

## Exciton dispersion in a structure of $N$ coupled multiple quantum wells

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We derive analytic expressions for the exciton dispersion in  $N$  coupled quantum wells. The Förster-type interaction is taken as a mechanism of interwell coupling. The explicit  $N$  dependence is obtained, which shows that for any  $N$  the dispersion is continuous in all directions and the isolated quantum-well value is reached in the limit of  $k_{\parallel}, q_{\nu} \rightarrow 0$  as is physically expected.

Multiple-quantum-well (MQW) engineering has recently reached a high level of progress (see, for example, Refs. 1–3). This allows one to test the many properties of MQW structures, which may be exploited for future high-performance devices, because the physics associated with MQW's is still far from a complete understanding. In particular, how the different QW's are coupled to each other and what the consequences are of such couplings prove to be topics for discussions. Besides tunneling mechanism in MQW's with narrow barriers, a surge of research in the so-called Förster-type (or dipole-dipole) coupling between QW's separated by a wide barrier has arisen quite recently.<sup>4–9</sup>

In this Comment we wish to readdress an issue that has been dealt with in Ref. 4. The motivation is that the coupled exciton-photon mode splitting of MQW's in a semiconductor quantum microcavity has been observed.<sup>10–12</sup> The experiments indicate that the splitting tends to saturate when  $N$  (the number of QW's) exceeds a certain value. To explain this feature one has to know, among other things, the  $N$  dependence of the exciton energy in an  $N$  coupled QW structure. Our purpose is twofold: (i) to derive from first principles the explicit dependence on the number  $N$  of QW's which would be further applicable to finite-size MQW structures placed inside a quantum microcavity<sup>10–12</sup> and (ii) to check the validity of the result reported in Ref. 4. The authors of Ref. 4 considered a MQW structure of infinite size (i.e.,  $N = \infty$ ). With the Förster-type interwell coupling taken into account, it was shown in Ref. 4 that the eigenenergy of the MQW structure has an unusual behavior near  $k_{\parallel}, q_{\nu} = 0$  ( $\vec{k}_{\parallel}$  is the in-plane wave vector while  $q_{\nu}$  is that along the growth direction). The exciton dispersion suffers a direction dependence near  $k_{\parallel}, q_{\nu} = 0$ , namely,

$$\lim_{q_{\nu} \rightarrow 0} \left( \lim_{k_{\parallel} \rightarrow 0} \mathcal{E}(q_{\nu}, k_{\parallel}) \right) = E(0), \quad (1)$$

while

$$\lim_{k_{\parallel} \rightarrow 0} \left( \lim_{q_{\nu} \rightarrow 0} \mathcal{E}(q_{\nu}, k_{\parallel}) \right) = E(0) + \frac{\text{const}}{d}, \quad (2)$$

where  $E$  is the exciton energy of a separate QW,  $\mathcal{E}$  is that of the whole coupled MQW structure, and  $d = l + b$  is the period of the structure with  $l$  ( $b$ ) being the well (barrier)

width. According to Eqs. (1) and (2) the energy  $\mathcal{E}$  approaches two distinct values when the wave vector tends to zero and, as the authors of Ref. 4 noticed, it changes continuously in the neighborhood. As a consequence of that behavior, one expects an intrinsic additional source to the linewidth:<sup>4</sup>

$$\Gamma \propto \frac{1}{l^3} + \frac{b}{d} + \dots \quad (3)$$

The second term on the right-hand side of Eq. (3) appears due to the splitting in Eqs. (1) and (2) and was estimated to be of the order of meV for GaAs-Al<sub>x</sub>Ga<sub>1-x</sub>As MQW's. The above finding seems to be of a fundamental interest so that it is desirable to verify to what extent it is valid. In practice, the number of QW's is always finite, typically<sup>10</sup>  $N = 30 - 100$  in usual devices or even much less for a quantum microcavity configuration. We will show in this comment that for any  $N$  no such splitting as found in Ref. 4 occurs.

