Renormalized Frequency Shift of Superradiant Excitons in Thin Semiconductor Films

Yung-Chang Lee,[a] Der-San Chuu, and Wai-Ning Mei[b]
Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan, Republic of China
(Received 27 April 1992)

The radiative frequency shift of an exciton in a thin semiconductor film, like its radiative level width, is shown to be superradiatively enhanced. Unlike the latter, however, a finite frequency shift can only be obtained after proper renormalization for the correlated system. The shift is found to be inversely proportional to the square of the factor $k_0 d$ and proportional to the film thickness T; $k_0 = \frac{E_{\mathbf{q}n}}{\hbar c}$, $E_{\mathbf{q}n}$ being the exciton energy gap and d the lattice constant of the semiconductor. Therefore, the coherent frequency shift can be observed experimentally if one varies the thickness, or the exciton energy gap $E_{\mathbf{q}n}$ by imposing high pressure.

PACS numbers: 71.35.+z, 71.45.–d, 78.65.–s

Recently, much attention has been focused on superradiance of systems with restricted geometries. Historically, the idea of the superradiant state for a system of atoms was introduced by Dicke [1]. The coherent radiation phenomenon for the atomic system was intensively investigated in the late sixties [1–4]. It was found that the radiative correction to the emission frequency of these systems usually involves divergences that have to be removed by renormalization [4, 5]. An adaptation of the general prescription for renormalizing such radiative frequency shift for a many-particle system was proposed by Lee and Lin [5]. Turning to a system of atoms arranged in a three-dimensional lattice with one of the atoms excited initially, Lee and Lee [6] found that the photon would be totally trapped in the system if it were not for the broken symmetry due to lattice imperfections and the finite boundary. Furthermore, when the longitudinal Coulomb interaction among the atoms that is partially responsible for the propagation of the exciton in a solid is taken into account in a three-dimensional bulk crystal, the exciton is found to couple with the photon to form a polariton and thus does not decay radiatively [7].

However, when the exciton is in a thin crystal film [8–16], the crystal symmetry in the direction normal to the film plane is broken in an essential way, rendering the radiative decay of the exciton possible. It was shown [9, 10] that the remaining crystal symmetry parallel to the plane of the film led to phase coherence between the exciton and photon fields everywhere in the film. Consequently, a trade-off between the degrees of freedom of the radiation field and those of the atoms in the crystal film was shown to be responsible for the superradiative enhancement of the exciton decay in the optical region [10]. Recently, many investigations on the radiative linewidth of excitons in quantum wells have been performed [11–17]. The effect of phonons on the cooperative radiative decay of excitons in small aggregates has attracted strong interest [13]. The size-dependent radiative decay time of excitons in microcrystallites [14] and the radiative dynamics crossover from the small thickness, superradiant exciton regime to the bulk crystal, polariton regime were studied extensively [16]. Recently, radiative lifetimes of superradiant decay of excitons in InAs quantum sheets were measured [17]. However, the coherent frequency shift of an exciton in thin films has received little attention so far. For one thing, the radiative frequency shift of an exciton involves ultraviolet divergences arising from virtual photons of large momenta. Furthermore, in calculating the frequency shift of the exciton in a thin film due to coherence effect alone, apparent infrared divergences of the logarithmic type turn up in the limit of zero $\mathbf{q}$, the center-of-mass momentum of the exciton. In this Letter, we show how the radiative frequency shift of the exciton in a semiconductor thin film should be properly renormalized before a meaningful comparison between theoretical results and experimental observations can be made. The renormalized frequency shift, much like the linewidth, will be seen to also be superradiatively enhanced due to coherence effect.

