Decay rate and renormalized frequency shift of superradiant excitons: Crossover from two-dimensional to three-dimensional crystals

Yueh-Nan Chen and Der-San Chuu*
Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan
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The superradiant decay rate and renormalized frequency shift of Wannier excitons in a semiconductor film of N layers are studied. It is found that both the decay rate and renormalized frequency shift show oscillatory dependence on layer thickness. The crossover from the superradiant exciton to the bulk polariton when varying N from 1 to ∞ is also examined.

Since Dicke's pioneer work[1] on coherent spontaneous radiation of gas, the coherent effect for spontaneous radiation of various systems has attracted extensive interest both theoretically and experimentally.[2,3] The coherent radiation phenomena for the atomic system was intensively investigated in the late 1960's.[4–6] It was found that the distances between atoms in crystal are much smaller than the ones in gas, so the coherent effect would be much greater than that in gas.[7] But as it was well known,[8] the excitons will couple with photons to form a polariton—an eigenstate of the combined system consisting of the crystal and the radiation field which does not decay radiatively. What makes the excitons trapped in the bulk crystal is the conservation of crystal momentum. If one considers a thin film,[9,10] the excitons can undergo radiative decay as a result of the broken crystal symmetry. The decay rate of excitons in a thin film is enhanced by a factor of $(\lambda/d)^2$ compared to a lone exciton in an empty lattice, where λ is the wave length of emitted photon and d is the lattice constant of the film.

Recently, with the advance of microfabrication technologies such as molecular beam epitaxy, it has become possible to fabricate various structure of microcrystal with fine quality and novel properties, such as quantum well, superlattice, quantum dot, and quantum wire. The excitons in quantum well can exhibit the behavior between purely three dimensions (3D) and 2D. Many investigations on the radiative linewidth of excitons in quantum wells have been performed. Cebulla *et al.*[11] found an abnormal increase of excitonic radiative life time with the decrease of well width below 5 nm for $In_xGa_{1-x}As/InP$ quantum well. The radiative lifetimes of excitons in InAs quantum sheets were measured by Brandt *et al.*,[12] they observed the increasing of radiative life-time with the decreasing of well thickness. Hanamura[13] investigated theoretically the radiative decay rate of quantum dot and quantum well. The obtained results are similar to that of Lee and Liu's[10] for thin films. Knoester[14] obtained the dispersion relation of Frankel excitons of quantum slab. The oscillating dependence of the radiative width of the exciton-like polaritons with the lowest energy on the crystal thickness was found. Recently, Björk *et al.*[15] examined the relationship between atomic and excitonic superradiance in thin and thick slab geometries. They demonstrated that superradiance can be treated by a unified formalism for atoms, Frenkel excitons, and Wannier excitons. In a previous work of Agranovich *et al.*,[16] a detailed microscopic study of Frenkel exciton-polariton in crystal slabs of arbitrary thickness is performed.

In reality, superradiance is accompanied by frequency shifts, as pointed out in Ref. 17. However, the coherent frequency shift of an exciton has received little attention—especially that concerning the crossover from two-dimensional to three-dimensional case. One of the reasons is the difficulty arose from divergent nature of frequency shift, both infrared and ultraviolet. In this paper, we approach this problem by using the method of renormalization proposed by Lee *et al.*[17] in a semiconductor slab of arbitrary thickness. In addition we also show the decay rate of Wannier excitons as a function of layer thickness. It will be found the higher-order modes have larger maximum decay rates, as predicted by G. Björk *et al.*[15]

Consider a crystal slab with simple cubic structure which consists of N layers. The state of the Wannier exciton can be specified as $|\mathbf{k},n,l,m\rangle$, where $\mathbf{k}=(\mathbf{q},k_z)$ is the exciton wave number with $\mathbf{q}(k_z)$ parallel(normal) to the slab and n,l, and m are quantum numbers for internal structure of the exciton. Here, k_z takes the values $k_z=2\pi n_z/Nd$, with d the lattice spacing and n_z an integer that is limited to one Brillouin zone $(n_z=0,1,\ldots,N-1)$. The Hamilton for the exciton is

