Photoluminescence decay times in strong-coupling semiconductor microcavities

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A theoretical investigation of the dynamics in a photoluminescence experiment in strong-coupling semiconductor microcavities is presented. Radiative recombination rates of microcavity polaritons in the strong-coupling regime and their scattering rates with acoustic phonons are used to study the polariton dynamics in a photoluminescence experiment, as a function of the temperature and the cavity detuning. It is found that the leaky modes of the distributed Bragg reflectors enclosing the cavity constitute the main sink of radiation, and that these modes dominate the whole recombination process. As a result, the rise and decay times are almost independent of the cavity detuning. Moreover, these times are close to those of the bare exciton at any temperature, thus showing little cavity influence on the overall photoluminescence dynamics.

I. INTRODUCTION

Semiconductor microcavities of increasingly better quality have been recently realized in many laboratories.[1–4] In these structures, quarter-wavelength stacks of transparent semiconductors, known as distributed Bragg reflectors (DBR's), are used to efficiently confine the light in the cavity.[5] The resulting cavity mode may then be coupled to an exciton resonance by inserting a quantum well (QW) inside the cavity. This coupling becomes strong for large cavity finesses, and mixed exciton-cavity photon modes result, split by up to a few meV in the III-V materials considered.[2,3] Radiative emission and splitting of these modes may be tailored by changing the properties of the cavity, and radiative recombination rates may be sizably enhanced. A main point to be addressed is the modification of the well-known bare-exciton dynamics[6] in a photoluminescence (PL) experiment due to photon confinement. In fact, the nature of the lowest radiative levels is fundamentally changed in the strong-coupling microcavity. Moreover, a sizable decrease of the PL decay times is desirable for optical applications. In this paper we present a theoretical investigation on this point, and make a comparison with experimental results. The mixed exciton-cavity photon modes typical of strong coupling are effectively described as microcavity polaritons (MCP's), and have been a main subject of theoretical[7,8] and experimental investigations.[2,3] The problem of the PL dynamics involves exciton-polariton relaxation by scattering with phonons. It is known that scattering by phonons is inhibited in the region of strong exciton-photon coupling.[9] The problem of the competition between the relaxation rates and the enhanced radiative recombination rates of the cavity polaritons arises. We find that photon-scattering rates are actually insufficient to provide full thermalization of the emission in the strong-coupling region. However, the phonon-scattering rates are essentially unaltered in the excitonlike part of the dispersion of the lower polariton branch and are thus able to provide partial thermalization. In a portion of this dispersion, polaritons radiate into the leaky modes of the DBR's.[8] The existence of these modes is due to the low reflectivity of the DBR's at large angles. We have found that the rise and decay times of the PL signal are determined by these leaky modes only, and are thus practically uninfluenced by the cavity structure. In fact, rise and decay times close to those of a corresponding bare-exciton structure are found for any temperature and cavity detuning. Comments and comparison with recent experimental results are, finally, discussed.

II. RATE EQUATION MODEL OF THE DYNAMICS

In this work we consider a realistic one-wavelength semiconductor microcavity having a single 150-Å GaAs QW embedded at its center. DBR mirrors consisting of 24 and 20 pairs of quarter-wavelength stacks, respectively, enclose the cavity. A detailed description of the structure and materials constituting such a strong-coupling microcavity may be found elsewhere.[2] Since our system is planar, full translational symmetry on the QW plane allows us to introduce an in-plane wave-vector label denoted by $\mathbf{k}$ for the states of the system, which is used throughout this work. For each $\mathbf{k}$, the mixed exciton-cavity photon modes resulting from the electromagnetic interaction are calculated exactly within a quantum model.[8] We used a calculated value of 40×10^{-5} Å^{-2} for the QW oscillator strength.[10] The resulting dispersion $\omega_{\mathbf{k}}$ and total radiative broadening $\Gamma_{\mathbf{k}}$ of the polariton modes are shown in Figs. 1(a), 1(b), and 1(c). Four different detunings $\delta=\omega_{\rm cav}-\omega_{\rm exc}=0$, 3, -3, and -6 meV, where $\omega_{\rm cav}$ is the cavity resonance frequency at normal incidence, and $\hbar\omega_{\rm exc}$ the exciton energy at normal incidence, have also been considered. From Fig. 1(a) we see that the Rabi splitting Ω of the polariton modes for zero detuning and at normal incidence is slightly larger than 3 meV. Therefore, the cavity is in the strong-coupling regime. We recall that the radiative broadening is nonvanishing in the radiative region only. This region is defined by the condition $k<k_0=n_{\rm sub}\omega/c$, where $n_{\rm sub}$ is the substrate index of refraction and ω is the energy of the considered excitation. We notice that the radiative emission rate at $k=0$ is the same for the upper and lower polari-