Consider first a Wannier exciton in a very thin film consisting of one layer in the x-y plane. The generalization to the case of a thicker film with larger width T will be made later. We will assume a two-band model for the band structure of the relevant semiconductor, the ramification of which will be discussed later. Suppose that at $t = 0$ the system is in an excitonic $|\,\mathbf{q}, n\rangle$ state, where $\mathbf{q}$ is the crystal momentum in the x-y plane characterizing the motion of the exciton and n is the internal structure quantum number for the exciton with energy $E_{\mathbf{q},n} \equiv \hbar c k_0$. For time $t > 0$, the system evolves in time and generally returns to its ground state by emitting a photon [9]. The time dependence is characterized by a frequency shift and a decay rate. The frequency shift of the exciton can be expressed as [9]

$$\Omega_{\mathbf{q},n} = \mathrm{Re}\sum_k \frac{|\,D_{\mathbf{k},\mathbf{q}n}\,|^2}{\hbar^2(ck_0 - ck - i0^+)} = \Omega_{\mathbf{q},n}(0) + \sum_{l\neq 0} \exp(-i\mathbf{q}\cdot\boldsymbol{l})\Omega_{\mathbf{q}n}(\boldsymbol{l}), \quad (1)$$

where $\Omega_{\mathbf{q}n}(\boldsymbol{l})$ is the energy correction (except for $\hbar$) due to the emission and its subsequent reabsorption of a virtual photon by the $\mathbf{q}n$ exciton which, in the meantime,

has traveled a distance of lattice vector $\boldsymbol{l}$. Naturally a sum over $\boldsymbol{l}$ has to be carried out. Splitting the $\boldsymbol{l}=0$ term from this sum we see that the first term $\Omega_{\mathbf{q}n}(0)$ represents the frequency shift of a lone exciton isolated in space as if the surrounding lattice had been stripped off. It suffers from the ultraviolet divergence just like the Lamb shift of a hydrogen atom and thus needs to be renormalized. The second term in Eq. (1) represents the frequency shift due to the coherence effect and can be seen to be convergent [5]. The radiative decay rate of the exciton $\gamma_{\mathbf{q}n}$ is similarly given by

$$\gamma_{\mathbf{q}n}=\mathrm{Im}\sum_{k}\frac{|\,D_{\mathbf{k},\mathbf{q}n}\,|^2}{\hbar^2(ck_0-ck-i0^+)}$$
$$=\Gamma_{\mathbf{q}n}(0)+\sum_{l\neq 0}\exp(-i\mathbf{q}\cdot\boldsymbol{l})\Gamma_{\mathbf{q}n}(\boldsymbol{l}). \tag{2}$$

In Eqs. (1) and (2),

$$D_{\mathbf{k},\mathbf{q}n}=\left[\frac{2\pi e^2\hbar c}{m^2c^2NkV}\right]^{1/2}\sum_{l}\exp[i(\mathbf{k}-\mathbf{q})\cdot\boldsymbol{l}](\hat{\boldsymbol{\varepsilon}}_{\mathbf{k}\sigma}\cdot\mathbf{X}_n), \tag{3}$$

where the exponential phase factor is due to the propagation of the virtual $\mathbf{k}$ photon and of the $\mathbf{q}n$ exciton over the same distance $\boldsymbol{l}$, and

$$\mathbf{X}_n=\sum_{\boldsymbol{\rho}}F_n^*(\boldsymbol{\rho})\int d^2\tau\, w_c^*(\boldsymbol{\tau}-\boldsymbol{\rho})(-i\hbar)\,\nabla_\tau\, w_v(\boldsymbol{\tau}) \tag{4}$$

is the effective transition dipole matrix element between the electronic Wannier state w_c in the conduction band and the Wannier hole state w_v in the valence band. Its presence signifies the repeated deexcitation and reexcitation of the exciton as it evolves in time, being punctuated by the virtual-photon emissions and readsorptions. Here $F_n(\boldsymbol{\rho})$ is the two-dimensional hydrogenic wave function with $\boldsymbol{\rho}$ being the separation between the electron and hole of the exciton. The vectors $\boldsymbol{l}$ and $\boldsymbol{\rho}$ are in the x-y plane. The second term in Eq. (1) represents the coherence effect since it involves a sum over nonvanishing lattice vectors $\boldsymbol{l}$. If the lattice spacing d is much larger than the characteristic radiation wavelength λ_0, this term becomes insignificant owing to the then wildly fluctuating individual terms in the summand. For any finite d/λ_0, the quantities $\Omega_{\mathbf{q}n}(\boldsymbol{l})$ and $\Gamma_{\mathbf{q}n}(\boldsymbol{l})$ can be obtained as [10]