$$H_{ex}=\sum_{\mathbf{k}nlm} E_{\mathbf{k}n} c^{\dagger}_{\mathbf{k}nlm} c_{\mathbf{k}nlm}, \tag{1}$$

where $c^{\dagger}_{\mathbf{k}nlm}$ and $c_{\mathbf{k}nlm}$ are the creation and destruction operators of the exciton, respectively. The Hamiltonian of the photon is

$$H_{ph}=\sum_{\mathbf{q}'k'_z\lambda} \hbar c(q'^2+k'^2_z)^{1/2} b^{\dagger}_{\mathbf{q}'k'_z\lambda} b_{\mathbf{q}'k'_z\lambda}, \tag{2}$$

where $b^{\dagger}_{\mathbf{q}'k'_z\lambda}$ and $b_{\mathbf{q}'k'_z\lambda}$ are the creation and destruction operators of the photon, respectively. The wave vector $\mathbf{k}'$ of the photon were separated into two parts: $\mathbf{q}'$ is the parallel component of $\mathbf{k}'$ on the crystal plane such that $k'^2=q'^2+k'^2_z$.

The interaction between the exciton and the photon can be expressed as

$$\begin{aligned} H' = \sum_i \sum_{\mathbf{q}'k'_z\lambda} \frac{e}{mc} \sqrt{\frac{2\pi\hbar c}{(q'^2+k'^2_z)^{1/2} v}} \\ \times [b^{\dagger}_{\mathbf{q}'k'_z\lambda} \exp(i\mathbf{q}'\cdot\boldsymbol{\tau}_i + ik'_z\tau_z) + b_{\mathbf{q}'k'_z\lambda} \\ \times \exp(-i\mathbf{q}'\cdot\boldsymbol{\tau}_i - ik'_z\tau_z)](\boldsymbol{\epsilon}_{\mathbf{q}'k'_z\lambda}\cdot\mathbf{p}_i), \end{aligned} \tag{3}$$

where $\tau=(\tau_i,\tau_z)$ is a position vector of the electron i in the crystal slab, $\mathbf{p}_i$ is the corresponding momentum operator of the electron i, and $\boldsymbol{\epsilon}_{\mathbf{q}'k_z'\lambda}$ is the polarization vector of the photon.

The essential quantity involved is the matrix element of H' between the ground state $|G\rangle$ and the Wannier exciton state $|\mathbf{k},n,l,m\rangle$. The Wannier exciton state can be expressed as

$$|\mathbf{k},n,l,m\rangle=\sum_{\mathbf{l},\boldsymbol{\rho},I,J} U^*_{\mathbf{k}nlm}(\mathbf{l},\boldsymbol{\rho},I,J)|c,(\mathbf{l}+\boldsymbol{\rho},I+J);v,(\mathbf{l},J)\rangle, \tag{4}$$

and the interaction matrix elements can be written as

$$\langle\mathbf{k},n,l,m|H'|G\rangle=\sum_{\mathbf{l},\boldsymbol{\rho},I,J}\langle c,(\mathbf{l}+\boldsymbol{\rho},I+J);v, \times(\mathbf{l},J)|U^*_{\mathbf{k}nlm}(\mathbf{l},\boldsymbol{\rho},I,J)H'|G\rangle, \tag{5}$$

in which the excited state $|c,(\mathbf{l}+\boldsymbol{\rho},I+J);v,(\mathbf{l},J)\rangle$ is defined as

$$|c,(\mathbf{l}+\boldsymbol{\rho},I+J);v,(\mathbf{l},J)\rangle=a^\dagger_{c,(\mathbf{l}+\boldsymbol{\rho},I+J)}a_{v,(\mathbf{l},J)}|G\rangle, \tag{6}$$

where $a^\dagger_{c,(\mathbf{l}+\boldsymbol{\rho},I+J)}$ ($a_{v,(\mathbf{l},J)}$) is the creation (destruction) operator for an electron (hole) in the conduction (valence) band at site $(\mathbf{l}+\boldsymbol{\rho},I+J)[(\mathbf{l},J)]$, and $\mathbf{l}$ and $\boldsymbol{\rho}$ (I and J) are the two-(one-) dimensional vector parallel (normal) to the layer plane. The expansion coefficient $U^*_{\mathbf{k}nlm}(\mathbf{l},\boldsymbol{\rho},I,J)$ is the exciton wave function in the N layer crystal slab:

