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Excitonic polaritons in one-dimensional photonic crystals

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Nonlocal investigations are presented for the exciton-photon coupling in one-dimensional periodic alternating layered structures consisting of two kinds of slabs (photonic crystals), in which excitons are present only in one of the two slabs. The lower branch of the excitonic polariton for this system is found to split into many small bands separated by small band gaps. This phenomenon is explained as the band splitting caused by the coherent interference of polaritonic waves in the periodic systems. The present nonlocal study also demonstrates a double exciton-photon coupling, in which the upper branch of the polariton couples again with the size-quantized exciton states. [S0163-1829(98)50904-6]

Currently, there is a strong interest in the optical properties of semiconductor nanostructures.[1] Most of the optical processes in these structures have so far been treated as a process that could occur in an isolated quantum structure (e.g., a single quantum well). The optical response of the whole system is therefore obtained by simply summing up the responses of the individual structures. In contrast to this, some new aspects may be anticipated for the optical responses of quantum structures when these structures are not isolated anymore and therefore some couplings are presumed between them. Several features arising from this cooperativity have been reported elsewhere: superradiant emission[2] and optical linear and nonlinear responses.[3] In this context, it possibly brings a departure in optical materials science and optical devices to answer the question as to what could be the optical processes in periodically arranged quantum structures. The coherent interference of the photon in ordered[4] (photonic crystals) and disordered[5] systems appears to give an impetus to the above study from a slightly different point of view. In this paper, we investigate using the nonlocal exciton theory the exciton-photon coupling (polariton effects) in one-dimensional photonic crystals. The intention of this investigation is to isolate the kinetics of excitonic polaritons that may be exhibited in the whole system of regularly arranged quantum structures.

The model of the periodic system we employ here is an alternating layered structure that consists of CuCl and NaCl slabs. Let the number of the layers be infinite, its period l, and let the CuCl slab thickness be d. We focus on the photon energy near the exciton resonance of CuCl. The excitons that might be created in NaCl slabs can therefore be neglected because of the pronounced energy separation. Namely, it is sufficient to consider that excitons are present only in CuCl slabs. Here, we consider CuCl slabs so thick that the scheme of the exciton center-of-mass quantization holds. Unnecessarily thick slabs, however, make the exciton levels unseparable. Hence, in this study, we employ the slabs with an appropriate thickness (see the next paragraph). This structure requires us to treat the exciton in it with the nonlocal theory because of the spatial dispersion of the exciton. When electromagnetic waves propagate in the direction (z axis) perpendicular to the layers, the electric field $E_x(z)$ has to satisfy the following Maxwell equation with the exciton polarization:

$$\frac{d^2E_x}{dz^2}+\left(\frac{\omega}{c}\right)^2\left(\varepsilon(z)E_x(z)+4\pi S\int dz'\chi(z,z')E_x(z')\right)=0, \tag{1}$$

where the integral in this equation is the nonlocal exciton polarization [denoted by $P_x(z)$]. Here, $\chi(z,z')$ is the nonlocal polarizability of the exciton, S is the area of the region we are considering, and $\varepsilon(z)$ is the dielectric constant that is a periodic function of z with the same period as that of the structure. Ordinary notations are used for other parameters. Here, we assume for simplicity that no direct interactions are present between excitons in different slabs. This implies that the nonlocality of the exciton works within each slab but it does not extend to other slabs. In the light of this assumption, we require the nonlocal polarizability $\chi(z,z')$ to have the periodicity as follows. First, $\chi(z,z')$ must have a finite value when both z and z' are in the identical CuCl slab, while it must vanish for any other combinations of z and z' positions. Second, once $\chi(z,z')$ is defined in a slab, then $\chi(z,z')$ in other slabs must have the same form as previously defined. The nonlocal polarizability thus defined in a slab has the form

$$\chi(z,z')=2\left(\frac{e}{m_0\omega}\right)^2|\boldsymbol{p}_{\rm vc}|^2|\phi_{1s}(0)|^2S^{-1}\times\sum_{n=0}^{\infty}\frac{\varphi_n(z)\varphi_n(z')^*}{\hbar\omega_n-\hbar\omega-i\gamma}, \tag{2}$$

