# Spontaneous resonance between bound and delocalized excitons caused by enhanced radiative corrections

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Light-induced coupling between bound and delocalized excitons in a single semiconductor thin film has been investigated. A planar defect with a negative $\delta$-function-type potential acts as a potential well for the center-of-mass motion of an exciton. In the nano-to-bulk size regime, enhanced radiative corrections of particular delocalized states attain the binding energy of the bound state, which leads to unconventional resonant states of the excitonic system. We demonstrate that such radiative couplings and their quantum interference enhance light absorption and asymmetric optical spectra around the bound level.

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## I. INTRODUCTION

A bound exciton in a semiconductor, a localized state of an exciton trapped mainly by impurities or defects, has been a ubiquitous subject since early times in material science, photophysics, and application for optoelectronic devices, and it continues to be attracting researchers. Recent developments in experiments to overcome the unintentional dopants and defects are opening up veiled potentials of this system such as low-threshold lasers [1] and single-photon sources [2,3] utilizing bound exciton transitions. Therefore, controlling individual bound states in high-quality samples has been receiving much attention in the past decade [4].

In contrast, optical responses of excitons in high-quality samples are dominated by delocalized excitons. Particularly in the nano-to-bulk size regime, a large interaction volume between a delocalized state and a radiation field leads to an increase in a level shift and a spectral broadening (radiative corrections [5–9]). Importantly, not only light-exciton coupling but also exciton-exciton coupling via radiation is enhanced [10,11]. We expect such coupling to occur between bound and delocalized states as well. If this is true, the bound exciton transition may be enhanced through quantum interference of the coupled states reflecting their greatly different spectral widths. This is a good analogy of the energy concentration of light into localized nano-objects positioned nearby metallic nanostructures [12–15], though the physical origin of the spectral broadening is totally different.

Although numerous studies have been performed on the optical properties of bound excitons [16–23], the correlation between bound and delocalized excitons has been sparsely discussed so far. In many cases, they have been regarded as independent because of the large energy separation and the orthogonality of their wave functions. However, in the nano-to-bulk size regime, we believe that such assumptions are no longer guaranteed for the above reasons. In this paper, we propose a scheme to enhance the bound exciton transition with special attention paid to radiation-induced coupling with delocalized states. Bound excitons in ZnO, III-V semiconductors such as GaAs and GaN have been extensively studied, and also the planer defects and $\delta$-doping layer are known as origins of bound excitons, exhibiting large coupling with light [24–27]. Thus, as a possible system, we consider a thin film with a planar defect that acts as a potential well for the translational motion of excitons and provides a bound state. With regard to material parameters, we employ those of GaN for numerical demonstrations. Although GaN-based optoelectronic devices have already been commercialized, defect engineering is required to maximize their potential performances [28–30].

As a result, we find that the large radiative shift of a particular delocalized exciton attains the binding energy of the bound exciton, which causes a spontaneous resonance. Because of the resonant coupling, radiation is partially transferred from the delocalized state to the bound state, resulting in radiative broadening. Furthermore, accompanying quantum interference between the coupled states leads to an enhancement of the bound exciton absorption. We also demonstrate that these anomalous behaviors are reflected in coherent optical signals as an asymmetric spectrum.

The rest of this paper is organized as follows: Section II explains the calculation model of the excitonic system and nonlocal response theory to clarify the radiative coupling of bound and delocalized exciton states. In Sec. III, we show results of an eigenmode analysis and investigate some anomalous behaviors of these coupled states focusing on quantum interference. Our results and a discussion are summarized in Sec. IV.

## II. THEORETICAL FRAMEWORK

### A. Model of the excitonic system

We consider a thin film structure with a thickness much larger than the excitonic Bohr radius and that is periodic along the film surface. In this condition, the relative motion of an exciton can be treated in the same way as those in the bulk, although the center-of-mass (c. m.) motion is confined in the thickness ($z$) direction. A planer defect positioned at $z = l$ acts as a potential well for the excitonic c. m. motions. The Hamiltonian of the excitonic system in the $z$ direction can be

