Double quantum well in a semiconductor microcavity: Three-oscillator model and ultrafast radiative decay

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A complete study of excitons in double quantum wells embedded in a semiconductor microcavity is presented. In the strong coupling regime we predict the possibility of observing three peaks in optical spectra when the two quantum well excitons have different resonance energies, and discuss the best structure parameters for observing this phenomenon. In the weak coupling regime we show that a radiative lifetime as short as ≈ 100 fs can be obtained by taking advantage of the coherence between the wells in addition to the microcavity effect. This ultrashort radiative lifetime is shown to be observable even in the presence of disorder.

Coherent optical phenomena in semiconductors are currently of great interest. In particular, tailoring of the excitonic radiative dynamics in semiconductor heterostructures is being intensively studied on both theoretical[1–4] and experimental[5–8] sides. In bulk semiconductors, due to crystal momentum conservation, stationary exciton-polariton states are formed:[9,10] thus the excitonic radiative lifetime depends on the dimensions of the sample and/or on the presence of scattering processes and is usually in the ns range.[11–15] In low-dimensional systems like quantum wells (QW's), on the other hand, an exciton with a given in-plane wave vector has an intrinsically short radiative lifetime due to the lack of wave-vector conservation along the growth direction.[1,16,17] Such a lifetime is of the order of 10 ps.[1,2] Very short lifetimes are indeed observed in QW systems under resonant excitation,[5,7,8] although the simple "polariton" picture based on wave-vector conservation has to be modified to include the effect of disorder.[2,3,7]

The radiative properties of the exciton can be substantially different when the single QW (SQW) is embedded in a microcavity (MC).[6,18] In the strong-coupling regime[19] Rabi oscillations occur between exciton and photon modes (which correspond to the formation of quasistationary cavity polaritons). In the weak-coupling regime, an irreversible process takes place and the excitonlike solution can exhibit an enhanced or deenhanced radiative decay. The problem of a QW exciton in a MC has been recently investigated in Ref. 20 and a two-oscillator model was found to hold for the coupled exciton-cavity system in the whole range of mirror reflectivities.

A MC is not the only way to have enhanced radiative emission. As shown by Citrin,[21] electromagnetic (e.m.) coupling between two identical QW's in free space can modify the radiative decay and can lead to a lifetime that is half the lifetime of an isolated QW. A further enhancement of the radiative emission can be achieved in a multiple-quantum-well (MQW) system, in which there is a particular state that is coherently coupled with light and that decays with an enhanced ("superradiant") decay rate, while all the other states are "subradiant" or dark.[22,23] The minimum lifetime that can be obtained in a MQW is about 1/6 of that of a SQW (Ref. 23) and is achieved when the MQW thickness is of the order of the wavelength of light (as the MQW thickness goes to infinity, stationary superlattice polaritons are recovered[24]). The coupled emission of excitons in MQW's is a phase-coherent phenomenon, which can be washed out by dephasing processes. In particular vertical disorder (i.e., the effect of having slightly different resonance energies for the excitons in the different QW's) is likely to destroy the effect in real structures. We should also mention the related work on Bragg MQW's.[25]

The results summarized above concerning a SQW embedded in a MC on one side, and a MQW in free space on the other, suggest the possibility of further enhancing the superradiant behavior by placing several identical QW's in a MC. This problem has been already partially investigated in Refs. 20 and 26. In those works, however, e.m. coupling within the MQW was not fully taken into account (i.e., multiple interference effects between the light reflected within the MQW were neglected).

Here we develop a complete study of a double QW (DQW) in a MC, which takes fully into account polariton effects between the wells; we give analytical expressions both in strong- and weak-coupling regimes and further investigate the effect of vertical disorder on the emission properties, due for example to well-width fluctuations.

Our approach is based on the semiclassical theory of the exciton-cavity system and a transfer matrix (TM) formulation is adopted. Each layer in the structure is characterized by a local, frequency-independent dielectric constant with the only exception of the QW's, which are described by a nonlocal susceptibility. The formalism is described in Refs. 20 and 24. For simplicity we consider the case of normal incidence.