where only the resonant term is taken.[6] Here, $\phi_{1s}(0)$ is the $1s$ exciton wave-function value of the relative motion at the origin, $\boldsymbol{p}_{\rm vc}$ is the momentum matrix element of the optical transition between the conduction and the valence bands, and γ is the damping factor. For a hard wall, the wave function of the exciton center-of-mass motion $\varphi_n(z)$ must be a sinusoidal function and therefore the nth exciton energy level is given by $\hbar\omega_n=\hbar\omega_{1s}+(\hbar^2/2m)[\pi(n+1)/d]^2$ for $n=0,1,2,\ldots$, where ω_{1s} is the exciton resonance and m is the exciton mass. Equation (1) is an integrodifferential equation with periodically varying coefficients. The ordinary procedure for an equation like this is to solve it using the Bloch theorem. To the author's knowledge, however, it does not always seem general that the integrodifferential equation has a solution of the Bloch type. The group-theoretic approach to the integrodifferential operator can verify it to be true indeed provided that the kernel (with two variables) possesses some kind of periodicity. Then, to confirm it in a practical problem, let us tentatively assume a solution of the Bloch type,

$$E_x(z)=e^{iKz}\sum_G F_x(G)e^{iGz}, \tag{3}$$

where $F_x(z)$ is a periodic function with the period of l and it is expanded into a Fourier series (G is the reciprocal lattice). The wave number K thus introduced describes the motion throughout the whole periodic structure but it is not implied for that in one slab. Substituting Eq. (3) into $P_x(z)$ and using the periodicity of $\chi(z,z')$ defined before, we find after a careful check that the exciton polarization $P_x(z)$ also has the form of the Bloch function. This result appears to be physically correct and reassures the existence of the Bloch-type solution for the kernel having the periodicity we imposed on the system. The next step of calculation is the same as that for the photonic crystals,[4] although it is more complicated in the present case because of the exciton term. Substituting Eq. (3) into Eq. (1), we obtain a matrix for determining the eigenvalue ω for an arbitrary wave number K. This matrix is a function of ω and K and hence the eigenvalue problem cannot be solved using the standard method we find in a computer library, except for a simple case.[7] Since the point is to get nontrivial solutions, then we determined the eigenvalues by searching the zeros of the determinant for the relevant matrix.

In the numerical calculation, we employed the periodic structure in which $l=813$ and $d=731$ Å. These lengths were chosen for the following reasons. First, we required the exciton resonance to be located in the lowest photonic band, i.e., the lowest band obtained by neglecting the exciton term in Eq. (1). Second, in order to manifest the nonlocal effects of the exciton in the slab, the slab thickness d must be of the same order as or larger than the wavelength λ; namely, λ/d should be small. The structural parameters satisfying the two above requirements simultaneously are restricted within some combinations of λ and d. For the λ and d values thus chosen, the λ/d value is estimated at 2.42. Other parameters used are as follows:[8] the mass and the resonance of a CuCl exciton are $m=2.5m_0$ and $\hbar\omega_{1s}=3.2025$ eV, respectively, the dielectric constants are 5.00 for CuCl and 2.46 for NaCl, and the exciton-photon coupling constant is $\Delta_{\mathrm{LT}}=5.5$ meV. We set the damping factor γ as 0 in the practical calculations.

Prior to showing the results of the exciton-photon coupling, let us ascertain where the exciton resonance is located in the photonic band for the structure mentioned in the preceding paragraph. The inset of Fig. 1 shows the photonic bands (solid lines) in the first Brillouin zone (BZ) obtained by neglecting the exciton effects, together with the exciton resonance location (dotted line). In all figures of this paper, the ω and K values are normalized in the unit of $2\pi c/l$ and $2\pi/l$, respectively. The bulk exciton resonance ω_{1s} for CuCl is located near the top of the first band. The wavelength at the point at which the resonance crosses the photonic band is estimated at $\lambda\sim1770$ Å.

The energy dispersion of the polariton in the above periodic structure is shown in Fig. 1 for the first BZ. Since we are interested in the energy region near the exciton resonance, the ordinate of the upper figure of Fig. 1 is magnified in the vicinity of the resonance. The lower figure is, on the contrary, scaled down to cover a wider energy range than in the upper one and the two are joined together continuously.