We start with a Hamiltonian describing a structure of  $N$  QW's coupled by an effective dipole-dipole interwell term  $D_{|n-n'|}$  as follows (tunneling is neglected):

$$H^{(N)} = \sum_{n=1}^N \left[ E(k_{\parallel}) a_{n\vec{k}_{\parallel}}^{\dagger} a_{n\vec{k}_{\parallel}} + \sum_{n'=1, n' \neq n}^N D_{|n-n'|}(k_{\parallel}) a_{n'\vec{k}_{\parallel}}^{\dagger} a_{n\vec{k}_{\parallel}} \right], \quad (4)$$

where  $a_{n\vec{k}_{\parallel}}$  denotes the annihilation operator of an exciton in well  $n$  and with in-plane wave vector  $\vec{k}_{\parallel}$  and  $D_{|n-n'|}$  has the form<sup>4,6,9</sup>

$$D_{|n-n'|}(k_{\parallel}) = \alpha k_{\parallel} e^{-k_{\parallel}|n-n'|d} \quad (5)$$

with  $\alpha = 4\pi\mu^2|\Phi_{2D}(0)|^2/\epsilon$ ,  $\mu$  the interband dipole moment,  $\epsilon$  the medium dielectric constant, and  $\Phi_{2D}(0)$  the real-space two-dimensional exciton envelope function taken at the origin  $\vec{r} = 0$ .

Due to interwell coupling, the exciton can hop from well to well and the state of the whole coupled structure differs from that of  $N$  isolated QW's. In general, the overall state vector is constructed by linearly superposing exciton states of individual QW's. If there are only a few QW's, the eigenvector of  $H^{(N)}$  is searched for in the form

$$\Psi_{\nu \vec{k}_{\parallel}}^{(N)} = \sum_{n=1}^N u_{\nu n}(k_{\parallel}) a_{n \vec{k}_{\parallel}}^{\dagger} |0\rangle \quad (6)$$

with  $\nu = 1, 2, \dots, N$  and  $u_{\nu n}(k_{\parallel})$  the transformation coefficients. Use of Eq. (6) diagonalizes Eq. (4) and yields an  $N$ -order equation for  $N$  new eigenenergies. This kind of work has been done, e.g., in Refs. 6 and 9 for a coupled double QW structure, i.e., for  $N = 2$ . Here, to get closest to Ref. 4, we assume  $N$  to be large enough to impose a cyclic boundary condition on the system state vector. Then, making use of the periodicity of the structure, the eigenvector of  $H^{(N)}$  can be built as

$$\Psi_{q_{\nu} \vec{k}_{\parallel}}^{(N)} = \frac{1}{\sqrt{N}} \sum_{n=1}^N e^{iq_{\nu} n d} a_{n \vec{k}_{\parallel}}^{\dagger} |0\rangle, \quad (7)$$

where the  $N$  independent discrete values of  $q_{\nu}$  are determined by the cyclic boundary condition, which implies

$$q_{\nu} = \frac{2\pi\nu}{Nd}, \quad \nu = 0, 1, 2, \dots, (N-1). \quad (8)$$

The eigenenergy of  $H^{(N)}$  is then found by solving the equation

$$H^{(N)} \Psi_{q_{\nu} \vec{k}_{\parallel}}^{(N)} = \mathcal{E}^{(N)}(q_{\nu}, k_{\parallel}) \Psi_{q_{\nu} \vec{k}_{\parallel}}^{(N)}. \quad (9)$$

Putting Eqs. (4) and (6) into Eq. (9) and using the commutation relation  $[a_{n k_{\parallel}}, a_{n' k_{\parallel}}^{\dagger}] = \delta_{nn'}$  and that  $a_{n \vec{k}_{\parallel}} |0\rangle = 0$ , we readily get

$$\mathcal{E}^{(N)}(q_{\nu}, k_{\parallel}) = E(k_{\parallel}) + \sum_{\rho \neq 0} D_{|\rho|}(k_{\parallel}) e^{iq_{\nu} \rho d}. \quad (10)$$