 

written as [31]

$$H_{\rm ex} = -\frac{\hbar^2}{2M}\frac{\partial^2}{\partial z^2} - Va\delta(z-l), \tag{1}$$

where $M$ is the effective mass of the exciton and $a$ is a lattice constant. As a planar defect, we assume a $\delta$-function-type potential with a strength $V$ ($>0$). The eigenenergies $\epsilon_k$ and eigenfunctions $\phi_k(z)$ of the kinetic term are already known. By using a complete set $\{\phi_k(z)\}$, the eigenfunction of $H_{\rm ex}$ can be expanded as $\psi_\kappa(z) = \sum_k C_{\kappa k}\,\phi_k(z)$. The eigenvalue equation $H_{\rm ex}\psi_\kappa(z) = E_\kappa\psi_\kappa(z)$ has a nontrivial solution under the following relation:

$$-\frac{1}{Va} = \sum_k \frac{|\phi_k(l)|^2}{E_\kappa - \epsilon_k}. \tag{2}$$

By applying $\phi_k(z) = \sqrt{\frac{1}{L}}e^{ikz}$ and $\epsilon_k = \hbar^2k^2/(2M)$, the binding energy of a bound exciton at the bulk limit is obtained as $\Delta_b = -E_{\rm bound} = M(Va)^2/(2\hbar^2)$.

For a thin film with thickness $d$, we take the boundary condition $\phi_k(z) = 0$ at $z = 0$ and $d$. Then the eigenfunction of $H_{\rm ex}$ is obtained as

$$\psi_\lambda(z) = N_\lambda\{\cos\kappa_\lambda(d-l-z) - \cos\kappa_\lambda(d-|l-z|)\}, \tag{3}$$

where $\lambda$ is an index to label every quantized exciton state, $N_\lambda$ is a normalization constant, and $\kappa_\lambda$ is a quantized wave number that satisfies $E_\lambda = \hbar^2\kappa_\lambda^2/(2M)$. A bound state can exist under the following condition:

$$Va > \frac{\hbar^2}{2M}\frac{1}{l(1-l/d)}. \tag{4}$$

Figure 1 shows wave functions of (a) the bound state with $\Delta_b = 7.1$ meV and (b) some delocalized states. The thickness $d$ is 452 nm and the defect is positioned at the center of the sample. The bound state is strongly localized around the defect, in contrast to the delocalized states, although their spatial structures are also modulated near the defect.

In Ref. [31], various multiple scattering behaviors of exciton polaritons have been investigated in a system where the planar defect acts as a positive potential wall for excitons. In this paper, we pay attention to the bound state and clarify the radiative coupling with other delocalized exciton states. Although the defect position $l$ affects the excitonic wave functions and their interference structures in the optical spectra, it does not play an essential role in the radiative coupling. To avoid confusing discussions of $l$ dependences, we use a fixed $l = d/2$ throughout the paper.

### B. Radiative coupling of exciton states

To describe the self-consistent interplay between the spatial structures of radiation fields and excitonic waves, we apply nonlocal response theory [32,33]. According to linear response theory, the first-order polarization in the site representation is written as

$$\mathcal{P}(z,\omega) = \int \chi(z,z',\omega)\,\mathcal{E}(z',\omega)dz'. \tag{5}$$