$$U^*_{\mathbf{k}nlm}(\mathbf{l},\boldsymbol{\rho},I,J)=\frac{1}{\sqrt{N'}}\frac{1}{\sqrt{N}}\exp(i\mathbf{q}\cdot\mathbf{r}_c+ik_zr_c')F_{nlm}(\boldsymbol{\rho},z), \tag{7}$$

where the coefficient $1/\sqrt{N'}$ is for the normalization of the state $|\mathbf{k},n,l,m\rangle$, $r_c'=[m_e^*(I+J)+m_h^*J]/(m_e^*+m_h^*)$, and $\mathbf{r}_c$ is the center of mass of the exciton in the x-y plane. Here, m_e^* and m_h^* are, respectively, the effective masses of the electron and the hole. $F_{nlm}(\boldsymbol{\rho},z)$ is the hydrogenic wave function in the quantum well (with well width $L=Nd$), and can be determined by variational method.[18]

After summing over $\mathbf{l}$ and J, we have

$$\langle\mathbf{k},n,l,m|H'|G\rangle=\sum_{k_z'\lambda\mathbf{g}}\frac{1}{\sqrt{N}}\frac{1-e^{-i(k_z-k_z')Nd}}{1-e^{-i(k_z-k_z')d}}\frac{e}{mc} \times\sqrt{\frac{2\pi\hbar c}{((\mathbf{q}+\mathbf{g})^2+k_z'^2)^{1/2}v}} \times[b_{(\mathbf{q}+\mathbf{g})k_z'\lambda}(\boldsymbol{\epsilon}_{(\mathbf{q}+\mathbf{g})k_z'\lambda}\cdot\mathbf{A}_{(\mathbf{q}+\mathbf{g})k_znlm}) +b^\dagger_{-\mathbf{q}k_z'\lambda}(\boldsymbol{\epsilon}_{-(\mathbf{q}+\mathbf{g})k_z'\lambda}\cdot\mathbf{A}_{(\mathbf{q}+\mathbf{g})k_znlm})], \tag{8}$$

where

$$\mathbf{A}_{\mathbf{q}k_znlm}=\sqrt{N'}\sum_{\boldsymbol{\rho},I}F^*_{nlm}(\boldsymbol{\rho},z)\int d^3\boldsymbol{\tau}w_c[\boldsymbol{\tau}-(\boldsymbol{\rho},I)] \times\exp\left(i\mathbf{q}\cdot\left(\tau_i-\frac{m_e^*\boldsymbol{\rho}}{m_e^*+m_h^*}\right) +i\left(k_z'\tau_z-\frac{m_e^*I}{m_e^*+m_h^*}k_z\right)\right)(-i\hbar\boldsymbol{\nabla})w_v[\boldsymbol{\tau}] \tag{9}$$

and $\mathbf{g}$ is the reciprocal lattice vector in the layer plane.

Hence the interaction between the exciton and the photon (in the resonance approximation) can be written in the form

$$H'=\sum_{\mathbf{q}k_znlm\mathbf{g}}\sum_{k_z'}D_{(\mathbf{q}+\mathbf{g})k_z'nlm}b_{(\mathbf{q}+\mathbf{g})k_z'}c^\dagger_{(\mathbf{q}+\mathbf{g})k_znlm}+\text{h.c.}, \tag{10}$$

where

$$D_{\mathbf{q}k_z'nlm\mathbf{g}}=\frac{1}{\sqrt{N}}\frac{1-e^{-i(k_z-k_z')Nd}}{1-e^{-i(k_z-k_z')d}}\frac{e}{mc}\sqrt{\frac{2\pi\hbar c}{((\mathbf{q}+\mathbf{g})^2+k_z'^2)^{1/2}v}} \times\boldsymbol{\epsilon}_{(\mathbf{q}+\mathbf{g})k_z'\lambda}\cdot\mathbf{A}_{(\mathbf{q}+\mathbf{g})k_znlm}. \tag{11}$$

