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Decay rate of a Wannier exciton in low-dimensional systems

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Abstract. – The superradiant decay rate of a Wannier exciton in a one-dimensional system is studied. The crossover behavior from 1D chain to 2D film is also examined. It is found that the decay rate shows an oscillatory dependence on the channel width L . When the quasi-1D channel is embedded with planar microcavities, it is shown that the dark mode exciton can be examined experimentally.

Since Dicke [1] pointed out the concept of superradiance, 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 were intensively investigated in the late sixties [4–6]. One of the limiting cases of superradiance is the exciton-polariton state in solid-state physics. When a Frenkel exciton couples to the radiation field in a small system which contains N lattice points, it represents one excited atom in some site and $N - 1$ unexcited atoms in the others. According to Dicke's theory, the decay rate of the system will be enhanced by a factor N . But, as was well known in a 3D bulk crystal, the excitons will couple with photons to form polaritons—the eigenstate of the combined system consisting of the crystal and the radiation field which does not decay radiatively [7]. What makes the excitons trapped in the bulk crystal is the conservation of crystal momentum. If one considers a linear chain or a thin film, the exciton can undergo radiative decay as a result of the broken crystal symmetry. The decay rate of the exciton is enhanced by a factor of λ/d in a linear chain [8] and $(\lambda/d)^2$ for 2D exciton-polariton [9, 10], where λ is the wavelength of the emitted photon and d is the lattice constant of the linear chain or the thin film.

Lots of investigations on the radiative linewidth of the excitons have been performed. The first observation of a superradiant short lifetime has been performed by Ya. Aaviksoo *et al.* [11] on surface states of the anthracene crystal. Later, Deveaud *et al.* [12] measured the radiative lifetime of free excitons in GaAs quantum wells and observed the enhanced radiative recombination of the excitons. Hanamura [13] investigated theoretically the radiative decay rate of quantum dot and quantum well. The obtained results are in agreement with that of

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Lee and Liu's [10] prediction for thin films. Knoester [14] obtained the dispersion relation of Frankel excitons of quantum slab. The oscillating dependence of the radiative width of the excitonlike 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 Agranovich *et al.*'s work [16], a detailed microscopic study of Frenkel exciton-polariton in crystal slabs of arbitrary thickness was performed.

For lower-dimensional systems, Ivanov and Haug [17] predicted the existence of exciton crystal, which favors coherent emission in the form of superradiance, in quantum wires. Manabe *et al.* [18] considered the superradiance of interacting Frenkel excitons in a linear chain. Recently, with the advances of the modern fabrication technology, it has become possible to fabricate the planar microcavities incorporating quantum wires [19]. This makes it interesting to study this problem more carefully. In this paper, we will investigate the radiative decay of the Wannier exciton in a quantum channel and the crossover behavior from 1D chain to 2D film. Application to real semiconductor microcavity will also be considered.

Let us first consider a linear chain with lattice spacing d . The state of the Wannier exciton can be specified as $|k_z, n\rangle$, where k_z is the exciton wave number in the chain direction and n is the quantum number for the internal structure of the exciton. The Hamiltonian for the exciton is

$$H_{\text{ex}} = \sum_{k_z n} E_{k_z n} c_{k_z n}^\dagger c_{k_z n}, \quad (1)$$

where $c_{k_z n}^\dagger$ and $c_{k_z n}$ are the creation and destruction operators of the exciton, respectively. The Hamiltonian of the free photon is

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

where $b_{\mathbf{q}' k'_z \lambda}^\dagger$ and $b_{\mathbf{q}' k'_z \lambda}$ are, respectively, the creation and destruction operators of the photon, and λ runs through the indices of x, y , and z . The wave vector \mathbf{k}' of the photon was separated into two parts: k'_z is the parallel component of \mathbf{k}' along the linear chain such that $k'^2 = q'^2 + k_z'^2$.