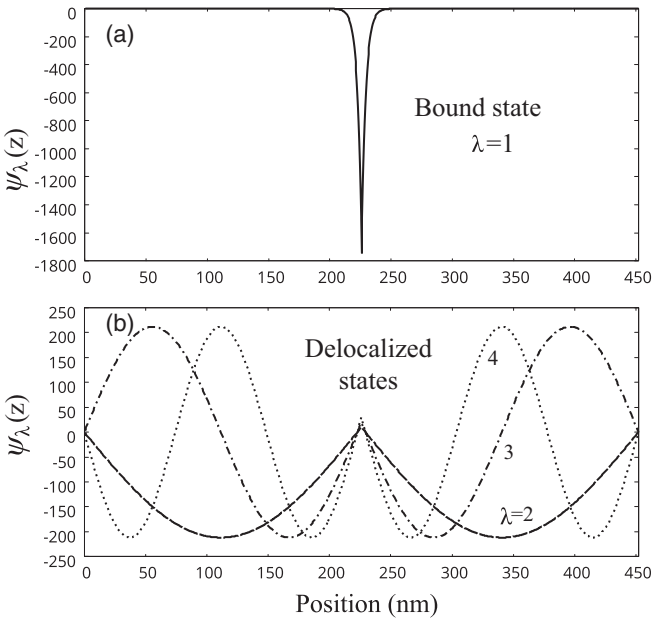


FIG. 1. Excitonic wave functions of (a) a bound state with the binding energy $\Delta_b = 7.1$ meV, and (b) some delocalized states. The lowest energy state is the bound state $\lambda = 1$, followed in order by $\lambda = 2$ (dashed line), $\lambda = 3$ (the dashed dotted), and $\lambda = 4$ (the dotted line). The thickness is 452 nm and the defect is positioned at the center of the sample.

In this expression, the resonant term of the nonlocal susceptibility is written as

$$\chi(z,z',\omega) = \sum_\lambda \frac{|\mu_\lambda|^2\psi_\lambda(z)\psi_\lambda^*(z')}{E_T + E_\lambda - \hbar\omega - i\Gamma}, \tag{6}$$

where $E_T$ is the transverse exciton energy, and $\Gamma$ is the nonradiative damping constant. Practically, $\Gamma$ for the bound state is different from that for delocalized states. In this paper, however, we use the same value to clarify the role of the radiative widths, not the nonradiative ones. $|\mu_\lambda|$ is the coupling strength between light and excitons at the bulk limit. For delocalized excitons ($\lambda \geqslant 2$), $|\mu_\lambda|$ relates to the longitudinal-transverse (LT) splitting energy $\Delta_{LT}$ as $|\mu_{\lambda\geqslant 2}|^2 = \epsilon_b\Delta_{LT}/(4\pi)$, where $\epsilon_b$ is the background dielectric constant of the material. In contrast, $|\mu_1|$ is determined from the radiative lifetime of the bound exciton, as explained in the last paragraph of this section. For GaN, nearly degenerate excitonic bands (called A, B, and C excitons) appear in the optical spectra [19,34,35]. In the present demonstration, however, we focus on the single (A) band exciton, avoiding nonessential contributions of the multiband excitons. Elaborate analysis considering the radiative coupling between multiband excitons [11] is necessary for evaluating the absolute value of the optical signal, although we do not consider it in the present study.

If we assume the normal incidence for simplicity, the polarization field from the resonant contribution should be determined self-consistently from the following Maxwell equation [11]:

$$\mathcal{E}(z,\omega) = \mathcal{E}^{(0)}(z,\omega) + 4\pi\left(\frac{\omega}{c}\right)^2\int \mathcal{G}(z,z',\omega)\,\mathcal{P}(z',\omega)\,dz', \tag{7}$$

where $\mathcal{E}^{(0)}(z,\omega)$ is the background electric field and $c$ is the velocity of light in vacuum. $\mathcal{G}(z,z',\omega)$ is the retarded Green's function of the Maxwell equation for a thin film structure [36]. By considering Eqs. (5), (6), and (7), the following closed linear equation set is obtained:

$$\boldsymbol{S}(\omega)\boldsymbol{X}(\omega) = \boldsymbol{F}^{(0)}(\omega), \tag{8}$$

where $F_\lambda^{(0)}(\omega) = |\mu_\lambda| \int \psi_\lambda^*(z)\, \mathcal{E}^{(0)}(z,\omega)\, dz$ indicates the interaction between an exciton and the background electric field and

$$X_\lambda(\omega) = \frac{|\mu_\lambda|}{E_T + E_\lambda - \hbar\omega - i\Gamma_\lambda} \int \psi_\lambda^*(z)\, \mathcal{E}(z,\omega)\, dz \tag{9}$$

indicates the polarization amplitude of each exciton component. The coefficient matrix $\boldsymbol{S}(\omega)$ is written as

$$\boldsymbol{S}(\omega) = (E_T - E_\lambda - \hbar\omega - i\Gamma_\lambda)\boldsymbol{I} + \boldsymbol{Z}(\omega), \tag{10}$$

where $\boldsymbol{I}$ is a unit matrix. The radiative correction matrix $\boldsymbol{Z}(\omega)$ is obtained as

