

Note on superradiance of excitonic molecules

Vladimir M. Agranovich

Institute of Spectroscopy, USSR Academy of Sciences, Troitsk, Moscow Region 142092, USSR

and

Shaul Mukamel¹

Department of Chemistry, University of Rochester, Rochester, NY 14627, USA

Received 15 March 1990; accepted for publication 3 May 1990

Communicated by A.A. Maradudin

The radiative decay of excitonic molecules can proceed in two channels: decay to a single exciton and a photon or a direct transition to the ground state with the emission of a photon with the entire biexciton energy (approximately twice the exciton energy). The latter channel is induced by anharmonicity and is normally much weaker. We show that in geometrically restricted systems (surfaces, monolayers, microcrystallites, quantum wells), and at low temperatures, the latter channel is superradiant whereas the former is not. This may provide a new method for the direct observation of biexcitons.

Nonlinear optical spectroscopy of bulk semiconductors and semiconductor microstructures (quantum wells and quantum dots) provides an excellent probe for the structure and the dynamics of highly excited states. Linear spectroscopy probes the dynamics of single quasiparticle (exciton, polariton) and its theoretical description as a harmonic mode (a boson) is well understood. In nonlinear optics we create several quasiparticles which interact. The kinematic interaction is the source for the breakdown of the boson approximation, and together with the dynamical interaction it results in the nonlinear optical response. Consequently nonlinear optics provides a very sensitive means for studying these nonlinear interactions. One of the most important effects in optical nonlinearities of semiconductors is the formation of bound states of two excitons, known as biexcitons or excitonic molecules. Biexciton formation usually results in a new fluorescence line, and in a dramatic enhancement of two photon absorption [1,2]. In addition, virtual biexciton states [3,4] make an important contribution to nonlinear sus-

ceptibilities. The important role of biexcitons in semiconductors has been confirmed by numerous theoretical and experimental studies (for a recent review see ref. [5]). The formation of biexcitons in molecular crystals has been discussed as well [6,7]. However, so far their existence has never been confirmed experimentally. The excitons in molecular crystals are either Frenkel or charge transfer excitons (as opposed to the Wannier excitons in semiconductors). The weaker exciton-exciton interaction or the more efficient exciton-exciton annihilation in this case are possible reasons for the difficulty in biexciton observation.

In this Letter we point out a new possibility for observing biexcitons in two-dimensional microstructures. The main idea is that in two-dimensional systems (interfaces, thin films, quantum wells or molecular monolayers) at low temperature, the biexciton will exhibit a superradiant radiative decay, very similar to what was considered [8-17] and first observed [18] for ordinary excitons. This may allow the observation of fluorescence in which the entire biexciton energy is transferred to a photon $\hbar\omega = E_2(k)$. This channel can therefore compete with

¹ Camille and Henry Dreyfus Teacher/Scholar.

the decay to an exciton and a photon with $\hbar\omega \approx E_2(k) - E_1(k')$ which is normally observed in semiconductors. In molecular crystals fast superradiant radiative decay may compete with exciton-exciton annihilation. For simplicity we discuss below the case of molecular crystals with Frenkel excitons. The molecular crystal Hamiltonian which includes exciton-exciton interactions was derived in ref. [6] (see also ref. [7]).

Adopting a two level model for each molecule (with a ground state 0 and an excited state f, with gas phase electronic transition energy $\Delta\epsilon_f$), the Hamiltonian is given by

$$\hat{H} = \epsilon_0 + \hat{H}_1 + \hat{H}_2 + \hat{H}_3 + \hat{H}_4, \quad (1)$$

where ϵ_0 is the ground state energy of the crystal, and

$$\hat{H}_1 = \sum'_{nm} \langle f0 | \hat{V}_{nm} | 00 \rangle (P_{nf}^\dagger + P_{nf}), \quad (2)$$