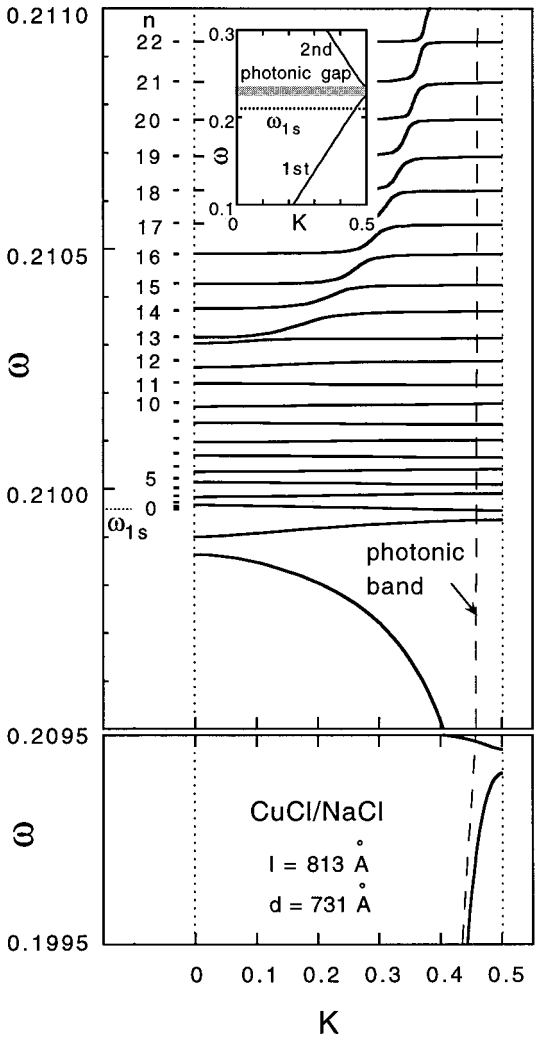

FIG. 1. Polariton dispersion of the periodic structure. Locations of size-quantized exciton levels are indicated by bars together with their index n. The inset shows the dispersion of the photonic crystal (i.e., neglecting the exciton effects) and the location of the exciton resonance ω_{1s}. In all figures of this paper, ω and K are normalized by $2\pi c/l$ and $2\pi/l$, respectively.

The steep broken line is the photonic band neglecting exciton effects, which is identical to the first band in the inset. In the presence of exciton-photon coupling, the energy dispersion is found to consist of many small bands that are separated by small band gaps. Note that these bands are dispersive (i.e., having finite group velocities). In the higher-energy region, we observe the phenomena of the dispersion curves anticrossing, i.e., the two curves approaching and then moving apart due to the repulsion between them. The band energy values in the higher-energy region coincide well with the size-quantized exciton levels, the positions of which are indicated by bars in Fig. 1 together with their index n. This coincidence, however, is gradually declining with the decrease in the energy. In the lower-energy region, there is evidently no correspondence between them.

In order to make clear what has happened to excitonic polaritons in periodic structures, we expanded the polariton dispersion near the resonance into the extended K zone up to the eighth BZ [see Fig. 2(a)]. The upper figure of Fig. 2(a) is again more magnified than the lower one. The mutual arrangement of the small bands described by solid lines in this

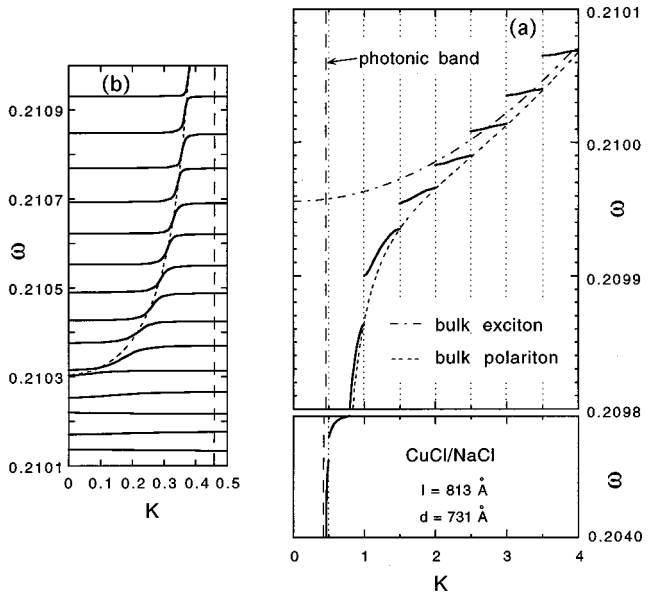

FIG. 2. Polariton dispersion replotted from Fig. 1 and magnified in the vicinity of the exciton resonance. (a) Dispersion expanded into the extended K zone at lower energies and (b) dispersion redrawn in the first Brillouin zone at higher energies. Bulk polariton dispersions using the renormalized exciton mass are drawn by broken lines in both (a) and (b).