$$Z_{\lambda'\lambda}(\omega) = -4\pi \left(\frac{\omega}{c}\right)^2 |\mu_{\lambda'}\mu_\lambda| \times \iint \psi_{\lambda'}^*(z)\mathcal{G}(z,z',\omega)\psi_\lambda(z')\, dz dz', \tag{11}$$

which indicates the coupling between respective exciton components via radiation. A bound and a delocalized exciton are radiatively coupled through this term. The roots of $\det|\boldsymbol{S}(\omega)| = 0$ provide the eigenmodes of the exciton-radiation-coupled system [6]. The real part $\mathrm{Re}[\hbar\omega_\xi]$ gives the eigenenergy including the radiative shift from the bare exciton energy, and the imaginary part $-\mathrm{Im}[\hbar\omega_\xi]$ gives the radiative width, where $\xi$ is the index of the quantized exciton-radiation-coupled states. Maxwell's boundary conditions for the electromagnetic field at the surfaces provide the reflectance $R(\omega)$ and the transmittance $T(\omega)$, simultaneously [32]. The total absorption of the system is obtained from $A(\omega) = 1 - R(\omega) - T(\omega)$.

The value of $|\mu_1|$ can be determined from the radiative lifetime of the bound state at the bulk limit through the following procedure: From Eq. (11), the self-interaction of the bound state under the long-wavelength approximation is obtained as $Z_{11}^{\mathrm{LWA}}(\omega) = -4\pi(\frac{\omega}{c})^2|\mu_1|^2\mathcal{G}(l,l,\omega)|\int \psi_1(z)dz|^2$. By using the Green's function and the wave function for the bulk, the radiative lifetime of the bound state, $\tau_{\mathrm{bulk}}$, is approximately obtained as $\tau_{\mathrm{bulk}} \sim \frac{\hbar}{-2\mathrm{Im}[Z_{11}^{\mathrm{LWA}}(E_T-\Delta_b/\hbar)]} = \frac{\sqrt{\epsilon_b}McVa}{16\pi|\mu_1|^2(E_T-\Delta_b)}$.

## III. RESULTS AND DISCUSSION

We use material parameters of bulk GaN [35]: $E_T = 3.4791$ eV, $\Delta_{LT} = 1.34$ meV, $\epsilon_b = 8.6$, and $M = 0.5m_0$ where $m_0$ is a static electron mass. In addition, we use the radiative lifetime of the bound state as $\tau_{\mathrm{bulk}} = 130$ ps and the binding energy as $\Delta_b = 7.1$ meV, which have been estimated from the photoluminescence of the neutral donor-bound exciton in GaN [18].

Figure 2 shows the thickness-dependent energy structure of (a) bare exciton states and (b) exciton-radiation coupled states measured from $E_T$. The bound level is less dependent on thickness larger than 20 nm, while from Eq. (4) it disappears for very small thicknesses less than 5 nm though it is not shown in the figure. Compared with the bound state, delocalized states receive much greater radiative corrections, reflecting their large interaction volume and spatial phase relation with a radiation field [6,11]. In particular, the states with $\xi \geqslant 3$ are redshifted in turn with increase in the thickness, which induces a spontaneous resonance between the bound and delocalized states with very small anticrossing properties. For the states with $\xi = 5$ and 6, the radiative shifts reach and exceed the binding energy $\Delta_b$ at particular thicknesses. In these thicknesses, the spatial structures between light and the particular exciton state (e.g., $\lambda = 6$ state at 452 nm) are well matched, activating quantum interference of the bound and delocalized exciton states. In contrast, the exciton states with $\lambda > 6$ are spatially mismatched to the radiation field. This is why the resonant coupling between bound and higher-order states is relatively small. However, for thicker samples, we can find the optimum conditions where the higher-order states ($\lambda > 6$) are spatially matched to the light waves.