$$\begin{aligned} \hat{H}_2 = \sum_n (\Delta\epsilon_f + \mathcal{D}_f) P_{nf}^\dagger P_{nf} \\ + \sum'_{n,m} M_{nm}^f [P_{mf}^\dagger P_{nf} + \frac{1}{2}(P_{nf}^\dagger P_{mf}^\dagger + P_{nf} P_{mf})], \end{aligned} \quad (3)$$

$$\begin{aligned} \hat{H}_3 = \sum'_{n,m} (\langle 0f | \hat{V}_{nm} | ff \rangle - \langle f0 | \hat{V}_{nm} | 00 \rangle) \\ \times (P_{nf}^\dagger + P_{nf}) P_{mf}^\dagger P_{mf}, \end{aligned} \quad (4)$$

$$\hat{H}_4 = \frac{1}{2} \sum_{n,m} \Phi_{nm} P_{nf}^\dagger P_{nf} P_{mf}^\dagger P_{mf}, \quad (5)$$

where

$$\begin{aligned} \Phi_{nm} = \langle ff | \hat{V}_{nm} | ff \rangle + \langle 00 | \hat{V}_{nm} | 00 \rangle \\ - 2 \langle f0 | \hat{V}_{nm} | f0 \rangle, \end{aligned} \quad (6)$$

$$\mathcal{D}_f = \sum_n (\langle 0f | \hat{V}_{nm} | 0f \rangle - \langle 00 | \hat{V}_{nm} | 00 \rangle),$$

$$M_{nm}^f = \langle 0f | \hat{V}_{nm} | f0 \rangle. \quad (7)$$

Here P_{nf}^\dagger (P_{nf}) is the creation (annihilation) operator for an excitation on the n th site. These operators satisfy the Pauli commutation relations. \hat{V}_{nm} represents the intermolecular interaction and its matrix element is defined by

$$\langle \alpha\beta | \hat{V}_{nm} | \gamma\delta \rangle = \int \varphi_n^\alpha \varphi_m^\beta \hat{V} \varphi_n^\gamma \varphi_m^\delta d\tau_n d\tau_m,$$

where φ_n^α represents the state where site n is in state α . The number operator for the excitations is $\hat{N} \equiv \sum_n P_{nf}^\dagger P_{nf}$. If we neglect in eq. (1) the H_1 and H_3

terms, we obtain a zero-order Hamiltonian which conserves the number of excitons (i.e. commutes with \hat{N}):

$$H_0 \equiv \epsilon_0 + \hat{H}_2 + \hat{H}_4. \quad (8)$$

The last term in this Hamiltonian is responsible for the appearance of a bound two-exciton state (biexciton). We suppose below that the exciton-exciton interaction energy (eq. (6)) Φ_{nm} is sufficiently large, so that a bound two-exciton state does exist. We will take into account the mixing effect of states with different numbers of excitons.

The single-particle and two-bound-particles eigenstates and eigenvalues of H_0 with wavevector \mathbf{k} are

$$\psi_1^0(\mathbf{k}) = \sum_n \psi_n(\mathbf{k}) P_{nf}^\dagger | 0 \rangle, \quad E \equiv E_1(\mathbf{k}),$$

$$\psi_2^0(\mathbf{k}) = \sum_{n,m} \psi_{nm}(\mathbf{k}) P_{nf}^\dagger P_{mf}^\dagger | 0 \rangle, \quad E \equiv E_2(\mathbf{k}),$$

where ψ_n and $\psi_{nm} = \psi_{mn}$ are the wave functions of the exciton and the biexciton respectively in the coordinate representation.