Now, we assume that at time $t=0$ the crystal slab is initially characterized by the presence of a Wannier exciton in the mode $\mathbf{q}+\mathbf{g},k_z,n,l,m$. For time $t>0$, the state $|\psi(t)\rangle$ can be written as

$$|\psi(t)\rangle=f_0(t)|\mathbf{q}+\mathbf{g},k_z,n,l,m;0\rangle +\sum_{k_z'}f_{G;(\mathbf{q}+\mathbf{g})k_z'}(t)|G;(\mathbf{q}+\mathbf{g})k_z'\rangle, \tag{12}$$

where $|\mathbf{q},k_z,n,l,m\rangle$ is the state with a Wannier exciton in the mode $\mathbf{q}+\mathbf{g},k_z,n,l,m$ in the crystal slab but no photons, and $|G;(\mathbf{q}+\mathbf{g})k_z'\rangle$ means the crystal slab in its ground state and a photon of $\mathbf{q}+\mathbf{g},k_z'$ in the radiation field.

By the method of Heitler and Ma in the resonance approximation, the probability amplitude $f_0(t)$ can be expressed as[10]

$$f_0(t)=\exp\left(-i\Omega_{(\mathbf{q}+\mathbf{g})k_znlm}t-\frac{1}{2}\gamma_{(\mathbf{q}+\mathbf{g})k_znlm}t\right), \tag{13}$$

where

$$\gamma_{(\mathbf{q}+\mathbf{g})k_znlm}=2\pi\sum_{k_z'}|D_{(\mathbf{q}+\mathbf{g})k_z'nlm}|^2\delta(\omega_{(\mathbf{q}+\mathbf{g})k_znk_z'}) \tag{14}$$

and

$$\Omega_{(\mathbf{q}+\mathbf{g})k_znlm}=\mathcal{P}\sum_{k_z'}\frac{|D_{(\mathbf{q}+\mathbf{g})k_z'nlm}|^2}{\omega_{(\mathbf{q}+\mathbf{g})k_znk_z'}} \tag{15}$$

with $\omega_{(\mathbf{q}+\mathbf{g})k_znk_z'}=E_{qk_zn}/\hbar-c\sqrt{(\mathbf{q}+\mathbf{g})^2+k_z'^2}$. Here $\gamma_{(\mathbf{q}+\mathbf{g})k_znlm}$ and $\Omega_{(\mathbf{q}+\mathbf{g})k_znlm}$ are, respectively, the decay rate and frequency shift of the exciton.

If we neglect the Umklapp process, the Wannier exciton decay rate in the optical region can be calculated straightforwardly and is given by

$$\gamma_{\mathbf{q}k_z nlm}=\begin{cases} G^{-}_{k_z n_z}\dfrac{3}{2}\dfrac{\pi N'}{Ak_0^3}\gamma_0\dfrac{(k_0^2-q^2)|\hat{\chi}_{nlm}\times\hat{z}|^2+|\hat{\chi}_{nlm}\times\mathbf{q}|^2}{(k_0^2-q^2)^{1/2}}, & \sqrt{q^2+k_z^2}<k_0, \\ 0, \quad \text{otherwise} \end{cases} \tag{16}$$

where A is the area of the layer plane, $k_0=E_{qk_z n}/\hbar$, $\overline{k_z}=(k_0^2-q^2)^{1/2}$,

$$\boldsymbol{\chi}_{nlm}=\sum_{\boldsymbol{\rho},I} F^*_{nlm}(\boldsymbol{\rho},z)\int d^3\boldsymbol{\tau} w_c[\boldsymbol{\tau}-(\boldsymbol{\rho},I)](-i\hbar\boldsymbol{\nabla})w_v[\boldsymbol{\tau}], \tag{17}$$

$$\gamma_0=\frac{4e^2\hbar k_0}{3m^2c^2}|\boldsymbol{\chi}_{nlm}|^2, \tag{18}$$

and

$$G^{-}_{k_z n_z}=\frac{1}{N}\frac{\sin^2(\pi n_z-Nd\overline{k_z}/2)}{\sin^2(\pi n_z/N-d\overline{k_z}/2)}, \quad n_z=0,1,2\cdots. \tag{19}$$