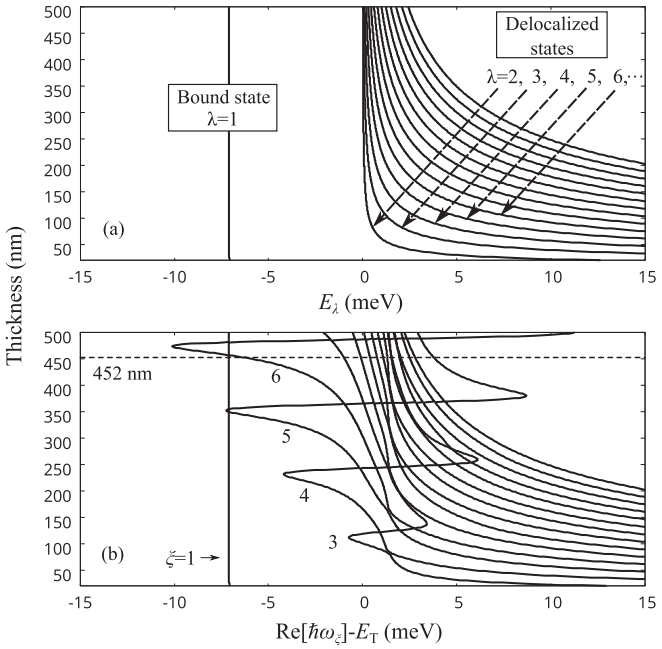


FIG. 2. Thickness-dependent energy structure of (a) bare exciton states $E_\lambda$ and (b) exciton-radiation coupled states measured from $E_T$.

The radiative width of the bound state ($\xi = 1$) tends to be enhanced at around the energetically resonant thicknesses as shown in Fig. 3(a) (where the local maximal values appear at slightly lesser thicknesses than the resonant ones). At 452 nm, the value takes a local maximum of 7.22 $\mu$eV. However, for such thicknesses, delocalized states exhibit relatively small radiative widths, although the values are still far greater than those of the bound state, as shown in Fig. 3(b) (where, at 452 nm, the value for $\xi = 6$ is 2.47 meV, which is a factor of ~342 greater that for $\xi = 1$). Therefore, we expect that the coupled states with such different types of spectral structures change the optical response around the bound state through their quantum interference depending on the thickness.

To evaluate the energy concentration of light, we define the bound-state absorption $\Delta A(\omega)$ as

$$\Delta A(\omega) = A(\omega) - A_{|\mu_1|=0}(\omega), \tag{12}$$

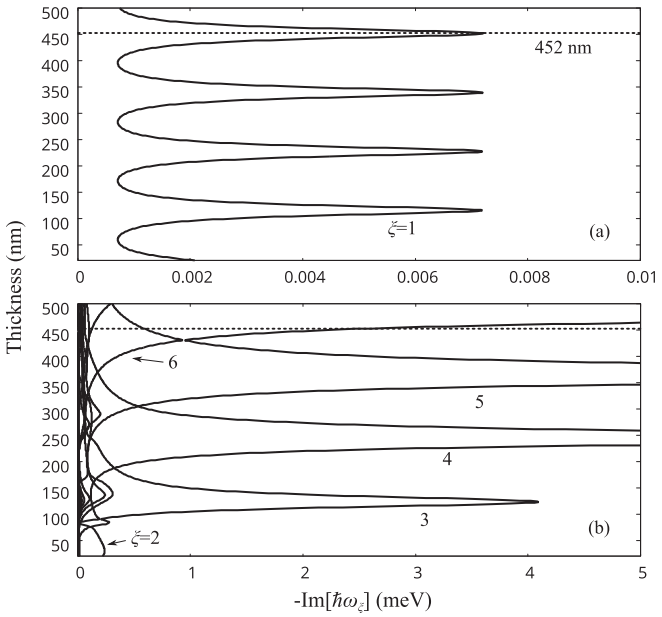


FIG. 3. Thickness-dependent radiative widths of exciton-radiation coupled states with (a) an index $\xi = 1$ (bound state) and (b) indices $\xi \geqslant 2$ (delocalized states).

where $A_{|\mu_1|=0}(\omega)$ is the total absorption without the bound exciton transition, i.e., $|\mu_1| = 0$. [Note that $A_{|\mu_1|=0}(\omega)$ is not zero even around the bound level because there are small absorptions by delocalized states.] By comparing this value with the absorption without the delocalized exciton transitions, the presented resonant effects on the absorption spectra can be clarified. Figure 4 shows the thickness dependence of $\Delta A(\omega)$ for $\Gamma = 0.4$ meV. The dotted lines indicate the total absorptions without delocalized exciton transitions, i.e., $|\mu_{\lambda\geqslant 2}| = 0$ for respective thicknesses. Enhancement of light absorption around the bound level is obvious because spectral broadening and peak increase occur simultaneously. Furthermore, the amount of increase with increasing thickness is much greater compared with that without delocalized exciton transitions (dotted lines). The spectral broadening is partially explained by the enlarged radiative width shown in Fig. 3(a). However, such drastic enhancement cannot be fully explained only from the viewpoint of the radiative width, particularly in terms of the peak increase.