We have considered a Fabry-Pérot MC with dielectric mirrors, called distributed Bragg reflectors (DBR's), with a DQW placed at the center. For frequencies close to the center of the stop band (ω_m) an approximate, parametric form of the reflection coefficient $r(\omega)$ of a DBR, which allows a realistic description of the frequency dependence of the phase, is[20,27]

$$r(\omega) = \sqrt{R}e^{i[\frac{n}{c}L_{\rm DBR}(\omega-\omega_m)+\varphi_m]}. \quad (1)$$

$L_{\rm DBR}$ represents a mirror penetration depth and is usually much larger than the cavity length. Expressions for R and $L_{\rm DBR}$ are given in Refs. 20 and 27.

Once the TM of the whole structure is known, the poles of the transmission coefficient (i.e., the zeros of the element T_{22} of the TM) give the dispersion as well as the radiative lifetime of the coupled exciton-radiation modes. In this way we find two decoupled solutions, such as for the DQW in free space, for which decoupled symmetric and antisymmetric solutions are found[21]. If we call symmetric (antisymmetric) the solution that tends to the symmetric (antisymmetric) one in the limit $r \to 0$ and indicate it by S (A) we have

$$\left(\frac{\Gamma_0}{\Delta}\right)_S = -\frac{1}{2}\frac{ir^2e^{2ikL_C}e^{-ikl/2} - 2re^{ikL_C}\sin(kl/2) - ie^{ikl/2}}{\cos(kl/2)(r^2e^{2ikL_C}e^{-ikl} + 2re^{ikL_C} + e^{ikl})}, \quad (2)$$

$$\left(\frac{\Gamma_0}{\Delta}\right)_A = -\frac{1}{2}\frac{r^2e^{2ikL_C}e^{-ikl/2} + 2re^{ikL_C}\cos(kl/2) + e^{ikl/2}}{\sin(kl/2)(r^2e^{2ikL_C}e^{-ikl} + 2re^{ikL_C} + e^{ikl})}. \quad (3)$$

Here $\Delta = \omega - \omega_0 + i\gamma$, with ω_0 and γ frequency and nonradiative broadening of the exciton; $l = L_W + L_B$, where L_W, L_B are the thickness of the well and of the barrier, respectively; L_C is the cavity length; $\Gamma_0 = \frac{1}{4\pi\epsilon_0}\frac{\pi}{n}\frac{e^2}{m_0c}f_{xy}$ is the radiative broadening for the exciton in a SQW (f_{xy} is the oscillator strength per unit area), and $k = n\omega/c$. For $r \to 0$ (no mirrors) the results of Ref. 21 for the polariton dispersion of a DQW in free space are recovered.

For notational simplicity we restrict ourselves from now on to the case of a λ cavity; we further assume that the center of the stop band of a DBR coincides with the frequency of the cavity mode and that the phase of $r(\omega)$ is zero at $\omega = \omega_m$. In this way the field intensity is maximum at the center of the MC, where the DQW is positioned. We use the parametrized expression of $r(\omega)$ and expand the right-hand side (RHS) of (2) and (3) close to resonance. For the A solution, the RHS in (3) is never zero at $\omega = \omega_m$ for any value of R, so that we can treat the A solution in the whole range of reflectivity values by evaluating the RHS at $\omega = \omega_m$ (when $\omega_0 = \omega_m$, this coincides with the exciton-pole approximation, and is equivalent to lowest-order perturbation theory). For $r \to 1$ we find

$$\omega_A = \omega_0 - \Gamma_0 \sin k_0 l - i\gamma\ , \quad (4)$$

which means that the A solution is not coupled at all with the radiation field. On the contrary, this approximation does not suffice for the S solution when $R \to 1$: in fact for $r = 1$ and $\omega = \omega_m$ the numerator in the RHS of (2) becomes zero (i.e., the radiative shift of the exciton becomes very large), which is a signature of the crossover to the strong-coupling regime. The high reflectivity case for the S solution requires an expansion of the RHS in (2) up to first order in $\omega - \omega_m$. For $R \approx 1$ this leads to the model of two damped coupled harmonic oscillators:

$$(\omega - \omega_0 + i\gamma)(\omega - \omega_m + i\gamma_c) = V_S^2, \quad (5)$$

where the parameters V_S and γ_c are given by

$$V_S = \sqrt{\frac{2\Gamma_0 c}{nL_{\rm eff}}(1+\cos k_0 l)} \qquad \gamma_c = \frac{c}{nL_{\rm eff}}(1-\sqrt{R}), \quad (6)$$

$k_0 = \frac{n}{c}\omega_0$, and $L_{\rm eff} = L_c + L_{\rm DBR}$ is an effective cavity length. We recall (see Ref. 20) that the same result was found also in the case of a SQW embedded in a MC, but in that case, $V = \sqrt{\frac{2\Gamma_0 c}{nL_{\rm eff}}}$. This means that when a DQW is present, cooperative effects due to e.m. coupling between the wells arise. The degree of cooperation is measured by an effective number of wells given by $n_{\rm QW} = 1 + \cos k_0 l$.[20,26] As discussed in Ref. 20, when $|V_S| > |\gamma_c - \gamma|/2$ the strong-coupling regime occurs.