Here, χ^*_{nlm} represents the effective dipole matrix element for an electron to jump from the excited Wannier state in the conduction band back to the hole state in the valence band. We see from Eq. (16) that $\gamma_{\mathbf{q}k_z nlm}$ is proportional to $1/(k_0 d)^2$.[17] This is just the superradiance factor implying the coherent contributions from atoms within half a wavelength or so.[19]

We now pay attention to the factor $G^{-}_{k_z n_z}$ in Eq. (16). For the limiting case $N=1$ (monolayer), the factor $G^{-}_{k_z n_z}=1$ and the wave function $F_{nlm}(\boldsymbol{\rho},z)$ becomes two-dimensional hydrogenic wave function. It means our result can reduce to the thin film case.[10] In Fig. 1 we have plotted the factor $G^{-}_{k_z n_z}$ as a function of layer numbers for $n_z=0$ mode. In ploting the figure, $G^{-}_{k_z n_z}$ is expressed in terms of

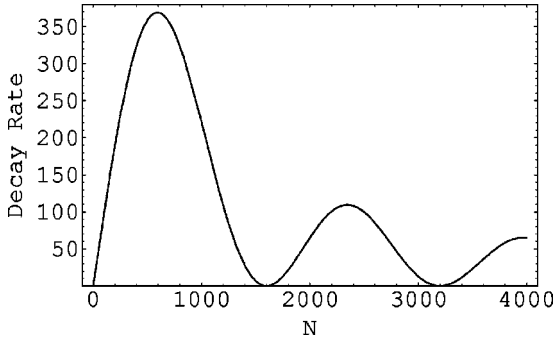

FIG. 1. Decay rate of a slab with superradiant Wannier excitons as a function of layer numbers. The vertical and horizontal units are

$$\frac{3}{2}\frac{\pi N'}{Ak_0^3}\gamma_0\frac{(k_0^2-q^2)|\hat{\chi}_{nlm}\times\hat{z}|^2+|\hat{\chi}_{nlm}\times\mathbf{q}|^2}{(k_0^2-q^2)^{1/2}}$$

and layer numbers N, respectively. In this and following graphs we have assumed $\overline{k_z}=2\pi/\lambda$, $\lambda=8000$ Å, and lattice spacing $d=5$ Å.

$$\frac{3}{2}\frac{\pi N'}{Ak_0^3}\gamma_0\frac{(k_0^2-q^2)|\hat{\chi}_{nlm}\times\hat{z}|^2+|\hat{\chi}_{nlm}\times\mathbf{q}|^2}{(k_0^2-q^2)^{1/2}},$$

therefore the coordinate of the figure now represents the decay rate formally. In addition, we have also assumed $\overline{k_z}=2\pi/\lambda$, $\lambda=8000$ Å, and lattice spacing $d=5$ Å in the numerical calculations. With the increasing of layer numbers, the decay rate shows the oscillatory and decreasing behavior. There is also a maximum value of the factor $G^{-}_{k_z n_z}$ when $N=592$, and the oscillation period is 1600(layers). This is similar to the Knoester's result[14] which comes from the interference between the radiation fields emitted by the various excitons in the slab. In sufficiently thick slab, it becomes radiatively stable polariton result. Figure 2 shows the factor $G^{-}_{k_z n_z}$ of the higher-order mode ($n_z=1,2$). One can see from the figure the maximum decay rate occurred at larger N. Due to the k_z vector conservation, the higher-order modes had larger maximum decay rate. Our result is also in good agreement with the recent study of optical properties transition from thin film to bulk.[15]

Now let us present out results for the renormalized frequency shift. The frequency shift in Eq. (15) can be expressed as

$$\Omega_{(\mathbf{q}+\mathbf{g})k_z nlm}=\frac{2\pi e^2}{\hbar m^2c^2 v}\mathcal{P}\sum_{\mathbf{g}k'_z\lambda}\frac{1}{N}\frac{\sin^2(\pi n_z-Ndk'_z/2)}{\sin^2(\pi n_z/N-dk'_z/2)}\times\frac{|\boldsymbol{\epsilon}_{(\mathbf{q}+\mathbf{g})k'_z\lambda}\cdot\boldsymbol{\chi}_{nlm}|^2}{\sqrt{(\mathbf{q}+\mathbf{g})^2+k_z'^2}}\frac{1}{k_0-\sqrt{(\mathbf{q}+\mathbf{g})^2+k_z'^2}}. \tag{20}$$

