.

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