Typically the matrix elements of H_1 and H_3 are much smaller than the spacing between the eigenstate of H_0 . (In anthracene, for example, the former are $\sim 1000 \text{ cm}^{-1}$ whereas the latter is 25000 cm^{-1} so that their ratio is $\sim 10^{-2}$). This justifies treating H_1 and H_3 as a small perturbation. We thus define the perturbed two-particle bound state:

$$\psi_2(\mathbf{k}) = \psi_2^0(\mathbf{k}) + \sum_s \frac{\langle \psi_2^0(\mathbf{k}) | \hat{H}_1 + \hat{H}_3 | \psi_s^0(\mathbf{k}) \rangle}{E_2(\mathbf{k}) - E_s(\mathbf{k})}.$$

We expect H_1 to be larger than H_3 . We thus get

$$\begin{aligned} \langle \psi_2^0(\mathbf{k}) | \hat{H}_1 | \psi_1^0(\mathbf{k}) \rangle \\ = \sum_{n_1, m_1, n_2} \psi_{nm}^*(\mathbf{k}) \langle 0 | P_{nf} P_{mf} P_{n_1 f}^\dagger P_{n_2 f}^\dagger | 0 \rangle \\ \times \langle 0f | \hat{V}_{n_1, m_1} | 00 \rangle \psi_{n_2 f}^0(\mathbf{k}) \\ = 2 \sum_{n, m, m_1} \psi_{nm}^*(\mathbf{k}) \langle 0f | \hat{V}_{nm_1} | 00 \rangle \psi_{mf}^0(\mathbf{k}). \end{aligned}$$

We now recall that

$$\psi_{nm}(\mathbf{k}) = \frac{1}{\sqrt{N}} \exp[-\frac{1}{2}i\mathbf{k} \cdot (\mathbf{r}_n + \mathbf{r}_m)] \varphi(|n-m|), \quad (9)$$

where \mathbf{r}_n is the position of site n and $\varphi(n-m)$ is the wave function for the biexciton internal motion,

normalized as $\sum_m |\varphi(|n-m|)|^2 = 1$. Note that for our model $\psi_{nn} = 0$ so that $\varphi(0) = 0$. We thus obtain

$$\begin{aligned} & \sum_m \psi_{nm}^*(\mathbf{k}) \psi_{mf}^0(\mathbf{k}) \\ &= \frac{1}{N} \sum_m \varphi^*(|n-m|) \exp[\frac{1}{2}i\mathbf{k} \cdot (\mathbf{r}_n - \mathbf{r}_m)] \\ &\equiv \frac{1}{N} \Phi^*(\mathbf{k}), \end{aligned}$$

where

$$|\Phi(\mathbf{k})| \approx |\Phi(0)| = \left| \sum_m \varphi(|n-m|) \right| \sim 1.$$

We thus get

$$\begin{aligned} & 2 \sum_{n,m,m_1} \psi_{nm}^*(\mathbf{k}) \psi_{mf}^0(\mathbf{k}) \langle 0f | \hat{V}_{nm_1} | 00 \rangle \\ &= 2\Phi^*(\mathbf{k}) \sum_n \langle 0f | \hat{V}_{nm_1} | 00 \rangle \equiv D, \end{aligned}$$

where D is approximately the sum of energies of interaction of the transition $0 \rightarrow f$ moments (dipole, quadruple, etc.) of molecule n with other molecules of the crystal in the ground state. Therefore, D is of the order of the energy shift of electronic excitation between the gas phase and condensed matter transitions \mathcal{D}_f (eq. (7)). We thus have

$$\psi_2(\mathbf{k}) \approx \psi_2^0(\mathbf{k}) + \frac{D}{E_1(\mathbf{k})} \psi_1^0(\mathbf{k}).$$

It follows from this expression that the transition dipole of the biexciton to the ground state will be related to that of the exciton by

$$\langle \psi_2(\mathbf{k}) | P | 0 \rangle \approx \langle \psi_1(\mathbf{k}) | P | 0 \rangle \frac{D}{E_1(\mathbf{k})}. \quad (10)$$

Since $|D/E_1(\mathbf{k})| \ll 1$, the biexciton state will have a weak borrowed oscillator strength to the ground state due to the mixing with ψ_1^0 (assuming that the single-exciton transition is allowed).