As seen from above, the frequency shift suffers from ultraviolet divergence when k'_z and $\mathbf{g}$ are large, and has infrared

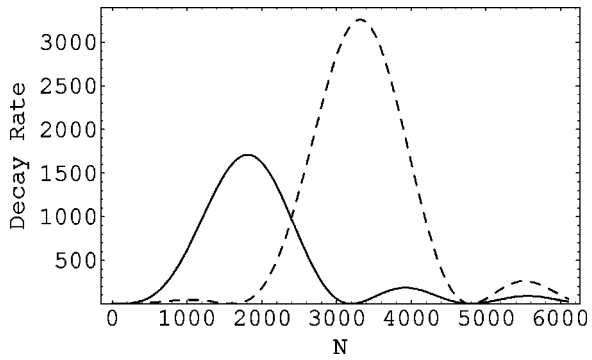

FIG. 2. Decay rate of the higher-order mode. The solid line corresponds to $n_z=1$, whereas the dashed line is $n_z=2$. The vertical and horizontal units are

$$\frac{3}{2}\frac{\pi N'}{Ak_0^3}\gamma_0\frac{(k_0^2-q^2)|\hat{\chi}_{nlm}\times\hat{z}|^2+|\hat{\chi}_{nlm}\times\mathbf{q}|^2}{(k_0^2-q^2)^{1/2}}$$

and layer numbers N, respectively.

divergence when the denominator approaches zero. Recently, Lee, Chuu, and Mei[17] employed the usual renormalization procedure to find that the ultraviolet divergence comes from the inclusion of Umklapp process. Therefore, the renormalized result ($\mathbf{q}\sim 0$ and $n_z=0$) can be written as

$$\Omega^{\rm ren}_{\mathbf{q}k_z nlm}=\frac{2\pi e^2 N'}{\hbar m^2 c^2 v}\mathcal{P}\sum_{k_z'\lambda}\frac{1}{N}\frac{\sin^2(Ndk_z'/2)}{\sin^2(dk_z'/2)}$$

$$\times\frac{|\boldsymbol{\epsilon}_{\mathbf{q}k_z'\lambda}\cdot\mathbf{A}_{\mathbf{q}k_z nlm}|^2}{k_z'}\frac{1}{k_0-k_z'}. \tag{21}$$

We now turn to the infrared divergent problem. As seen from Eq. (21), there is divergent problem when $k_z'\sim 0$ or $k_z'\sim k_0$. This can be overcome by substituting $-i\hbar\boldsymbol{\nabla}$ by $-imck_z'\boldsymbol{\tau}$ (Ref. 17) in Eq. (17) when k_z' is small. It is equivalent to the dipole-interaction form, $H'\sim\mathbf{r}\cdot\mathbf{E}$. With this treatment, we have

$$\Omega^{\rm ren}_{\mathbf{q}k_z nlm}=N'\mathcal{P}\sum_{k_z'\lambda}B_{\mathbf{q}k_z' nlm}\left(\frac{1}{k_0-k_z'}\right)\frac{1}{N}\frac{\sin^2(Ndk_z'/2)}{\sin^2(dk_z'/2)} \tag{22}$$

with

$$B_{\mathbf{q}k_z' nlm}=\begin{cases}\dfrac{2\pi e^2}{m^2c^2\hbar k_z' v}|\boldsymbol{\epsilon}_{\mathbf{q}k_z'\lambda}\cdot\boldsymbol{\chi}_{nlm}|^2, & \text{when is } k_z' \text{ large}\\ \dfrac{2\pi e^2 k_z'}{\hbar v}|\boldsymbol{\epsilon}_{\mathbf{q}k_z'\lambda}\cdot\mathbf{d}_{nlm}|^2, & \text{when is } k_z' \text{ is small}\end{cases} \tag{23}$$

where

$$\mathbf{d}_{\mathbf{q}k_z nlm}=\sum_{\boldsymbol{\rho},I}F^*_{nlm}(\boldsymbol{\rho},z)\int d^3\boldsymbol{\tau}w_c[\boldsymbol{\tau}-(\boldsymbol{\rho},I)](-i\hbar\boldsymbol{\nabla})w_v[\boldsymbol{\tau}]. \tag{24}$$

