Decay rate and renormalized frequency shift of a quantum wire Wannier exciton in a planar microcavity

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Super-radiant decay rate and frequency shift of a Wannier exciton in a one-dimensional quantum wire are studied. It is shown that the dark-mode exciton can be examined experimentally when the quantum wire is embedded in a planar microcavity. It is also found that the decay rate is greatly enhanced as the cavity length L_c is equal to the multiple wavelength of the emitted photon. Similar to its decay-rate counterpart, the frequency shift also shows discontinuities at resonant modes.

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Historically, the idea of super-radiance was introduced by Dicke.¹ Later, the coherent radiation phenomena for the atomic system was intensively investigated.²⁻⁶ One of the limiting cases of super-radiance is the exciton-polariton state in solid-state physics. But as it was well known in a three-dimensional (3D) bulk crystal,⁷ the excitons will couple with photons to form polaritons—the eigenstate of the combined system consisting of the crystal and the radiation field that does not decay radiatively. 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 chain⁸ and $(\lambda/d)^2$ for 2D exciton polariton,^{9,10} where λ is the wavelength of emitted photon and *d* is the lattice constant of the linear chain or the thin film.

First observation of super-radiant short lifetimes of excitons has been performed by Aaviksoo et al.11 on surface states of the anthracene crystal. Later, Deveaud et al.¹² measured the radiative lifetimes of free excitons in GaAs quantum wells and observed the enhanced radiative recombination of the excitons. Hanamura¹³ investigated theoretically the radiative decay rate of quantum-dot and quantum-well excitons. The results obtained by Hanamura are in agreement with that of Liu and Lee's (Ref. 10) prediction for thin films. Knoester¹⁴ obtained the dispersion relation of Frenkel excitons of quantum slab. An 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 super-radiance in thin and thick slab geometries. They demonstrated that super-radiance can be treated by a unified formalism for Frenkel excitons and Wannier excitons. In work of Agranovich et al.'s work¹⁶ a detailed microscopic study of Frenkel exciton-polariton in crystal slabs of arbitrary thickness was performed.

For lower dimensional systems, Ivanov and Haug¹⁷ predicted the existence of an exciton crystal, which favors coherent emission in the form of super-radiance in quantum wires. Manabe *et al.*¹⁸ considered the super-radiance 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 Although some of the theoretical papers discussed the exciton-polariton splitting of quantum wires embedded in a microcavity,²⁰ the spontaneous emission of the exciton as a function of cavity length has received no attention. In this paper, we will investigate the radiative decay of the Wannier exciton in one-dimensional quantum wires embedded in planar microcavities. It will be shown that some interesting quantities may be measured by making use of the properties of the microcavity.

For simplicity, let us first approximate the quantum wire as a linear chain with lattice spacing d in a free space. As it was well known, the Sommerfeld factor is smaller than unity in a one-dimensional system.²¹ The strong Coulomb interaction moves the oscillator strength out of the continuum states into the exciton resonance. Practically the entire oscillator strength is accumulated in the ground-state exciton. Thus, we can assume a two-band model for the band structure of the system safely as long as the thermal energy is smaller than the binding energy of the exciton. In this case, the state of the Wannier exciton can be specified as

$$|k_z,n\rangle = \sum_{l\rho} \frac{1}{\sqrt{N}} \exp(ik_z r_c) F_n(l), \qquad (1)$$

where the coefficient $1/\sqrt{N}$ is for the normalization of the state $|k_z, n\rangle$, k_z is the crystal momentum along the chain direction characterizing the motion of the exciton, n is the quantum number for the internal structure of the exciton, and, in the effective mass approximation, $r_c = [m_e^*(l+\rho) + m_h^*\rho]/(m_e^* + m_h^*)$ is the center of mass of the exciton. $F_n(l)$ is the hydrogenic wave function with $l+\rho$ and ρ being the positions of the electron and hole, respectively. Here, m_e^*

and m_h^* are the effective masses of the electron and hole, respectively. The Hamiltonian for the exciton is

$$H_{ex} = \sum_{k_{z}n} E_{k_{z}n} c_{k_{z}n}^{\dagger} c_{k_{z}n}, \qquad (2)$$

where $c_{k_z n}^{\mathsf{T}}$ and $c_{k_z n}$ are the creation and destruction operators of the exciton, respectively. $E_{k_z n}$ is the exciton dispersion.