These results can be interpreted as follows. The S solution couples with the symmetric e.m. field with the same phase in the two QW's, so that the radiation-matter coupling is now reinforced with respect to a SQW. On the contrary, the coupling between the exciton and the e.m. field in one QW takes place with a phase that is the opposite of that with the other QW, so that a cancellation of the coupling occurs for the A mode (see Fig. 1).

We now consider the weak-coupling regime and restrict ourselves for notational convenience to the long-wavelength limit. Expanding up to leading order in $k_0 l$, the A solution is found to give rise to a deenhanced emission rate:

$$\Gamma_A = (\Gamma_0/2)(k_0 l)^2[(1-\sqrt{R})/(1+\sqrt{R})]. \quad (7)$$

On the contrary, the S solution has an enhanced decay rate:

$$\Gamma_S = 2\Gamma_0[(1+\sqrt{R})/(1-\sqrt{R})]. \quad (8)$$

The factor 2 stems from the e.m. coupling between the wells, which emit in phase, whereas the enhancement factor $F = \frac{(1+\sqrt{R})}{(1-\sqrt{R})}$ is due to the presence of the MC and is the same found for a SQW.[3,6,18,20]

As discussed in Ref. 20 for the case of a SQW, the enhanced exciton decay of the symmetric solution reaches its maximum at the reflectivity value R_C, which marks the crossover from weak to strong coupling, and which is given for the DQW case by $1 - R_C = 4\sqrt{2}(nL_{\rm eff}n_{\rm QW}\Gamma_0/c)^{\frac{1}{2}}$ with $n_{\rm QW} = 1 + \cos kl \approx 2$. At this point, $\Gamma_S = \gamma_c/2 = \sqrt{2}(cn_{\rm QW}\Gamma_0/nL_{\rm eff})^{\frac{1}{2}}$. With the parameters of Ref. 20, which correspond to the cavity parameters of Ref. 19, this point corresponds to a criti-

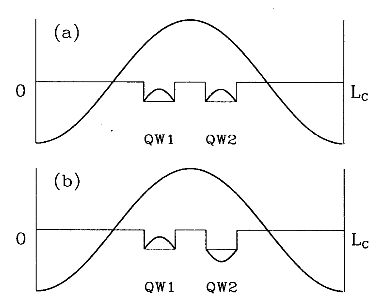

FIG. 1. Schematic picture of the electric field profile in a λ cavity with a symmetric mode, in interaction with (a) the symmetric state, and (b) the antisymmetric state of a DQW.

cal reflectivity $R_C \approx 0.8$, which gives a radiative lifetime $\tau = 1/(2\Gamma_S) \approx 140$ fs. This is much shorter than both momentum scattering and dephasing times, which are in the ps range.[8]

The main problem with the coupled recombination is that the energy difference between the excitons located in different wells (due mainly to well-width fluctuations) acts as a dephasing mechanism. Since the energy mismatch of the wells is typically much larger than Γ_0 (which is usually of the order of 0.03 meV), observation of ultrafast decay in free space seems problematic. In a MC, however, the situation is expected to be more favorable, because now vertical disorder competes with the enhanced decay rate produced by the cavity. In order to quantify this expectation, we now study the model situation in which the two QW excitons have different energies ω_{01}, ω_{02} (although the energy difference arises from different well widths, we still take the two wells to have the same thickness). For simplicity we assume the same nonradiative broadening γ. Setting $T_{22} = 0$, we find

$$\omega_{1,2} = \frac{\omega_{01}+\omega_{02}}{2} - i\gamma - i\Gamma_0 \frac{1 + r^2 e^{2ikL_C} + 2re^{ikL_C}\cos kl}{1 - r^2 e^{2ikL_C}} \pm \frac{1}{2}\sqrt{(\omega_{01}-\omega_{02})^2 - 4\Gamma_0^2 \frac{(e^{ikl} + r^2 e^{2ikL_C} e^{-ikl} + 2re^{ikL_C})^2}{(1 - r^2 e^{2ikL_C})^2}} . \qquad (9)$$

These relations contain no approximations and apply in the whole range of reflectivity values. It is clear that S and A solutions are no more decoupled in the presence of vertical disorder.