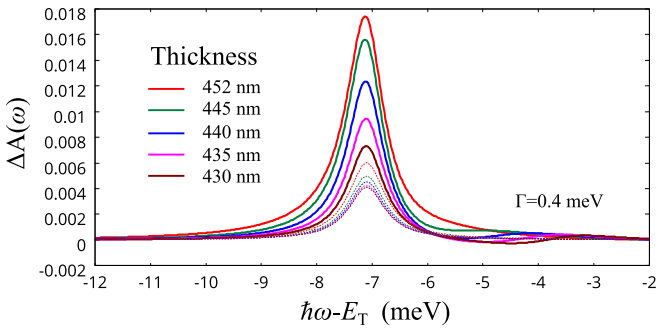


FIG. 4. Thickness dependence of a bound exciton absorption $\Delta A(\omega)$ for $\Gamma = 0.4$ meV. The dotted lines indicate a total absorption without delocalized exciton transitions, i.e., $|\mu_{\lambda\geqslant 2}| = 0$ for respective thicknesses.

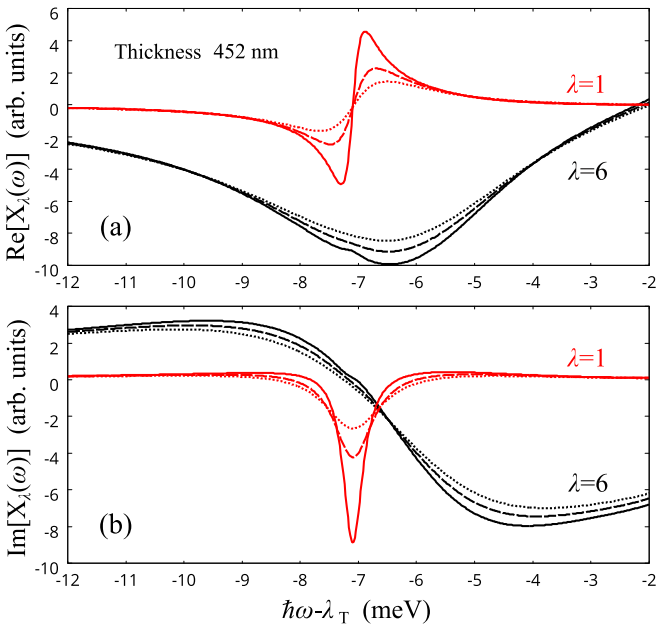


FIG. 5. Γ dependence of (a) real parts and (b) imaginary parts of $X_1(\omega)$ and $X_6(\omega)$ in a 268-nm-thick film. The solid, dashed, and dotted lines indicate $\Gamma = 0.2$, 0.4, and 0.6 meV, respectively.

To reveal the mechanism of the absorption enhancement, we investigated the polarization amplitude of each exciton component $X_\lambda(\omega)$. At 452 nm, the states with $\lambda = 1$ and $\lambda = 6$ are resonantly coupled via radiation, as shown in Fig. 2(b). The Maxwell electric field is described by a superposition of every excitonic polarization component. Therefore, optical responses around the bound level are considered to reflect quantum interference between $\lambda = 1$ and $\lambda = 6$ states. Figure 5 shows the Γ dependence of (a) real parts and (b) imaginary parts of $X_1(\omega)$ and $X_6(\omega)$. The characteristic structure of $X_1(\omega)$ with its narrow spectral width is totally covered by the broad spectral width of $X_6(\omega)$, which comes from the large radiative corrections. Importantly, the polarization amplitude for $\lambda = 6$ is slightly suppressed at around the bound level with the decrease in Γ, while that for $\lambda = 1$ becomes more pronounced. This indicates that radiation dominated by the delocalized state ($\lambda = 6$) is partially transferred to the bound state ($\lambda = 1$). Such a type of interaction relates to a Fano resonance [37,38], where discrete levels coupled to continuous bands exhibit asymmetric structures in the spectra. The absorption enhancement as demonstrated in Fig. 4 is analogous to the energy concentration of light into localized nano-objects positioned near metallic nanostructures [12,13,15] derived from Fano-type quantum interference. The presented mechanism is physically different from such systems because the spectral broadening and coupling enhancement are not nonradiative processes but purely radiative ones.