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

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

where m is the electron mass, $\boldsymbol{\tau}_i$ is a position vector of the electron i in the linear chain, \mathbf{p}_i is the corresponding momentum of the electron i operator, 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 $|k_z, n\rangle$. We know that the interaction matrix elements of H' can be written as

$$\langle k_z, n | H' | G \rangle = \sum_{l, \rho} \langle c, l + \rho; v, l | U_{k_z n}^*(l, \rho) H' | G \rangle, \quad (4)$$

because the Wannier exciton state can be expressed as

$$|k_z, n\rangle = \sum_{l, \rho} U_{k_z n}^*(l, \rho) |c, l + \rho; v, l\rangle, \quad (5)$$

in which the excited state $|c, l + \rho; v, l\rangle$ is defined as

$$|c, l + \rho; v, l\rangle = a_{c, l + \rho}^\dagger a_{v, l} |G\rangle, \quad (6)$$

where $a_{l + \rho}^\dagger (a_{v, l})$ is the creation (destruction) operator of an electron in the conduction band (c) (valence band (v)) at lattice site $l + \rho(l)$. The expansion coefficient $U_{k_z n}^*(l, \rho)$ is the exciton wave function in the linear chain:

$$U_{k_z n}^*(l, \rho) = \frac{1}{\sqrt{N'}} e^{ik_z r_c} F_n(\rho), \quad (7)$$

where the coefficient $1/\sqrt{N'}$ is for the normalization of the state $|k_z, n\rangle$, and $F_n(\rho)$ and r_c represent the one-dimensional hydrogenic wave function and the center of mass of the exciton, respectively.

After summing over l in eq. (4), we have

$$\langle k_z, n | H' | G \rangle = \sum_{\mathbf{q}'\lambda} \frac{e}{mc} \sqrt{\frac{2\pi\hbar c}{(q'^2 + k_z^2)^{1/2} v}} [b_{\mathbf{q}'k_z\lambda} (\boldsymbol{\epsilon}_{\mathbf{q}'k_z\lambda} \cdot \mathbf{A}_{k_z n}) + b_{-\mathbf{q}'k_z\lambda}^\dagger (\boldsymbol{\epsilon}_{-\mathbf{q}'k_z\lambda} \cdot \mathbf{A}_{k_z n})], \quad (8)$$

where

$$\mathbf{A}_{k_z n} = \sqrt{N'} \sum_{\rho} F_n(\rho) \int d\boldsymbol{\tau} w_c(\boldsymbol{\tau} - \rho) \exp \left[ik_z \left(\boldsymbol{\tau} - \frac{m_e^* \rho}{m_e^* + m_h^*} \right) \right] (-i\hbar \nabla) w_v(\boldsymbol{\tau}), \quad (9)$$

$w_c(\boldsymbol{\tau})$ and $w_v(\boldsymbol{\tau})$ are, respectively, the Wannier functions for the conduction band and the valence band at site 0, and m_e^* and m_h^* are, respectively, the effective masses of the electron and hole. Hence the interaction between the exciton and the photon (in the resonance approximation) can be written in the form

$$H' = \sum_{k_z n} \sum_{\mathbf{q}'\lambda} D_{\mathbf{q}'k_z n} b_{k_z \mathbf{q}'\lambda} c_{k_z n}^\dagger + \text{h.c.}, \quad (10)$$

where

$$D_{\mathbf{q}'k_z n} = \frac{e}{mc} \sqrt{\frac{2\pi\hbar c}{(q'^2 + k_z^2)^{1/2} v}} \boldsymbol{\epsilon}_{\mathbf{q}'k_z\lambda} \cdot \mathbf{A}_{k_z n}. \quad (11)$$

Now, we assume that at time $t = 0$ one of the Wannier excitons is in the mode k_z, n . For time $t > 0$, the state $|\psi(t)\rangle$ can be written as

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

where $|k_z, n\rangle$ is the state with a Wannier exciton in the mode k_z, n in the linear chain, $|G; \mathbf{q}'k_z\rangle$ represents the state in which the electron-hole pair recombines and a photon in the mode \mathbf{q}', k_z

is created, and $f_0(t)$ and $f_{G;\mathbf{q}'k_z}(t)$ are, respectively, the probability amplitudes of the state $|k_z, n\rangle$ and $|G; \mathbf{q}'k_z\rangle$.