The Hamiltonian of free photons is

$$H_{ph} = \sum_{\mathbf{q}'k_z'} \hbar c (q'^2 + k_z'^2)^{1/2} b_{\mathbf{q}'k_z'}^{\dagger} b_{\mathbf{q}'k_z'}, \qquad (3)$$

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

In the resonance approximation,¹⁰ the interaction between the exciton and the photon can be written in the form

$$H' = \sum_{k_{z}n} \sum_{\mathbf{q}'} D_{\mathbf{q}'k_{z}n} b_{k_{z}\mathbf{q}'} c_{k_{z}n}^{\dagger} + \text{H.c.}, \qquad (4)$$

where

$$D_{\mathbf{q}'k_{z}n} = \frac{e}{mc} \sqrt{\frac{2\pi\hbar cN}{(q'^{2} + k_{z}^{2})^{1/2}v}} \epsilon_{\mathbf{q}'k_{z}} \chi_{k_{z}n}$$
(5)

with $\epsilon_{\mathbf{q}'k_z}$ being the polarization of the photon. In Eq. (5),

$$\chi_{k_{z}n} = \sum_{l} F^{*}(l) \int d\tau \omega_{c}(\tau - l) \exp\left[ik_{z}\left(\tau - \frac{m_{e}^{*}}{m_{e}^{*} + m_{h}^{*}}l\right)\right] \times \left(-i\hbar\frac{\partial}{\partial\tau}\right) \omega_{v}(\tau)$$
(6)

is the effective transition dipole matrix element between the electronic Wannier state ω_c in the conduction band and the Wannier hole state ω_v in the valence band.

Now, we assume that at time t=0 the Wannier exciton is in the mode k_z, n . For time t>0, the state $|\psi(t)\rangle$ for the whole system composed of the exciton and photons can be written as

$$|\psi(t)\rangle = f_0(t)|k_z, n; 0\rangle + \sum_{\mathbf{q}'} f_{G;\mathbf{q}'k_z}(t)|G;\mathbf{q}'k_z\rangle, \quad (7)$$

where $|k_z,n;0\rangle$ is the state with a Wannier exciton in the mode k_z,n in the linear chain without photons, and $|G;\mathbf{q}'k_z\rangle$ represents the state in which the electron-hole pair recombines and a photon in the mode \mathbf{q}',k_z is created.

By the method of Heitler and Ma in the resonance approximation, the probability amplitude $f_0(t)$ can be expressed as¹⁰

$$f_0(t) = \exp\left(-i\Omega_{k_z n} t - \frac{1}{2}\gamma_{k_z n} t\right),\tag{8}$$

where

$$\gamma_{k_{z^n}} = 2\pi \sum_{\mathbf{q}'} |D_{\mathbf{q}'k_{z^n}}|^2 \delta(\omega_{\mathbf{q}'k_{z^n}}) \tag{9}$$

$$\Omega_{k_{z}n} = \mathcal{P} \sum_{\mathbf{q}'} \frac{|D_{\mathbf{q}'k_{z}n}|^2}{\omega_{\mathbf{q}'k_{z}n}}$$
(10)

with $\omega_{\mathbf{q}'k_{z}n} = E_{k_{z}n}/\hbar - c \sqrt{q'^2 + k_z^2}$. Here $\gamma_{k_{z}n}$ and $\Omega_{k_{z}n}$ 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 can be calculated straightforwardly and is given by

$$\gamma_{k_{z}n} = \begin{cases} \frac{3\pi}{2k_{0}d} \gamma_{0} \frac{|\boldsymbol{\epsilon}_{\mathbf{q}'k_{z}}\chi_{n}|^{2}}{|\chi_{n}|^{2}}, & \sqrt{q'^{2}+k_{z}^{2}} < k_{0} \\ 0, & \text{otherwise,} \end{cases}$$
(11)

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

$$\chi_n = \sum_l F_n^*(l) \int d\tau w_c(\tau - l) \left(-i\hbar \frac{\partial}{\partial \tau} \right) w_v(\tau), \quad (12)$$

and

$$\gamma_0 = \frac{4e^2\hbar k_0}{3m^2c^2} |\chi_n|^2.$$
(13)

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. (11) that $\gamma_{k_{z^n}}$ is proportional to $1/(k_0 d)$. This is just the super-radiance factor coming from the coherent contributions of atoms within half a wavelength or so.^{8,18}

Now let us consider the quantum wire embedded in perfectly reflecting mirrors with cavity length L_c . If the mirror plane is parallel to the quantum wire, it means the exciton can only couple to discrete photon modes $[(2\pi/L_c)n_c]$, where n_c are integers] in the perpendicular direction, while the photon modes are still continuous in the other direction. Following the above derivation, the decay rate can be evaluated as

$$\gamma_{k_{z}=0,n} = \frac{2\pi e^{2}\hbar}{m^{2}c^{2}d} \sum_{n_{c}=1}^{N_{c}} \frac{1}{L_{c}} \frac{|\epsilon_{\mathbf{q}'k_{z}}\chi_{n}|^{2}}{\sqrt{k_{0}^{2} - \left(\frac{2\pi}{L_{c}}n_{c}\right)^{2}}}, \quad (14)$$

where N_c is an integer. Because of the conservation of energy, the value of N_c must be smaller than L_c/λ , where λ is the wavelength of the emitted photon.