$$\Omega_{\mathbf{q}n}(\boldsymbol{l})=\sum_{\mathbf{k}_2}J_{\mathbf{q}n}(\mathbf{k}_2)\exp(i\mathbf{k}_2\cdot\boldsymbol{l}) \tag{5}$$

$$J_{\mathbf{q}n}(\mathbf{k}_2)=P\sum_{k_z}\frac{|\,A_{\mathbf{k},\mathbf{q}n}\,|^2}{ck_0-c\sqrt{k_2^2+k_z^2}},\quad \mathbf{k}=\mathbf{k}_2+\hat{\mathbf{z}}k_z. \tag{6}$$

Also,

$$\Gamma_{\mathbf{q}n}(\boldsymbol{l})=\sum_{\mathbf{k}_2}G_{\mathbf{q}n}(\mathbf{k}_2)\exp(i\mathbf{k}_2\cdot\boldsymbol{l}), \tag{7}$$

where

$$G_{\mathbf{q}n}(\mathbf{k}_2)=\sum_{k_z}\pi\,|\,A_{\mathbf{k},\mathbf{q}n}\,|^2\,\delta(ck_0-c\sqrt{k_2^2+k_z^2}). \tag{8}$$

In Eqs. (6) and (8),

$$|\,A_{\mathbf{k},\mathbf{q}n}\,|^2=\frac{2\pi e^2}{m^2c\hbar kV}\,|\,\hat{\boldsymbol{\varepsilon}}_{\mathbf{k}\sigma}\cdot\mathbf{X}_n\,|^2 \tag{9}$$

which is valid in the optical limit $qa_0\ll 1$, a_0 being the size of the exciton. One may note from Eqs. (8) and (9) that $|\,A_{\mathbf{k},\mathbf{q}n}\,|^2$ and hence $J_{\mathbf{q}n}(\mathbf{k}_2)$ and $G_{\mathbf{q}n}(\mathbf{k}_2)$ are actually independent of $\mathbf{q}$ in the optical limit.

As asserted before, $\Omega_{\mathbf{q}n}(0)$ can easily be seen from Eqs. (5), (6), and (9) to diverge linearly due to the upper limit of the sum over $|\,\mathbf{k}\,|$. Renormalization is therefore in order. Since the usual method of renormalization is for one-electron systems, it must be suitably generalized to be applicable to the present case of a many-electron, two-band system. The spirit of mass renormalization is to use in the kinetic-energy term the finite mass of the free, physical (dressed) electron, which presumably consists of a bare electron dressed by a virtual photon cloud. Both the bare mass m_0 of the free electron and the energy correction due to the photon cloud turn out to be infinite, but largely canceling each other, leaving a finite physical electron mass m. When the electron is not free, as in an excitonic state of a semiconductor, the quantum state itself will be the result of the simultaneous action of the kinetic and the potential energy terms, the latter representing the environment the electron is put into. The effect of the removal of the ultraviolet divergence when the electron is free differs, of course, from the effect when the electron is in the environment of a potential. The difference between the two represents the effect of renormalization on the quantum state. Adapting to the present exciton case we have accordingly

$$\Omega_{\mathbf{q}n}^{\mathrm{ren}}=\Omega_{\mathbf{q}n}-\lim_{\substack{k_0\to 0\\ d\to\infty}}\Omega_{\mathbf{q}n}, \tag{10}$$

where the two limiting processes $k_0\to 0$ and $d\to\infty$ reduce the exciton to a free electron. The important advantages of such a renormalization procedure lie first in its being free of the ambiguities that might arise owing to the usually different perturbation schemes used in calculating the energy of the dressed exciton and that of the dressed free electron, and second in the inclusion of any many-body correlation effect in $\Omega_{\mathbf{q}n}$ itself.