This simple and well known argument explains why in three-dimensional semiconductor crystals biexciton decay proceeds through the channel $E_2 = E_1 + \hbar\omega$, where in the final state we have one exciton and one photon. However, for two-dimensional crystals and for small microcrystallites the situation may be very different. Let us consider two-dimensional crystals (a molecular monolayer). As was shown in ref. [8],

taking into account the retarded interactions strongly changes the two-dimensional exciton spectrum. Instead of a single branch, two polariton branches appear and one of them has a giant radiative width (fig. 1). (Other aspects of this effect were considered in refs. [9–17]). This radiative width Γ depends on the orientation of the transition dipoles and the wavenumber and its typical value is

$$\Gamma \approx \gamma \left(\frac{\lambda}{2\pi a} \right)^2,$$

where γ is the radiative width of the isolated molecule, a is the lattice constant, $\lambda = 2\pi c/\omega_0$, ω_0 is the transition frequency and c is the velocity of light. It should be stressed that this result, and more generally the influence of retardation on the spectrum of elementary excitations, is very different from the case of a three-dimensional crystal, where retarded interactions contribute only to the strong resonance renormalization of the dispersion curve for excitons (polaritons), but radiative decay is totally absent [19–21]. An ultrafast superradiative decay of anthracene films was first observed in recent picosecond measurements conducted by Aaviksoo et al. [18]. The lowest excited state in the surface monolayer in anthracene is blue shifted with respect to the lowest exciton in the bulk by $\sim 204 \text{ cm}^{-1}$. The electronic transition of the first monolayer lies between the bulk value and the isolated molecule value which

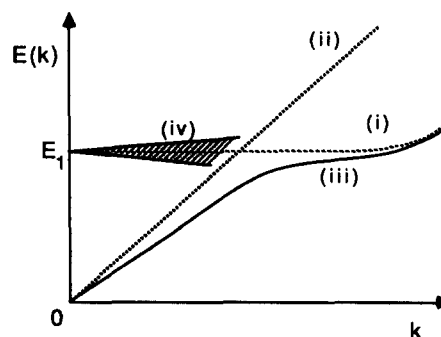


Fig. 1. Dispersion curves for two-dimensional excitons [8] when the molecular transition dipole is perpendicular to the plane. (i) Exciton dispersion without retardation. (ii) The photon dispersion $E = \hbar kc$. (iii) First polariton branch with vanishing radiative width. (iv) Second polariton branch with superradiant width. The finite width of curve (iv) represents the radiative width which grows as the crossing point is approached.

is blue shifted by $\sim 2000 \text{ cm}^{-1}$ with respect to the bulk. The electronically excited state of the surface molecular monolayer is clearly seen in emission at $1.8 k$. The monolayer next to the surface is blue shifted by $\sim 10 \text{ cm}^{-1}$ and the following one by $\sim 2 \text{ cm}^{-1}$. These shifts are now well understood [15]. Consequently, for temperatures low compared with the blue shift, the surface layer acts as an isolated monolayer and is an ideal system for investigations of two-dimensional excitons. Relative quantum yield measurements of the bulk and the surface emission indicate that the decay of the monolayer is purely radiative with a very small contribution of relaxation to the bulk. The picosecond timescales observed in these experiments are a beautiful example of superradiance in two-dimensional excitons. We should bear in mind that the state of biexcitons, like the state of single excitons, is determined by a single wavevector k and not by two wavevectors k_1, k_2 as is the case for two free excitons. This is why all results obtained in refs. [8–13] for superradiance of two-dimensional excitons may be directly used also for two-dimensional biexcitons. This means that biexcitons in a two-dimensional system will show a superradiant decay with radiative width Γ_{BE} :

$$\Gamma_{\text{BE}} = \gamma \left(\frac{D}{E_1(k)} \right)^2 \left(\frac{\lambda}{2\pi a} \right)^2. \quad (11)$$