The drastic enhancement of the bound exciton transition through the unique quantum interference with the delocalized states is expected to even change the coherent optical signals. In a similar manner to Eq. (12), we define changes in reflectance and transmittance by the bound exciton transition as

$$\Delta R(\omega) = R(\omega) - R_{|\mu_1|=0}(\omega) \quad (13)$$

$$\Delta T(\omega) = T(\omega) - T_{|\mu_1|=0}(\omega), \quad (14)$$

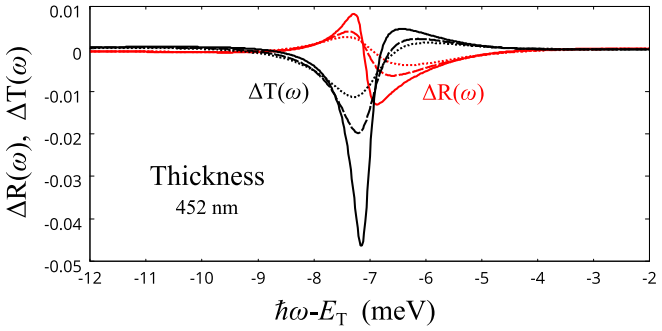


FIG. 6. Γ dependence of the changes in reflectance $\Delta R(\omega)$, and transmittance $\Delta T(\omega)$, by the bound exciton transition for 268-nm thickness. The solid, dashed, and dotted lines indicate $\Gamma = 0.2$, 0.4, and 0.6 meV, respectively.

where $R_{|\mu_1|=0}(\omega)$ and $T_{|\mu_1|=0}(\omega)$ are the reflectance and transmittance without bound exciton transition, respectively.

As shown in Fig. 6, they exhibit asymmetric spectral shapes, reflecting the superposition of $X_1(\omega)$ and $X_6(\omega)$ dominantly. By tuning the incident photon energy around the bound level, the signs of $\Delta R(\omega)$ and $\Delta T(\omega)$ are switchable. Also, the behavior can be considered as indirect evidence for the existence of the enhanced radiative corrections for delocalized states as shown in Fig. 2(b), although the asymmetric structures appear slightly even when only the bound state is excited because of interference with the background electric field.

## IV. CONCLUSION

By paying special attention to the radiative coupling between bound and delocalized states, we theoretically investigated optical responses of excitonic system in the semiconductor GaN with a planar defect. Enhanced radiative corrections attain the binding energy $\Delta_b$, which causes a spontaneous resonance between bound and particular delocalized states via radiation. The radiative widths of the bound state are enhanced around the resonant thicknesses owing to the transfer of radiation from the delocalized state. The drastic enhancement of the bound exciton absorption with the thickness can be explained by the enlarged radiative widths and anomalous quantum interference between resonant states. They are reflected even in the coherent optical signals as the asymmetric structures in the spectra, which can also be considered as indirect evidence to show the greatly enhanced radiative corrections for delocalized states. The presented results exhibit a striking contrast to conventional understandings of the optical response of bound excitons, in which they exist independently from delocalized states because of the large energy separation and the orthogonality of their wave functions.

The long range spatial coherence of delocalized excitons has already been realized for high-quality semiconductors whose thicknesses are several hundreds of nanometers [5,8,9]. With regard to the bound states, note that the planer structure is not essential for the proposed effect, but it would be possible to observe the similar effect in the case of other types of bound states. However, if the delta-doping technique developed in the growth technology of III-V semiconductors can be applied to the materials exhibiting the large radiative shift of excitons, it will be a good approach to experimentally verify the proposed effect with intentionally controlled spatial distributions of the bound states.

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