As can be seen from equation (14), the exciton modes with $k_0 < 2\pi/L_c$ have vanishing decay rate. These exciton modes do not radiate at all and photon trapping occurs.



FIG. 1. Decay rate of the superradiant exciton in a quantum wire embedded in cavities with leakages. The vertical and horizontal units are $2\pi e^2 \hbar |\boldsymbol{\epsilon}_{\mathbf{q}'k}\chi_n|^2/m^2 c^2 k_0 d$ and λ , respectively.

These *dark modes* also occur in a 2D thin film. However, it is hard to examine them directly because of the randomness of crystal momentum 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 might argue that the singularities in Eq. (14) are irrelevant in the weak-coupling region. However, there are always some leakages from the environment in realistic systems. In this case, the summation of the discrete modes in Eq. (14) becomes the integration of the continuous modes

$$\gamma_{k_z=0,n} = \frac{e^2 \hbar}{m^2 c^2 d} \int G(k_x) \frac{|\epsilon_{\mathbf{q}'k_z} \chi_n|^2}{\sqrt{k_0^2 - k_x^2}} dk_x, \qquad (15)$$

where the factor $G(k_x)$ contains the informations of the leakages. For good-reflecting mirrors, we further assume $G(k_x)$ has the form of Lorentzian distribution and can be expressed as

$$G(k_x) = N_G \sum_{n_c} \frac{1}{(k_x - 2\pi n_c/L_c)^2 + \Delta k_x^2},$$
 (16)

where N_G is the normalization constant and Δk_x is the line width. Figure 1 shows the numerical calculations of Eqs. (15) and (16) with Δk_x being equal to 1% of the fundamental mode $2\pi/L_c$. As can be seen in the figure, the singularities smear out because of the leakages. Therefore, as long as the corresponding band-gap frequency $E_{k,n}/\hbar$ is much larger than the decay rate at these peak values, the perturbation theory still works well. One also notes the peak value decreases with the increasing of cavity length. Therefore, the decay rate should approach to the free space limit in Eq. (11) as the cavity length becomes infinity. In the work of Ref. 19, Constantin *et al.* investigated the transition from nonresonant mode to resonant coupling between quantum-confined onedimensional carriers and two-dimensional photon states in a planar Bragg microcavity with a size of one wavelength incorporating strained In_{0.15}Ga_{0.85}/GaAs V-groove quantum wires. They found that when the excitonic transition energy is resonant with the cavity mode, the emission rate into this



FIG. 2. Renormalized frequency shift of the super-radiant exciton in a quantum wire as a function of the cavity length L_c . The vertical and horizontal units are $e^2\hbar |\boldsymbol{\epsilon}_{\mathbf{q}'k_z}\chi_n|^2/m^2c^2k_0d$ and λ , respectively.

mode is significantly enhanced. This significant feature is just the enhancement in Fig. 1 and can be easily explained by the present model.

To understand the emission rate increasing to resonance thoroughly, we now consider a Wannier exciton in a quantum ring embedded in perfectly reflecting mirrors with cavity length L_c . The circular ring is joined by the N_r lattice points with radius $\rho \sim N_r d/2\pi$, where d is the lattice spacing and the number of the lattice points is N_r . Following the above derivation, the decay rate of the quantum ring exciton can be expressed as

$$\gamma_{\nu} = \sum_{n_{c}} \frac{e^{2}\hbar}{m^{2}c^{2}L_{c}} \frac{\rho}{d} \\ \times |H_{\nu}^{(1)}(\sqrt{(2\pi/\lambda)^{2} - (2\pi n_{c}/L_{c})^{2}}\rho)|^{2} |\epsilon_{\mathbf{q}'k_{z}'} \cdot \boldsymbol{\chi}_{\nu}|^{2},$$
(17)

where ν is the exciton wave number in the circular direction and $H_{\nu}^{(1)}$ is the Hankel function. As can be seen from Eq. (17), the decay rate of a quantum ring exciton also shows the enhanced peaks as the cavity length L_c is equal to the multiple wavelength of the emitted photon. However, if one considers a Wannier exciton in a quantum dot embedded in the microcavity, the decay rate

$$\gamma \propto \sum_{n_c} \frac{e^2 \hbar}{m^2 c^2 L_c} \theta((2\pi/\lambda)^2 - (2\pi n_c/L_c)^2) |\boldsymbol{\epsilon}_{\mathbf{q}' k_z'} \boldsymbol{\chi}|^2,$$
(18)