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

$$f_0(t) = \exp \left[-i\Omega_{k_z} t - \frac{1}{2}\gamma_{k_z} t \right], \quad (13)$$

where

$$\gamma_{k_z} = 2\pi \sum_{\mathbf{q}'\lambda} |D_{\mathbf{q}'k_z n}|^2 \delta(\omega_{\mathbf{q}'k_z n}) \quad (14)$$

and

$$\Omega_{k_z} = \mathcal{P} \sum_{\mathbf{q}'} \frac{|D_{\mathbf{q}'k_z n}|^2}{\omega_{\mathbf{q}'k_z n}}, \quad (15)$$

with $\omega_{\mathbf{q}'k_z n} = E_{k_z n}/\hbar - c\sqrt{q'^2 + k_z^2}$. Here γ_{k_z} and Ω_{k_z} are, respectively, the decay rate and frequency shift of the exciton, and \mathcal{P} means the principal value of the integral.

The Wannier exciton decay rate in the optical region be calculated straightforwardly and is given by

$$\gamma_{k_z} = \begin{cases} \frac{3\pi}{2k_0 d} \gamma_0 \sum_{\lambda} \frac{|\epsilon_{k_z \lambda} \cdot \chi_n|^2}{|\chi_n|^2}, & k_z < k_0, \\ 0, & \text{otherwise,} \end{cases} \quad (16)$$

where $k_0 = E_{k_z n}/\hbar$,

$$\chi_n = \sum_{\rho} F_n^*(\rho) \int d\tau w_c(\tau - \rho) (-i\hbar\nabla) w_v(\tau), \quad (17)$$

and

$$\gamma_0 = \frac{4e^2 \hbar k_0}{3m^2 c^2} |\chi_n|^2. \quad (18)$$

Here, χ_n^* represents the effective dipole matrix element for an electron jumping from the excited Wannier state in the conduction band back to the hole state in the valence band, and γ_0 is the decay rate of an isolated atom. We see from eq. (16) that γ_{k_z} is proportional to $1/(k_0 d)$. This is just the superradiance factor coming from the coherent contributions of atoms within half a wavelength or so [8, 18, 20].

Now let us consider a quasi-1D channel with channel width $L = Nd$. The state of the Wannier exciton can be specified as $|\mathbf{k}, n, l\rangle$, where $\mathbf{k} = (k_z, k_x)$ is the exciton wave number with k_x (k_z) normal (parallel) to the channel direction and n, l is the quantum number for the internal structure of the exciton. Here, k_x takes the value $k_x = 2\pi n_x / Nd$, with n_x an integer that is limited to one Brillouin zone ($n_x = 1, 2, \dots, N-1$) [14, 19]. The Wannier exciton state can be expressed as

$$|\mathbf{k}, n, l\rangle = \sum_{l, \rho, I, J} U_{\mathbf{k}nl}^*(l, \rho, I, J) |c, (l + \rho, I + J); v, (l, J)\rangle, \quad (19)$$

and the interaction matrix elements can be written as

$$\langle \mathbf{k}, n, l | H' | G \rangle = \sum_{l, \rho, I, J} \langle c, (l + \rho, I + J); v, (l, J) | U_{\mathbf{k}nl}^* (l, \rho, I, J) H' | G \rangle, \quad (20)$$