It follows from Eqs. (10) and (1) that the renormalized frequency shift is

$$\Omega_{\mathbf{q}n}^{\mathrm{ren}}=\Omega_{\mathbf{q}n}^{\mathrm{ren}}(0)+\Omega_{\mathbf{q}n}^{\mathrm{coh}}, \tag{11}$$

where

$$\Omega_{\mathbf{q}n}^{\mathrm{ren}}(0)=\Omega_{\mathbf{q}n}(0)-\lim_{k_0\to 0}\Omega_{\mathbf{q}n}(0) \tag{12}$$

and

$$\Omega_{\mathbf{q}n}^{\mathrm{coh}}=\sum_{l\neq 0}e^{-i\mathbf{q}\cdot\boldsymbol{l}}\Omega_{\mathbf{q}n}(\boldsymbol{l}) \tag{13}$$

since $\lim_{d\to\infty}\Omega_{\mathbf{q}n}=\Omega_{\mathbf{q}n}(0)$.

As seen from Eq. (12), the renormalization affects only the part $\Omega_{\mathbf{q}n}(0)$, the frequency shift of the lone exciton—a positroniumlike object insulated from correlating with the rest of the crystal lattice. The coherent part $\Omega_{\mathbf{q}n}^{\mathrm{coh}}$ in

Eqs. (11) and (13) is then not touched by renomalization in principle. The separation of $\Omega_{\mathbf{q}n}$ into two terms in Eqs. (1) and (11), trivial as it seems, is essential conceptually to the singling out of the source of the quantum electrodynamical divergence in a correlated system and hence to its removal by renormalization.

On the other hand, it is much more expedient mathematically to carry out the lattice sum over all $\boldsymbol{l}$'s, including $\boldsymbol{l}=0$, as required in calculating the unrenormalized $\Omega_{\mathbf{q}n}$ of Eq. (1). By substituting Eq. (5) into (1) and approximating $J_{\mathbf{q}n}(\mathbf{k}_2)$ by $J_n(\mathbf{k}_2)$ as mentioned before we obtain

$$\Omega_{\mathbf{q}n}=\sum_{\mathbf{k}_2}J_n(\mathbf{k}_2)\sum_{\boldsymbol{l}}\exp[i(\mathbf{k}_2-\mathbf{q})\cdot\boldsymbol{l}]$$
$$=N\sum_{\mathbf{G}}J_n(\mathbf{q}+\mathbf{G}), \tag{14}$$

where $\mathbf{G}$ are the reciprocal lattice vectors in the k_x-k_y plane . We now examine the contribution of the umklapp (U) processes involving all the nonvanishing $\mathbf{G}$'s in Eq. (14), thereby going beyond the usual two-band model. Combining Eqs. (6) and (9) with (14) we find

$$\Omega_{\mathbf{q}n}=N\left[P\sum_{k_z}\sum_{\mathbf{G}}\mid A_{\mathbf{k},n}\mid^2\frac{1}{c(k_0-\sqrt{k_2^2+k_z^2})}\right], \tag{15}$$

where $\mathbf{k}_2\equiv\mathbf{q}+\mathbf{G}$ for abbreviation sake, and $A_{\mathbf{k},\mathbf{q}n}$ is now approximated as $A_{\mathbf{k},n}$ while N is the number of unit cells in the one-layer system. Focusing first on the contributions to $\Omega_{\mathbf{q}n}$ from large $\mid\mathbf{G}\mid\gg q$ and noting $\mid A_{\mathbf{k}n}\mid^2\sim 1/k$ from Eq. (9) we convert the sums into integrals via $\sum_{\mathbf{G}}\rightarrow\int\frac{d^2G}{(2\pi/a)^2}$ and $\sum_{k_z}\rightarrow\int\frac{dk_z}{2\pi/L_z}$ and obtain

$$\Omega_{\mathbf{q}n}\sim\frac{L_zNa^2}{(2\pi)^3}\int_{-\infty}^{\infty}dk_z\int_0^{\infty}\frac{2\pi G\,dG}{k(k_0-\sqrt{G^2+k_z^2})}$$
$$=\frac{V}{8\pi^3}\int\frac{d^3k}{k(k_0-k)}, \tag{16}$$

the last equality coming from identifying $\mathbf{k}_2=\mathbf{q}+\mathbf{G}\approx\mathbf{G}$. The integral in Eq. (16) obviously diverges at the upper limit.