For anthracene $D/E_1(k) \sim 10^{-2}$, so that without the superradiance $\Gamma_{\text{BE}} \approx 10^{-4}\gamma$. The quantum yield for this channel of direct biexciton emission is therefore very small. The third factor in eq. (11) however represents superradiance. For anthracene $(\lambda/2\pi a)^2 \sim 10^4$. In this case we see compensation of the smallness of $(D/E_1)^2$ by the large values of the superradiance factor $(\lambda/2\pi a)^2$. The radiative decay rate, Γ_{BE} , is therefore in this case comparable in magnitude to the single exciton decay γ . This, of course, need not be always the case. The anharmonicity factor $(D/E_1)^2$ and the superradiant enhancement $(\lambda/2\pi a)^2$ are two independent parameters and their relative magnitude may be very different. Our main point is that in two-dimensional systems there exists a new possibility for observing biexcitons which should be taken into account. In order to establish that, we need to consider also the influence of superradiance and two dimensionality on the other

main channels of biexciton decay. Let us consider the main process which is responsible for the biexciton fluorescence in 3D semiconductors. Suppose that the exciton has a dipole allowed transition to the ground state. The Hamiltonian of interaction with light is then

$$H' = - \sum_n \hat{p}_n \cdot \hat{E}(n),$$

where the electric field operator at position n is

$$\hat{E}(n) = \sum_{qj} (E_{qj} a_{qj} + E_{-qj}^* a_{-qj}^\dagger) \exp(i\mathbf{q}_1 \cdot \mathbf{r}_n),$$

the dipole moment operator of the n th molecule is

$$\hat{p}_n = p_{\text{of}}(P_{nf} + P_{nf}^\dagger).$$

The transition rate is given by Fermi's golden rule:

$$W_{\text{BE}}(\mathbf{k}_1) = \frac{2\pi}{\hbar} \sum_{\mathbf{Q}_1, \mathbf{q}_z, j} |\langle \psi_2^0(\mathbf{k}_1), 0 | \hat{H}' | \psi_1^0(\mathbf{Q}_1), j, \mathbf{k}_1 - \mathbf{Q}_1 + \mathbf{q}_z \rangle|^2 \times \delta(E_2(\mathbf{k}_1) - E_1(\mathbf{Q}_1) - \hbar c \sqrt{(\mathbf{k}_1 - \mathbf{Q}_1)^2 + q_z^2}).$$

Here $|\psi_2^0(\mathbf{k}_1), 0\rangle$ is the biexciton state and the radiation field is in the vacuum state (no photon). $|\psi_1^0(\mathbf{Q}_1), \mathbf{k}_1 - \mathbf{Q}_1 + \mathbf{q}_z\rangle$ is the final state with a $\psi_1^0(\mathbf{Q}_1)$ exciton and a single photon with a wavevector $\mathbf{k}_1 - \mathbf{Q}_1 + \mathbf{q}_z$. The parallel (in plane) wavevector is conserved and \mathbf{q}_z is arbitrary. Taking into account the structure of $\psi_2(\mathbf{k})$ and performing the summation over j , we get

$$\sum_j |\langle \psi_2^0(\mathbf{k}_1), 0 | \hat{H}' | \psi_1^0(\mathbf{Q}_1), \mathbf{k}_1 - \mathbf{Q}_1 + \mathbf{q}_z \rangle|^2 = \frac{8\pi E_1^2 |p_{\text{of}}|^2 |\Phi(0)|^2 (\mathbf{k}_1 - \mathbf{Q}_1)^2}{V \hbar q c q^2},$$

where $q^2 \equiv (\mathbf{k}_1 - \mathbf{Q}_1)^2 + q_z^2$.

If for simplicity we neglect the wavevector dependence of E_2 and E_1 , we get

$$W_{\text{BE}} \approx \frac{32 |\Phi(0)|^2}{3\hbar} |p_{\text{of}}|^2 \left(\frac{\omega_0}{c} \right)^3 \approx \gamma,$$

where $\hbar\omega_0 = E_1(0)$. This channel is therefore not affected by superradiance. As a result of the compensation in eq. (11), both processes $E_2 \rightarrow \hbar\omega$ and $E_2 \rightarrow E_1 + \hbar\omega$ will have approximately the same rate.