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

$$|c, (l + \rho, I + J); v, (l, J)\rangle = a_{c, (l + \rho, I + J)}^\dagger a_{v, (l, J)} | G \rangle, \quad (21)$$

where $a_{c, (l + \rho, I + J)}^\dagger$ ($a_{v, (l, J)}$) is the creation (destruction) operator for an electron in the conduction (valence) band at site $(l + \rho, I + J)$ ((l, J)), and l and ρ (I and J) are the vectors parallel (normal) to the channel direction. The expansion coefficient $U_{\mathbf{k}nl}^* (l, \rho, I, J)$ is the exciton wave function in the quantum channel:

$$U_{\mathbf{k}nl}^* (l, \rho, I, J) = \frac{1}{\sqrt{N'}} \frac{1}{\sqrt{N}} \exp [ik_z r_c + ik_x r'_c] F_{nl}(\rho, x), \quad (22)$$

where $r'_c = \frac{m_e^* (I + J) + m_h^* I}{m_e^* + m_h^*}$, r_c is the center of mass of the exciton in the channel direction, and $F_{nl}(\rho, x)$ is the hydrogenic wave function in the quantum channel. Following the derivation in the above, one can evaluate the decay rate straightforwardly:

$$\begin{aligned} \gamma_{\mathbf{k}nl} &= \frac{2\pi e^2 \hbar}{m^2 c v} |\epsilon_{\mathbf{k}\lambda} \cdot \mathbf{A}_{\mathbf{k}nl}|^2 \sum_{k'_y} \frac{1}{N} \frac{\sin^2(\pi n_y - N d k'_y / 2)}{\sin^2(\pi n_y / N - d k'_y / 2)} \times \\ &\times \left(\sum_{k'_x} \frac{\delta\left(\frac{E_{\mathbf{k}nl}}{\hbar} - c\sqrt{k_x'^2 + k_y'^2 + k_z^2}\right)}{\sqrt{k_x'^2 + k_y'^2 + k_z^2}} \right), \end{aligned} \quad (23)$$

where $E_{\mathbf{k}nl}$ is the exciton energy in the quasi-1D channel.

For convenience, the analysis of eq. (23) is addressed to the Frenkel exciton ($F_{nl}(0, 0)$ is equal to unity [10]). Generalization to the case of Wannier exciton is straightforward. In the $n_y = 0$ mode, there is an analytical solution for $N = 2$ and is given by

$$\gamma_{\mathbf{k}nl} \propto \frac{\gamma_0}{k_0 d} \left[1 + J_0 \left(d \sqrt{E_{\mathbf{k}nl}^2 / c^2 \hbar^2 - k_z^2} \right) \right], \quad (24)$$

where J_0 is the Bessel function of the zeroth order. For $N = 3$, there is also an analytical solution:

$$\gamma_{\mathbf{k}nl} \propto \frac{\gamma_0}{k_0 d} \left[3 + 2J_0 \left(2d \sqrt{E_{\mathbf{k}nl}^2 / c^2 \hbar^2 - k_z^2} \right) + 4J_0 \left(d \sqrt{E_{\mathbf{k}nl}^2 / c^2 \hbar^2 - k_z^2} \right) \right]. \quad (25)$$

As $N \rightarrow \infty$, the system becomes a crystal film, and the decay rate can be written as

$$\gamma_{\mathbf{k}nl} \propto \frac{\gamma_0}{(k_0 d)^2} \frac{1}{\sqrt{E_{\mathbf{k}nl}^2 / c^2 \hbar^2 - k_z^2}}, \quad (26)$$

where \mathbf{k} is the wave vector of the exciton in the crystal film. In fig. 1 we have plotted the decay rate as a function of N . In plotting the figure, we have assumed $k_0 = 2\pi/\lambda$, $\lambda = 8000 \text{ \AA}$, and lattice spacing $d = 5 \text{ \AA}$ in the numerical calculation. With the increasing of channel width, the decay rate shows an oscillatory behavior and approaches the 2D limit. The origin of the oscillation behavior can be seen more clearly by adding perfectly reflecting mirrors