We first consider the high-reflectivity limit. By rewriting (9) in a form similar to (2) and expanding up to first order in $\omega - \omega_m$ we obtain

$$\Delta_1 \Delta_2 (\omega - \omega_m + i\gamma_c) = (V_S^2/2)(\Delta_1 + \Delta_2) , \qquad (10)$$

where V_S is given by (6) and $\Delta_j = \omega - \omega_{0j} + i\gamma$, $j = 1, 2$. Relation (10) is also obtained by diagonalizing a 3×3 Hamiltonian, in which two oscillators of frequencies ω_{01} and ω_{02} are coupled with a third oscillator of frequency ω_m. For this reason we refer to (10) as to a three-oscillator model. For $\omega_{01} = \omega_{02}$ one solution is not coupled anymore, while for the other one relation (5) is recovered. Equation (10) leads to the expectation that three peaks can be observed in optical spectra in the strong-coupling regime. For simplicity we consider the symmetric situation $\omega_{01} = \omega_m + \delta$, $\omega_{02} = \omega_m - \delta$. In the particular case $\gamma = \gamma_c$, one solution is $\omega = \omega_m - i\gamma_c$, while the other two solutions are $\omega = \omega_m - i\gamma \mp \sqrt{\delta^2 + V_S^2}$. The strong-coupling regime persists until $|\gamma - \gamma_c| \approx V_S$. In Fig. 2 we show the calculated reflectivity for a λ cavity with an antisymmetric mode and two groups of three QW's at the maxima of the electric field (see inset). We assume 60-Å-wide GaAs-$Al_{0.4}Ga_{0.6}As$ QW's with $\hbar\omega_0 = 1.6$ eV and an oscillator strength $f_{xy} = 7.4 \times 10^{12} \text{cm}^{-2}$, giving $\hbar\Gamma_0 = 0.038$ meV. We take $n = 3.46$ and a HWHM $\gamma = 1.5$ meV. The two values of the energy difference 2δ correspond to a difference of one and two monolayers, respectively, between the two sets of wells. The matrix element for each set of three QW's is $V = 2.9$ meV, therefore $V_S = \sqrt{2}V = 4.1$ meV. It can be seen from Fig. 2 that the three peaks have comparable weights only when $\delta \approx V$. When $\delta \ll V$ only two peaks are found (this corresponds to the case previously treated of identical resonance energies). When $\delta \gg V$, on the other hand, the central peak has most of the weight, while the lateral peaks corresponding to the excitons have very little weight. Thus we conclude that the best condition for observing the three peaks is when $\delta \approx V$, i.e., when half of the energy difference matches the matrix element.

We then consider the weak-coupling regime. Evaluating the RHS of (9) at $\omega = \omega_m$ and expanding up to first order in $k_0 l$ (which is appropriate in the long-wavelength limit) two distinct behaviors are found to hold, depending on the ratio of vertical disorder to the enhanced radiative broadening (8). If the condition $|\omega_{01} - \omega_{02}| \gg \Gamma_S$ holds, we have for the radiative widths

$$\Gamma_1 \approx \Gamma_2 \approx \Gamma_0[(1+\sqrt{R})/(1-\sqrt{R})], \qquad (11)$$

i.e., the coupled decay due to the coherent emission of the two wells is washed out by strong disorder, so that the lifetime is now the same as for a SQW in a MC. On the other hand, if $|\omega_{01} - \omega_{02}| \ll \Gamma_S$ we find

$$\Gamma_1 \approx 2\Gamma_0[(1+\sqrt{R})/(1-\sqrt{R})], \qquad (12)$$

$$\Gamma_2 \approx \tfrac{1}{8}[(1-\sqrt{R})/(1+\sqrt{R})][(\omega_{01}-\omega_{02})^2/\Gamma_0]. \qquad (13)$$

Ultrafast, "superradiant" emission for the first solution (which, of course, becomes the S solution for $\omega_{01} = \omega_{02}$) is therefore expected when $|\omega_{01} - \omega_{02}| \ll \Gamma_S$. Since the decay rate of the S solution is enhanced by a large factor (e.g., $\Gamma_S \approx 36\Gamma_0 \approx 1$ meV for $R = 0.8$), it is clear that coupled emission between wells in a MC can occur, even when it is suppressed in free space.