Another exciton decay channel in molecular crystals which may compete with the superradiant emission is the bimolecular quenching of excitons. For two excitations separated by a distance R , the bi-

molecular quenching is given by twice the Forster rate $W_F(R) \equiv \gamma(R_F/R)^6$, where R_F is the Forster radius. Consequently, the bimolecular quenching rate of biexcitons is (see also eq. (9))

$$W_{\text{BMQ}} = 2 \sum_m |\varphi(|n-m|)|^2 W_F(|n-m|). \quad (12)$$

Since W_F decreases very rapidly with R , the most important contribution to eq. (12) is when the two particles are separated by the nearest neighbor spacing a . We thus have

$$W_{\text{BMQ}} \approx 2Z|\varphi(a)|^2 W_F(a).$$

Here Z is the number of nearest neighbors in the lattice. Since $\varphi(r)$ vanishes at $r=0$ and for $r \rightarrow \infty$, $|\varphi(r)|^2$ is a nonmonotonic function of r . If the biexciton radius is $R_{\text{BE}} \sim a$, then $|\varphi(a)|^2 \sim 1$. However, when $R_{\text{BE}} \gg a$, $|\varphi(a)|^2$ will depend on the behavior of $\varphi(r)$ for small r . Assuming that $\varphi(r) \sim r^\alpha$ for $r \ll R_{\text{BE}}$, we have $|\varphi(a)|^2 \approx (a/R_{\text{BE}})^{2+\alpha}$. In order to observe the direct biexciton decay we require therefore that the bimolecular quenching rate be smaller than the radiative decay rate of biexcitons, i.e.

$$2Z(R_F/a)^6(a/R_{\text{B}})^{2+\alpha} < \Gamma_{\text{BE}}/\gamma.$$

The present discussion may apply also to the observation of excitonic molecules of triplet excitons. In that case, the dipole transition is forbidden so that both radiative decay mechanisms are much slower. The argument regarding the quantum yield is however expected to hold.

Although the present calculation was made for molecular monolayers, the argument presented here applies to any geometrically restricted microstructure such as J aggregates, and small clusters (smaller than the optical wavelength) [22–25]. In these cases the $(\lambda/2\pi a)^2$ factor should be replaced by the cooperation number N_{eff} which denotes the number of molecules in a coherence volume [26,27]. In a small cluster of radius R , $N_{\text{eff}} \sim (R/a)^3$. For a semiconductor sphere $N_{\text{eff}} \sim (R/a_{\text{B}})^3$ where a_{B} is the exciton Bohr radius. The cooperation number may also depend on fluctuations and relaxation processes such as exciton phonon coupling and may be smaller than the total number of molecules in the cluster.

The support of the Science and Technology Center for Photoinduced Charge Transfer and the National

Science Foundation, the Air Force Office of Scientific Research, and the Petroleum Research Fund, administered by the American Chemical Society, is gratefully acknowledged.