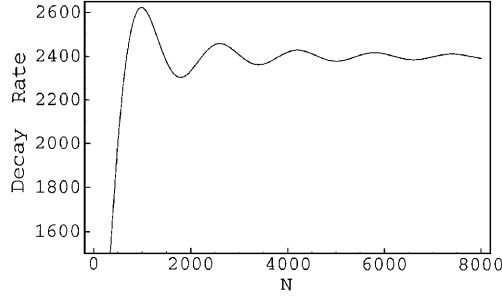


Fig. 1 – Decay rate of the superradiant exciton as a function of N (channel width $L = Nd$). The vertical and horizontal units are $\gamma_0/(k_0d)$ and N , respectively.

(microcavity with thickness L_c) above and below the quantum channel. If the mirror plane is parallel to the channel plane, it means the exciton can only couple to discrete photon modes ($k'_x = \frac{2\pi}{L_c}n_c$, where n_c is integer) in the perpendicular direction. Considering only the lowest mode $k'_x = \frac{2\pi}{L_c}$, the decay rate can be evaluated as

$$\gamma_{\mathbf{k}nl} = G_{Nk_z n_y} \frac{2\pi e^2 \hbar}{m^2 c^2 v} \frac{|\boldsymbol{\epsilon}_{\mathbf{k}\lambda} \cdot \mathbf{A}_{\mathbf{k}nl} t|^2}{\sqrt{k_0^2 - \left(\frac{2\pi}{L_c}\right)^2 - k_z^2}}, \quad (27)$$

where the oscillation factor

$$G_{Nk_z n_y} = \frac{1}{N} \frac{\sin^2\left(\pi n_y - \frac{Nd}{2} \sqrt{k_0^2 - \left(\frac{2\pi}{L_c}\right)^2 - k_z^2}\right)}{\sin^2\left(\pi n_y / N - \frac{d}{2} \sqrt{k_0^2 - \left(\frac{2\pi}{L_c}\right)^2 - k_z^2}\right)} \quad (28)$$

is similar to the quantum well result [14, 21] which comes from the interference between the radiation fields. As can be seen from eq. (27), the exciton modes with $k_0 < q = \sqrt{\left(\frac{2\pi}{L_c}\right)^2 + k_z^2}$ have a vanishing decay rate. These exciton modes do not radiate at all and photon trapping occurs. These dark modes also occur in a 2D thin film. However, it is hard to examine them directly because of the randomness of q in a thin film. With the recent developments of fabrication technology, it is now possible to fabricate the planar microcavities incorporating quantum wires [19]. If the thickness L_c is equal to the wavelength of the photon emitted by bare exciton (without external field), one can examine the dark mode directly by changing k_0 with external field.

One also notes that as the value of k_0 is equal to $\sqrt{\left(\frac{2\pi}{L_c}\right)^2 + k_z^2}$ (resonant mode), the decay rate goes to infinity. In the work of ref. [19], Constantin *et al.* investigated the transition from nonresonant mode to resonant coupling between quantum-confined one-dimensional carriers and two-dimensional photons states in a wavelength-long planar Bragg microcavity incorporating strained $\text{In}_{0.15}\text{Ga}_{0.85}/\text{GaAs}$ V-groove quantum wires. They found that when the excitonic transition energy is resonant with the cavity mode, the emission rate into this mode is significantly enhanced. This significant feature is just the singularity in eq. (27) and can be explained easily by the present model.

In summary, we have calculated the decay rate of the exciton in a quasi-1D channel with channel width L . For small channel width ($N = 1, 2$, and 3), analytical solutions can be

evaluated straightforwardly. Similar to the case of quantum well system, the decay rate of the exciton shows an oscillatory behavior with the increasing of channel width L . As $L \rightarrow \infty$, the decay rate approaches the 2D limit correctly. Second, when the quasi-1D channel is incorporated with planar microcavities, it becomes possible to examine the dark modes of the exciton. The distinguishing features are pointed out and may be observable in a suitably designed experiment.

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