It is immediately noted that if we calculate exactly $\Omega_{\mathbf{q}n}(0)$ of Eq. (1) by using the $\boldsymbol{l}=0$ form of Eq. (5), together with Eqs. (6) and (9), the result is just that given by Eq. (16). Hence we conclude that the ultraviolet divergence of $\Omega_{\mathbf{q}n}(0)$ is really the same as that from the U processes of large $\mathbf{G}$'s that contribute to the full $\Omega_{\mathbf{q}n}$. Once $\Omega_{\mathbf{q}n}(0)$ of the lone exciton is rendered finite by renormalization via Eq. (12), just like the familiar Lamb shift of single atoms, the U processes of large G's that arise from the unrestricted sum over $\boldsymbol{l}$ in Eq. (14) will also be rendered finite simultaneously.

A renormalized $\Omega_{\mathbf{q}n}(0)$, like the usual Lamb shift, is comparable in magnitude to any one of the many $\Omega_{\mathbf{q}n}(\boldsymbol{l})$'s. It would not play a dominant role in Eq. (11), in contrast to its infinite contribution in Eq. (1). We may now safely combine $\Omega_{\mathbf{q}n}^{\mathrm{ren}}(0)$ with $\Omega_{\mathbf{q}n}^{\mathrm{coh}}$ of Eq. (13) to return to an unrestricted sum over all $\boldsymbol{l}$'s in the manner of Eq. (14), except that the contributions from large G's should be removed at the same time to conform to renormalization. A two-band model which we adopted from the beginning automatically excludes the larger G's and is thus seen to be consistent with the above. Accordingly it follows from Eqs. (14) and (15) that

$$\Omega_{\mathbf{q}n}^{\mathrm{ren}}=NJ_n(\mathbf{q})$$
$$=N\left[P\sum_{k_z}\mid A_{\mathbf{q}k_z,n}\mid^2\frac{1}{c(k_0-\sqrt{q^2+k_z^2})}\right] \tag{17}$$

by taking only the $\mathbf{G}=0$ term. Similarly Eqs. (2) and (7) yield, innocently,

$$\gamma_{\mathbf{q}n}=2\pi NG_n(\mathbf{q})$$
$$=2\pi N\sum_{k_z}\mid A_{\mathbf{k},n}\mid^2\delta[c(k_0-\sqrt{q^2+k_z^2})]. \tag{18}$$

Nevertheless, at the risk of belaboring our point we emphasize that, had it not been for the renormalization in Eq. (12), the direct imposition of the two-band model on Eqs. (1) and (14) would not have been justified, especially after recognizing the infinite contribution from the single term $\Omega_{\mathbf{q}n}(0)$ alone in Eq. (1).

Analogous to the decay rate $\gamma_{\mathbf{q}n}$ in Eq. (18), the renormalized frequency shift in Eq. (17) is explicitly seen to be coherently enhanced by the same factor N as a result of the interaction of the phase-matched photon amplitude with the delocalized excitonic amplitude spread over all N atoms (or N unit cells, to be exact) in the entire x-y plane. This coherent participation of N atoms is achieved at the expense of a reduction of the number of photon modes involved. When a single excited atom radiates, all $\mathbf{k}$ photon modes are possible as long as $\mid\mathbf{k}_2\mid\leq\mid\mathbf{k}\mid\sim k_0$, and the number of such $\mathbf{k}_2$ modes is $\sim(\pi k_0^2)(\frac{A}{4\pi^2})$. On the other hand, when all N atoms in a delocalized $\mathbf{q}n$ exciton radiate, only the $\mathbf{k}$ photons with the same $\mathbf{k}_2=\mathbf{q}$ are allowed. It is this trading of the photonic degrees of freedom for the atomic degrees of freedom that leads to the superradiative enhancement factor of $N(\frac{Ak_0^2}{4\pi})^{-1}\sim\frac{1}{(k_0d)^2}$ for the planar exciton over the excited atom [10].