References

- [1] E. Hanamura, *Solid State Commun.* 12 (1973) 951.
- [2] G.M. Gale and A. Myszyzowicz, *Phys. Rev. Lett.* 32 (1979) 727.
- [3] V.M. Agranovich, N.A. Efremov and E.P. Kaminskaya, *Opt. Commun.* 3 (1971) 387; N.A. Efremov and E.P. Kaminskaya, *Fiz. Tverd. Tela* 14 (1972) 1185 [*Sov. Phys. Solid State*].
- [4] A. Maruani, J.L. Oudar, E. Batifol and D.S. Chemla, *Phys. Rev. Lett.* 41 (1978) 1372; A. Maruani and D.S. Chemla, *Phys. Rev.* 23 (1981) 841; Y. Masumoto and S. Shionoya, *J. Phys. Soc. Japan* 49 (1980) 2236; *Solid State Commun.* 38 (1981) 865.
- [5] M. Ueta, M. Kanzaki, K. Kobayashi, Y. Toyozawa and E. Hanamura, *Springer series in solid state sciences*, Vol. 60. *Excitonic process in solids* (Springer, Berlin, 1984).
- [6] V.M. Agranovich, *Theory of excitons* (Nauka, Moscow, 1968) [in Russian].
- [7] V.M. Agranovich and M.D. Galanin, *Electronic excitation energy transfer in condensed matter* (North-Holland, Amsterdam, 1982).
- [8] V.M. Agranovich and O.A. Dubovski, *Pis'ma Zh. Eksp. Teor. Fiz.* 3 (1966) 345 [*JETP Lett.* 3 (1966) 223].
- [9] V.I. Sugakov, *Fiz. Tverd. Tela (Leningrad)* 14 (1973) 1711 [*Sov. Phys. Solid State* 14 (1973) 1473].
- [10] M.R. Philpott, *J. Chem. Phys.* 61 (1974) 5306.
- [11] M.R. Philpott and P.G. Sherman, *Phys. Rev. B* 12 (1975) 5381.
- [12] Y.C. Lee and P.S. Lee, *Phys. Rev. B* 10 (1974) 344.
- [13] K.C. Liu, Y.C. Lee and Y. Shan, *Phys. Rev. B* 11 (1975) 978.
- [14] M. Orrit, C. Aslangul and Ph. Kottis, *Phys. Rev. B* 25 (1982) 726.
- [15] J.M. Turiel, Ph. Kottis and M.R. Philpott, *Adv. Chem. Phys.* 54 (1983) 303.
- [16] V.M. Agranovich, in: *Surface excitations*, eds. V.M. Agranovich and R. Loudon (North-Holland, Amsterdam, 1984) p. 551.
- [17] M. Orrit and Ph. Kottis, *Adv. Chem. Phys.* 74 (1988) 1.
- [18] Ya. Aaviksoo, Ya. Lippmaa and T. Reinot, *Opt. Spektrosk.* 62 (1987) 706 [*Opt. Spectrosc. (USSR)* 62 (1987) 419].
- [19] U. Fano, *Phys. Rev.* 103 (1956) 1202; 118 (1960) 451.
- [20] J.J. Hopfield, *Phys. Rev.* 112 (1958) 1555.
- [21] V.M. Agranovich, *Zh. Eksp. Teor. Fiz.* 37 (1959) 340 [*Sov. Phys. JETP* 10 (1960) 307].
- [22] D.V. Brumbaugh, A.A. Muentner, W. Knox, G. Mourou and B. Wittmershaus, *J. Lumin.* 32 (1984) 783; D. Mobius and H. Kuhn, *J. Appl. Phys.* 64 (1988) 5138;

- V. Sundstrom, T. Gillbro, R.A. Gadonas and A. Piskarskas, *J. Chem. Phys.* 89 (1988) 2754;
S. de Boer and D.A. Wiersma, *Chem. Phys. Lett.* 165 (1990) 45.
- [23] Y.R. Kim, M. Lee, J.R.G. Thorne, R.M. Hochstrasser and J.M. Ziegler, *Chem. Phys. Lett.* 145 (1988) 75.
- [24] J. Feldmann, G. Peter, E.O. Gobel, P. Dawson, K. Moore, C. Foxon and R.J. Elliot, *Phys. Rev. Lett.* 59 (1987) 2337.
- [25] T. Itoh, T. Ikehara and Y. Iwabuchi, *J. Lumin.* (1990), to be published;
M. Bawendi, M.L. Steigerwald and L.E. Brus, *Annu. Rev. Phys. Chem.* 44 (1990), in press.
- [26] J. Grad, G. Hernandez and S. Mukamel, *Phys. Rev. A* 37 (1988) 3838.
- [27] F.C. Spano and S. Mukamel, *J. Chem. Phys.* 91 (1989) 683;
Phys. Rev. A 40 (1989) 5783;
F.C. Spano, J.R. Kuklinski and S. Mukamel, to be published.