where θ is the step function, shows no peak, instead, only the plateau appears with the increasing of the cavity length. This is because the angular momentum (translational momentum) of the exciton in a quantum ring (wire) is conserved in circular (chain) direction, while the crystal symmetry is totally broken in a quantum dot. Due to the modification of the density of states of the photon in the microcavity, the decay rate of the exciton shows peaks in a quasi-one-dimensional system but *plateaus* in a quasi-zero-dimensional system. One should notice that such kind of peak maybe a useful feature

to realize the Aharonov-Bohm (AB) effect for an exciton in a quantum ring. Recently, Römer and Raikh²² studied theoretically the exciton absorption on a ring threaded by a magnetic flux. In order to see the AB oscillations, they suggested to measure the luminescence. In this case, however, the excitonic AB oscillations is very small and hard to be measured. Therefore, if one can incorporate the quantum ring with planar microcavities, the AB oscillations will be enhanced at these peaks.

A few remarks about frequency shift $\Omega_{k_z^n}$ in a quantum wire can be mentioned here. In perfect microcavities, the frequency shift can be expressed as

$$\Omega_{k_{z}=0,n} = \frac{e^{2}\hbar}{m^{2}c^{2}d} \sum_{n_{c}=1}^{N_{c}} \frac{1}{L_{c}} \int \frac{|\epsilon_{\mathbf{q}'k_{z}}\chi_{n}|^{2}}{\left[k_{0} - \sqrt{k_{x}^{2} + \left(\frac{2\pi}{L_{c}}n_{c}\right)^{2}}\right] \sqrt{k_{x}^{2} + \left(\frac{2\pi}{L_{c}}n_{c}\right)^{2}}} dk_{x}.$$
(19)

As pointed out in Ref. 24, the frequency shift suffers from divergences and has to be removed by renormalization. Following the renormalization procedure proposed by Lee *et al.*,²⁴ one can, in principle, obtain the frequency shift in Eq. (19). In the free-space limit, the frequency shift can be approximated as

$$\Omega_{k_z=0,n}^{ren} \sim -\gamma_{\text{single}} \left(\frac{1}{k_0 d}\right),\tag{20}$$

where γ_{single} is roughly equal to the decay rate of an isolated atom.²⁴ Similar to the decay rate in Eq. (11), the frequency shift is also super-radiatively enhanced by the coherent effect. The numerical calculations of Eq. (19) are shown in Fig. 2. As can be seen from the figure, the frequency shift has discontinuities when the cavity length is equal to the multiple wavelength of the emitted photon. This is because whenever the cavity length exceeds some multiple wavelength, it opens up another decay channel abruptly. These discontinuities should also smear out because of the leakages from the environment. One can also note that as the cavity length increases, the sign of the frequency shift changes from positive to negative and approaches the free-space limit. This kind of crossing also occurs in the quantum-well systems,²³ and can be realized by the competition between the negative and positive values of the integration in Eq. (19).

For usual semiconductors, the enhanced factor in Eq. (20) is about 10³ for Wannier excitons in the optical range. However, due to the extreme smallness of γ_{single} itself, observation of $\Omega_{k_z \sim 0,n}^{ren}$ is not expected to be easy. The discontinuous behavior at certain cavity length L_c may be a useful feature to observe this quantity. Besides, we suggest to investigate the semiconductor materials that have a larger excitonoscillator strength and thus a larger frequency shift. As for the magnitude of this shift, if the decay rate of the exciton is in the order of per picosecond, the radiative shift is about 10^{-1} meV. One can also vary the number N_0 of the wires. Due to the coherent effects, the measured frequency shift would be $N_0 \Omega_{k_z \sim 0,n}^{ren}$ if the wires are placed within the coherent length. However, one should also note the inhomogeneous broadening caused by the fluctuations in the wire sizes that cannot be kept absolutely constant, leading to the small band-gap fluctuation of quantum wires. The inhomogeneous broadening may be taken into account by assuming that the wires have a size distribution given by $f(E_{k_z})$ around a mean value $\overline{E_{k_z}}$. The average frequency shift is then $\overline{\Omega_{k_z}^{ren}}$ $= \int f(E_{k_z}) \Omega_{k_z \sim 0,n}^{ren} |_{E_{k_z}} dE_{k_z}$.

In summary, we have calculated the decay rate of the Wannier exciton in a quantum wire. When the quantum wire is incorporated into planar microcavities, it becomes possible to examine the dark modes of the exciton. Besides, the decay rate is greatly enhanced as the cavity length L_c is equal to the multiple wavelength of the emitted photon. The singularities in the decay rate smear out as a result of leakages from the environment. Furthermore, we have also calculated the renormalized frequency shift of the exciton. Similar to its decay rate counterpart, the frequency shift shows discontinuity at resonant modes. The distinguishing features are pointed out and may be observable in a suitably designed experiment.

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