Note that here  $\rho = n - n'$  measures the distance between well  $n$  and well  $n'$ . The sum over  $\rho$  in Eq. (10) is understood explicitly as

$$\sum_{\rho \neq 0} f_{\rho} = \sum_{\rho=1}^{(N-1)} (f_{\rho} + f_{-\rho}), \quad (11)$$

where  $f_{\rho}$  is an arbitrary function of  $\rho$ . The final result of our calculations is as follows:

$$\mathcal{E}^{(N)}(q_{\nu}, k_{\parallel}) = E(k_{\parallel}) + \alpha F^{(N)}(q_{\nu}, k_{\parallel}), \quad (12)$$

where

$$F^{(N)}(q_{\nu}, k_{\parallel}) = \frac{\cos(q_{\nu} d) - e^{-k_{\parallel} d} - e^{-k_{\parallel} d(N-1)} [1 - e^{-k_{\parallel} d} \cos(q_{\nu} d)]}{\cosh(k_{\parallel} d) - \cos(q_{\nu} d)} k_{\parallel}. \quad (13)$$

Our result Eq. (13) has two merits. The first merit is that it is valid for any  $N$  [of course, as we have assumed  $N$  large enough to use Eqs. (7) and (8) so that here, by any  $N$  we mean any large  $N$ , say,  $N \geq 10$ ]. In particular, it formally recovers the result of Ref. 4 when  $N$  tends to infinity. In this case, as it should be expected, Eq. (13) becomes coincident with Eq. (12) in Ref. 4 [note the misprint there:  $\cos(Kd)$  should be replaced by  $\cosh(Kd)$ ], i.e.,

$$F^{(\infty)}(q_{\nu}, k_{\parallel}) = \left[ \frac{\cos(q_{\nu} d) - e^{-k_{\parallel} d}}{\cosh(k_{\parallel} d) - \cos(q_{\nu} d)} \right] k_{\parallel}. \quad (14)$$

Yet, as will be seen shortly, this expression for  $N \rightarrow \infty$  is of use only as long as the limit of  $(k_{\parallel}, q_{\nu}) \rightarrow 0$  is not taken. The second merit of Eq. (13) concerns the limit of  $(k_{\parallel}, q_{\nu}) \rightarrow 0$ . Such a limit is of crucial importance in semiconductor physics since optical phenomena most likely occur near the origin in reciprocal space. Let us do this limit by writing explicitly the leading orders of the  $N$  unrelated parts but keeping the parts containing the  $N$  dependence as they are:

$$\begin{aligned} \lim_{q_{\nu} \rightarrow 0} \left( \lim_{k_{\parallel} \rightarrow 0} F^{(N)}(q_{\nu}, k_{\parallel}) \right) &= \lim_{k_{\parallel} \rightarrow 0} \left( \lim_{q_{\nu} \rightarrow 0} F^{(N)}(q_{\nu}, k_{\parallel}) \right) \\ &= \lim_{(q_{\nu}, k_{\parallel}) \rightarrow 0} \left\{ \frac{(1 - 1 + k_{\parallel} d) [1 - e^{-k_{\parallel} d(N-1)}]}{1 + \frac{(k_{\parallel} d)^2}{2} - 1 + \frac{(q_{\nu} d)^2}{2}} \right\} k_{\parallel} = 0. \end{aligned} \quad (15)$$