Unlike Eq. (18), the integration over k_z in Eq. (17) is not free of pitfalls yet. When Eq. (9) is substituted into Eq. (17), though there is no longer any ultraviolet divergence from large k_z, an infrared divergence from $\mid k_z\mid\sim 0$ apparently emerges in the important case of $\mathbf{q}\approx 0$. This originates with the $H'\sim\mathbf{A}\cdot\mathbf{p}$ perturbation Hamiltonian used [9] to obtain Eq. (9). This apparent divergence implies that the lowest-order theory of this perturbation is inadequate for the present purpose. Rather than going to successively higher orders we may cure this by regrouping via a canonical transformation [18] that converts the perturbation Hamiltonian into the dipole-interaction form, $H'\sim\mathbf{r}\cdot\mathbf{E}$. This would effectively change $\mid A_{\mathbf{k},n}\mid^2\sim\frac{\mid\mathbf{x}_n\mid^2}{k}$ of Eq. (9) into $\mid\tilde{A}_{\mathbf{k},n}\mid^2\sim k\mid\mathbf{d}_n\mid^2$, given more exactly by

$$|\tilde{A}_{\mathbf{k},n}|^2 = \frac{2\pi e^2 ck}{\hbar V}\,|\hat{\epsilon}_{\mathbf{k}\lambda}\cdot\mathbf{d}_n|^2, \tag{19}$$

where

$$\mathbf{d}_n = \sum_{\rho} F_n^*(\rho)\int d^2\tau\, w_c^*(\tau-\rho)\tau w_v(\tau). \tag{20}$$

A comparison between Eqs. (4) and (20) reveals that it is the change from the momentum operator $(-i\hbar)\nabla_\tau$ to the dipole operator τ that gives rise to the modified k dependence of $|\tilde{A}_{\mathbf{k},n}|^2$ from $|A_{\mathbf{k},n}|^2$. The judicious replacement of $|A_{\mathbf{k},n}|^2$ in Eq. (17) by $|\tilde{A}_{\mathbf{k},n}|^2$ for small k_z, with the dividing line defined to be where the two equal each other, enables us to carry out the k_z integration without further ado, yielding for a one-layer film the result

$$\Omega^{\rm ren}_{\mathbf{q}\sim 0,n} = -\frac{2e^2}{\hbar c}\frac{E_{q\sim 0,n}}{\hbar}\left|\frac{\hat{\epsilon}_k\cdot\mathbf{d}_n}{d^2}\right|^2. \tag{21}$$

It is not difficult to generalize the above one-layer result to that of a thin film of thickness T, containing $n_z = T/d$ layers (as long as $k_0T < 1$):

$$\Omega^{\rm ren}_{\mathbf{q}\sim 0,n} = -\frac{2e^2}{\hbar c}\frac{E_{q\sim 0,n}}{\hbar}\left|\frac{\hat{\epsilon}_k\cdot\mathbf{d}_n}{d}\right|^2\left(\frac{T}{d}\right)(1-k_0T). \tag{22}$$

The above expression can be rewritten as

$$\Omega^{\rm ren}_{\mathbf{q}\sim 0,n} = -\gamma_{\rm single}\left(\frac{1}{k_0d}\right)^2\left(\frac{T}{d}\right)(1-k_0T), \tag{23}$$

where

$$\gamma_{\rm single} = \Gamma_{\mathbf{q},n}(0) = \frac{2e^2}{\hbar c}\frac{E_{q\sim 0,n}}{\hbar}\,|\,k_0d_n\,|^2 \tag{24}$$

is the radiative decay rate of a single isolated exciton. Its presence in Eq. (23) is traceable to the neglect of the directional details [19] involving $|\hat{\epsilon}_k\cdot\mathbf{d}_n|^2$ in Eq. (22) for a first estimate.

In Eq. (23) the enhancement factor $(\frac{1}{k_0d})^2(\frac{T}{d})(1-k_0T)$ due to the coherence effect [5,9,16] is explicitly displayed. The extra factor involving the number of layers within a half wavelength appears since the excitonic amplitude spread among these layers is phase-matched almost layer by layer with the corresponding photonic amplitudes.