figure reminds us of the splitting of the electronic bands in what we call semiconductor superlattices.[9] In light of the above, an attempt was carried out as what follows in order to account for the phenomenon of small bands formation in Fig. 2(a). The polariton dispersion in the bulk material is ordinarily obtained by using the photonic and the excitonic dispersions. While it is natural to use the photonic band (dashed line) neglecting exciton effects as the photonic dispersion, the excitonic band in the bulk material does not seem to be suited for our purpose: this curve shows a too-small spatial dispersion. Accordingly, in order to take the lifting of exciton levels by the size quantization in the slab into account, we employed the excitonic dispersion (dash-dotted line) using the exciton mass renormalized by a factor of $(d/l)^2$. The lower branch of the polariton dispersion thus obtained is drawn by a broken line in Fig. 2(a). We find that this curve exactly reproduces the energy values of polariton bands at the edge of every BZ. This result is similar to what occurs in the electronic bands of semiconductor superlattices. These small bands are thus found to be interpreted as the bands which the lower branch of bulk polariton dispersion has split into by the coherent interference of the polaritonic waves in the periodic systems.

Now that the lower branch of the polariton is obtained in the form mentioned above, its counterpart, i.e., the upper branch, must also show up in the energy dispersion. This can be discovered in the higher-energy region in Fig. 1, which is replotted as Fig. 2(b). The broken line in Fig. 2(b) represents the upper polariton branch calculated by the same procedure as that for the lower branch. This line is shown to exactly reproduce the line for the anticrossing phenomena. This implies that the anticrossing phenomena are caused by the coupling of the size-quantized exciton states with the upper branch of the bulk polariton. Since the upper branch is already a result of the exciton-photon coupling, the phenomena can be regarded as the product of the sequentially occurring double exciton-photon coupling. The behavior of the polariton dispersion shown in Fig. 2 entirely results from the excitonic nonlocality as well as the structural periodicity. If the exciton in the slab were treated as an assembly of local oscillators with the resonance energies of size-quantized states, the photon would couple with each oscillator separately; the resulting polariton dispersion would be merely the sum of the independent polariton dispersions for every oscillator.

Possible coupling schemes in the present system may be grouped into the following three classes: (i) (photon+exciton)+periodicity, (ii) (exciton+periodicity) +photon, and (iii) (periodicity+photon)+exciton, where $(a+b)+c$ implies that a and b couple first followed by the coupling with c. This classification is valid when the coupling of one pair $(a\text{-}b)$ is much stronger than the other two (b-c and c-a). Among these coupling schemes, (iii) may be ruled out for the following reason. Scheme (iii) requires us to first build the photonic band and next couple it with an exciton. We studied the polariton dispersion in the periodic structure for which the exciton resonance was located in the middle of the photonic band gap. Despite the fact that there was not a photon (i.e., the counterpart in the coupling) near the exciton resonance, we obtained a polariton dispersion similar to that in Fig. 1 in this energy region. The coupling scheme must hence be different from (iii). The interpretations for Figs. 1 and 2 already mentioned in the preceding paragraphs are entirely based on scheme (i) and seem to have been successful. As mentioned, the bulk polariton is first created as a result of a strong coupling between a photon and an exciton and next its dispersion splits into bands by the subsequent coupling with the periodicity. This is exactly an analogue of the miniband formation in semiconductor superlattices.[9] What differs between the two? Needless to say, one treats electrons while another treats excitons. In superlattices, the phenomenon is caused by the electronic tunneling that occurs through thin barriers with finite potential heights. This tunneling could be regarded as *glue* that connects electronic states in different sites. Since we consider a hard wall, the tunneling of excitons never occurs. The *glue* in our case is undoubtedly the photon. The phenomenon may be intuitively explained as proceeding as follows: the photon, which when coupled with an exciton in a slab, modifies its field by this coupling; it propagates to the neighboring slab; it couples again with an exciton in this slab, and the process repeats itself. The exciton in a slab thus couples with the exciton in a different slab via the photon. The situation may be made clearer if we begin with darkness (very low optical density). In darkness, coupling scheme (ii) holds. The exciton shows constant discrete energy levels (instead of bands) at any K value. Therefore, the group velocity is zero and the exciton is localized in each slab. When the light is switched on and gradually increases its strength, scheme (ii) does not hold anymore and scheme (i) takes over control of the coupling instead and finally produces the energy dispersions shown before. The final states thus attained indicate that the exciton component localized in each slab in darkness could be construed as being delocalized with the assistance of the photon.

In conclusion, we have investigated, using the nonlocal

exciton theory, the kinetics of excitonic polaritons, which is exhibited in the whole system of regularly arranged quantum slabs. The energy dispersion of polaritons in this structure displays many small bands produced by the coherent interference of the polaritonic waves in the periodic systems. The upper branch of the polariton couples with the size-quantized exciton states, which can be regarded as a double exciton-photon coupling.

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