From Eq. (15) it is transparent that, for large but finite  $N$ , zero is obtained, independent of the order of carrying out the  $q_{\nu}$  limit and the  $k_{\parallel}$  limit. As for the formal limit of  $N \rightarrow \infty$  in Eq. (15), we encounter, from a mathematical point of view, an uncertainty in the product  $Nk_{\parallel}$  when at the same time  $k_{\parallel} \rightarrow 0$  and  $N \rightarrow \infty$ . *Mathematically*, we can suppose a situation when  $N$  tends to infinity more strongly than  $k_{\parallel}$  tends to zero, mak-

ing  $Nk_{\parallel}$  to be infinite. Such an imaginary circumstance gives the same result as in Ref. 4. Nevertheless, the above thing is unrealizable. *Physically*,  $k_{\parallel} \rightarrow 0$  is realized by decreasing the incidence angle  $\varphi$  of the pumping laser beam that excites the QW exciton. If the pumping field is directed to be perpendicular to the QW plane ( $\varphi = 0$ ), then  $k_{\parallel} = 0$ . On the other hand, the number  $N$  of QW's is restricted by the growth technique: it may

be as large as  $100$ ,<sup>10</sup> but it cannot be infinite. Based on this physical fact, the formal  $\lim_{(N \rightarrow \infty, k_{\parallel} \rightarrow 0)}$  in Eq. (15) is understood as the product  $Nk_{\parallel}$  tends to zero. In this sense Eq. (15) is applicable to any  $N$ , both finite and (formally) infinite. The point that makes these limits zero is the content in the square brackets in Eq. (15), which comes from the explicit accounting for  $N$ . If the formal limit of  $N \rightarrow \infty$  is made first in Eq. (13), i.e., if Eq. (14) is used, these square brackets would disappear and the  $\lim_{q_{\nu} \rightarrow 0} [\lim_{k_{\parallel} \rightarrow 0} F^{(\infty)}(q_{\nu}, k_{\parallel})]$  would vanish, whereas the  $\lim_{k_{\parallel} \rightarrow 0} [\lim_{q_{\nu} \rightarrow 0} F^{(\infty)}(q_{\nu}, k_{\parallel})]$  would give  $2/d$ , as obtained in Ref. 4. The resulting splitting is just a seeming fact caused by formal mathematics. This can be justified with some more physics as follows. The concrete interwell coupling considered here, i.e., the dipole-dipole coupling, does not contribute for the case  $k_{\parallel} = 0$  [see Eq. (5)]. When  $k_{\parallel} = 0$  the interwell connection is switched off, the QW's are decoupled, and thus, the outcome of the limit of  $k_{\parallel} \rightarrow 0$ , *whether it is taken before or after the limit*  $q_{\nu} \rightarrow 0$ , must correspond to the isolated QW's, i.e.,  $\mathcal{E}$  must tend to  $E$  ( $F^{(N)}$  must tend to zeros). Our strategy is therefore to use Eq. (13) in order to get the right physical limit when going from a coupled to an uncoupled regime. In the case of very large  $N$ , the formal limit  $N \rightarrow \infty$  is made at the end of the calculation. At this point it is worth mentioning that in the physics of semiconductor quantum microcavities,

which has just recently been developed (see, e.g., Refs. 10–12), the optical properties are sensitive to  $N$ . Hence knowledge of the explicit  $N$  dependence like Eq. (13) is highly necessary.

In conclusion, we have reconsidered the same problem of Ref. 4 but with emphasis paid to the explicit dependence on the number  $N$  of the QW's. Sometimes, if  $N$  is very large, the formal limit  $N \rightarrow \infty$  may not bring about qualitative changes and is convenient for theoretical calculations. However, sometimes *a priori* setting  $N = \infty$  at the very outset may lead to a seeming fact, as is the case considered here. We have proved that the direction-dependent behavior of the exciton dispersion in a coupled MQW structure discovered in Ref. 4 originates from treating infinite  $N$  at the outset, but not from the long-range character of the dipole-dipole interwell coupling as it was mentioned in Ref. 4. The dipole-dipole coupling, on the contrary, due to its  $k_{\parallel}$  dependence, requires  $\lim_{(k_{\parallel}, q_{\nu}) \rightarrow 0} \mathcal{E} = \lim_{(q_{\nu}, k_{\parallel}) \rightarrow 0} \mathcal{E} = E$ .

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