In the above treatment, the superradiative exciton modes ($q < k_0$) are considered. On the other hand, if q is larger than k_0, the frequency shift can be shown [5] to have a sign opposite to that in Eq. (22). However, these excitonic modes are not capable of radiative decay (they are called "trapped modes" in Ref. [10]). This is simply because the energy $\hbar ck_0$ of that exciton is not sufficient to produce a photon of momentum $\mathbf{k} = \mathbf{q} + \hat{\mathbf{z}}k_z$. Energies in these modes are thus trapped in the film.

One can see from Eq. (23) that the superradiative enhancement factor is huge, $\sim 10^7$ for Wannier excitons in the optical range. However, due to the extreme smallness of $\gamma_{\rm single}$ itself, observation of $\Omega^{\rm ren}_{q\sim 0,n}$ is not expected to be easy. Its linear dependence on the thickness T may be a useful feature that distinguishes it from the usual quantum well confinement. The most favorable range of T should be $a_B \ll T \ll \lambda_0$, where a_B is the Bohr radius of the exciton. The linear dependence on the excitonic energy $E_{\mathbf{q}n}$ also suggests the use of high pressure that could cause considerable changes in the band gap and hence in $E_{\mathbf{q}n}$.

In conclusion, we have shown in calculating the radiative frequency shift of an exciton in a thin semiconductor film that, first, a two-band model can only be justified after a proper renormalization adapted to removing the ultraviolet divergence pertaining to a correlated system has been made. Second, the renormalized frequency shift is superradiatively enhanced, analogously to its more familiar decay-rate counterpart, by the coherence effect. The distinguishing features are pointed out and may be observable in a suitably designed experiment.

One of the authors (Y.C.L.) would like to thank Professor B. Weinstein of SUNY at Buffalo for a helpful discussion and for pointing out the high pressure experiment. This work is partially supported by the National Science Council, Taiwan, ROC.

[a] Sabbatical leave from Department of Physics, SUNY at Buffalo, Buffalo, NY 14260.
[b] Sabbatical leave from Department of Physics, University of Nebraska, Omaha, NE 68101.

[1] R. H. Dicke, Phys. Rev. **93**, 99 (1954).
[2] V. Ernst and P. Stehle, Phys. Rev. **176**, 1456 (1968).
[3] Y. C. Lee and D. L. Lin, Phys. Rev. **183**, 147 (1969); **183**, 150 (1969), and references contained therein.
[4] F. Arecchi and D. Kin, Opt. Commun. (Netherlands) **2**, 324 (1970).
[5] Y. C. Lee and D. L. Lin, Phys. Rev. A **6**, 388 (1972).
[6] P. S. Lee and Y. C. Lee, Phys. Rev. A **8**, 1727 (1973).
[7] J. J. Hopfield, Phys. Rev. **112**, 1555 (1958).
[8] K. C. Liu, Y. C. Lee, and Y. Shan, Phys. Rev. B **11**, 978 (1975).
[9] K. C. Liu and Y. C. Lee, Physica (Amsterdam) **104A**, 131 (1980).
[10] Y. C. Lee and K. C. Liu, J. Phys. C **14**, L281 (1981).
[11] J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moore, C. Foxon, and R. J. Elliot, Phys. Rev. Lett. **59**, 2337 (1987).
[12] E. Hanamura, Phys. Rev. B **38**, 1228 (1988).
[13] F. C. Spano, J. R. Kukinski, and S. Mukamel, Phys. Rev. Lett. **65**, 211 (1990).
[14] T. Itch, M. Furnmiya, T. Ikehara, and C. Gourdon, Solid State Commun. **73**, 271 (1990).
[15] K. Misawa, H. Yao, T. Hayashi, and T. Kobayashi, J. Chem. Phys. **94**, 4131 (1991).
[16] J. Knoester, Phys. Rev. Lett. **68**, 654 (1992).
[17] O. Brandt, G. C. La Rocca, A. Heberle, A. Ruiz, and K. Ploog, Phys. Rev. B **45**, 3803 (1992).
[18] G. Baym, *Lectures On Quantum Mechanics* (Benjamin, New York, 1969), pp. 297 and 298.
[19] Y. C. Lee and P. S. Lee, Phys. Rev. B **10**, 344 (1974).