Equation (22) cannot be evaluated analytically. But for the thin film containing T layers, the renormalized frequency shift can be approximated as

$$\Omega^{\rm ren}_{\mathbf{q}k_z nlm}=-\frac{2e^2}{\hbar c}\frac{E_{n,\mathbf{q}\sim 0}}{\hbar}\left|\frac{\boldsymbol{\epsilon}_{k_z'}\cdot\mathbf{d}_{nlm}}{d}\right|^2\cdot T. \tag{25}$$

This is the just result obtained by Lee *et al.*[17] Similar to the decay rate, the above expression can be rewritten as

$$\Omega^{\rm ren}_{\mathbf{q}k_z nlm}=-\gamma_{\rm single}\left(\frac{1}{k_0 d}\right)^2 T, \tag{26}$$

where

$$\gamma_{\rm single}=\frac{2e^2}{\hbar c}\frac{E_{n,\mathbf{q}\sim 0}}{\hbar}|k_0 d_{nlm}|^2 \tag{27}$$

is the radiative decay rate of a single isolated exciton. The enhanced factor $(1/k_0 d)^2 T$ is due to the coherent effect.

In Fig. 3, we numerically calculated the frequency shift as a function of layer numbers. One can see from the figure the renormalized frequency shift also shows the oscillatory behavior, and the maximum frequency shift is reached about $N=1070$. In addition, the value of the frequency shift changes from negative to positive when the layer numbers is about 380. This negative phase delay between the decay rate in Fig. 1 and frequency shift in Fig. 3 comes from the factor $[1/(k_0-k_z')]$ in Eq. (22). As we integrate k_z' from zero to infinity, there is a competition between the negative and positive value of this factor. With the increasing of layer numbers, the frequency shift also decreases and gradually approaches to zero which agrees with the case of 3D limit. In spite of the oscillatory behavior, the oscillation period of frequency shift is not really a constant. In fact, the oscillation period, from the first maximum point to the next one, is 1675 layers, and gradually approaches to 1600 layers with the increasing of layer numbers. This is because as the layer number N is large, the k_z' in Eq. (22) must center around k_0 because of the conservation of momentum. Thus Eq. (22) can be approximated as

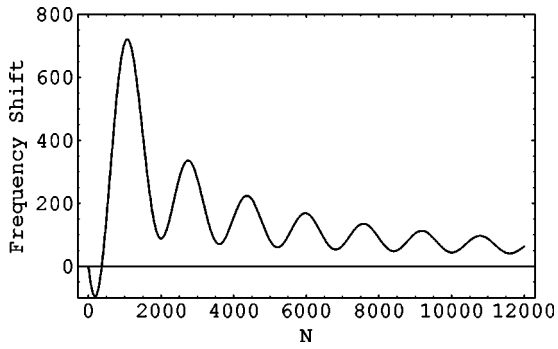

FIG. 3. The renormalized frequency shift of Wannier excitons as a function of layer numbers. The vertical and horizontal units are $\gamma_{\rm single}(1/k_0 d)^2$ and layer numbers N, respectively.

$$\Omega^{\rm ren}_{\mathbf{q}k_z nlm}\sim A(\overline{k_0})\frac{1}{N}\frac{\sin^2(Nd\overline{k_0})}{\sin^2(d\overline{k_0})}, \tag{28}$$

where $\overline{k_0}=k_0+\Delta$ is the mean value of the integration. As the layer number N becomes larger, the mean value $\overline{k_0}$ gets closer to k_0. This explains the asymptotic behavior of the oscillation period.

In conclusion, we have shown the decay rate and the frequency shift of Wannier exciton in a N-layer crystal slab. Similar to the case of Frenkel exciton, the decay rate of Wannier exciton shows oscillatory behavior with the increasing of layer numbers. By the renormalization method, we have also calculated the frequency shift as a function layer numbers. It is found that the frequency shift also shows oscillatory behavior. Both the decay rate and frequency shift can approach correct 2D and 3D limit.

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*Author to whom correspondence should be addressed. Fax: 886-3-5725230, Electronic address: dschuu@cc.nctu.edu.tw

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