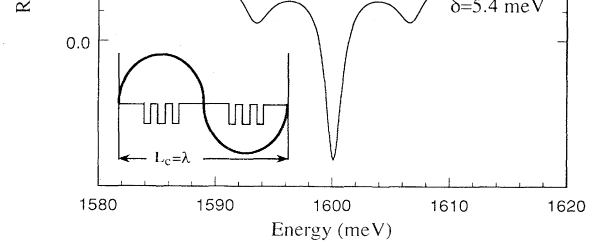

FIG. 2. Reflectivity of a λ cavity with an antisymmetric mode, containing two sets of three GaAs/$Al_{0.4}Ga_{0.6}As$ QW's (L_W=60 Å, L_B=100 Å) at the $\lambda/4$ and $3\lambda/4$ positions (see inset). The HWHM is $\gamma = 1.5$ meV. The three curves (two of which are offset for clarity) correspond to different values of the energy difference 2δ between the excitons in the two sets of QW's (see text).

Up to now we have developed a theory that allows the evaluation of the energies and decay times of the coupled modes. We now discuss how our predictions should be compared with a time-resolved transmission experiment. To this purpose, we calculate the response to a short Gaussian pulse tuned to the exciton energy; we require the time duration $\Delta\tau$ of the incoming wave packet and the radiative decay rate Γ_0 of the exciton to satisfy the condition $\Delta\tau \ll \Gamma_0^{-1}$ if the enhanced decay is to be observed.

We first discuss the temporal response of a DQW in free space. By Fourier transforming the linear response of the system to the time domain, we find for the transmitted electric field at times $t \gg \Delta\tau$:

$$E_t \propto \cos^2(k_0 l/2) e^{-iRe(\omega_S)t} e^{-\Gamma_S t} + \sin^2(k_0 l/2) e^{-iRe(\omega_A)t} e^{-\Gamma_A t} \quad (14)$$

(ω_S and ω_A are the complex energies of the S and A modes). Three relevant time scales characterize the output signal (which is proportional to $|E_t|^2$). The symmetric and antisymmetric solutions decay exponentially in a time $\tau_S = 1/(2\Gamma_S)$ and $\tau_A = 1/(2\Gamma_A)$, where Γ_S and Γ_A represent the radiative broadening of the S and A solutions, respectively. In the output signal a third term also arises, which oscillates with a frequency given by $Re(\omega_S - \omega_A)$. This term is the signature of the interwell e.m. coupling and is a typical beating effect.[8,28] The period of such an oscillation is, however, very long compared with τ_S and τ_A, so we do not think it could be evidenced in the experiment.

The ratio between the signals from the S and A modes is $R = \cot^4 k_0 l/2$, so that, in the long-wavelength limit, the strongest signal comes from the S solution. Embedding the DQW in a MC should further favor the S mode with respect to the A one, and the preceding ratio is expected to be now multiplied by F^4. In such a configuration we then predict that the short decay time of the S mode could be observed as the decay of the coherent signal in a time-resolved experiment.

Our conclusion about the coupled radiative decay for a DQW in a MC can be stated as follows: the initial pulse prepares the system in a state that nearly coincides with the S state, i.e., in a state in which the two QW excitons oscillate in phase. This leads to a short, coherent decay signal, which is observable for times smaller than the dephasing time T_2 [in our model, $T_2 \approx (\omega_{01} - \omega_{02})^{-1}$]. The same reasoning can be applied for understanding the effect of in-plane disorder: even if the excitons are heavily scattered and/or localized by in-plane disorder, the excitation pulse sets up a coherent superposition of all these oscillators, which decays with the superradiant decay rate Γ_S for $t < T_2$. These conclusions agree with those of Ref. 3. Thus the ultrafast radiative decay derived here can be observed as a short coherent signal in a time-resolved experiment in the transmitted or reflected directions, with the only condition that the decay time is shorter than the dephasing time. By putting several QW's in a MC, it will be possible to increase the effective cooperative number of QW's to about six,[23] thereby leading to a radiative lifetime of $\approx$ 80 fs.

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