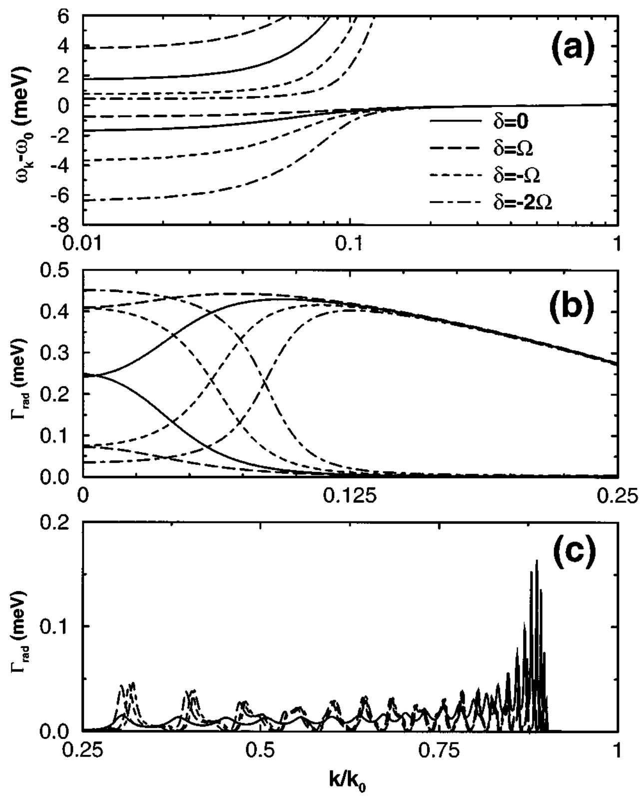

FIG. 1. Microcavity polariton dispersion $\omega_{\mathbf{k}}$ (a) and total radiative recombination rate $\Gamma_{\mathbf{k}}$ in the strong-coupling region (b), and in the leaky modes region (c), for a typical GaAs sample, at four different detunings δ. Parameters are given in the text.

tons only when $\delta=0$, and the lower branch radiative rates become larger for negative detunings and smaller for positive ones [Fig. 1(b)]. We also notice that the the calculation reproduces the radiative emission of the cavity polaritons into the leaky modes, for $k>0.25k_0$, Fig. 1(c), because the reflectivity of the DBR is exactly included in the calculation. It is clear from Fig. 1(c) that these modes remain practically unchanged at different detunings, with exception made for an overall minor energy displacement.

Next, we consider the PL process. In PL experiments, free electron-hole pairs are created by a short (a few picoseconds) exciting light pulse. These free pairs quickly cool down to low kinetic energies. Highly energetic bound excitons (at an energy close to the exciton binding energy) are then formed by acoustic-phonon emission. We do not describe these fast initial steps of the PL process as we are only interested in the subsequent relaxation dynamics. This dynamics takes place on much longer time scales. We therefore choose as an initial condition a sharply peaked excitonic population at the energy of the exciton continuum. Large initial $\mathbf{k}$ vectors, up to $10k_0$, have to be considered to describe this lower branch exciton-polariton distribution. The precise shape of the initial distribution slightly influences the rise times, but not the decay times. In fact, memory of this shape is rapidly lost during the initial rise of the PL signal. We also remark that coherence of the exciton polaritons is absent in a nonresonantly pumped experiment such as the one we are considering, because coherence of the free electron-hole pairs is rapidly washed out in the initial steps of relaxation.[11] Thus, the whole process may be conveniently described in terms of rate equations for populations of the different levels. Two dynamical mechanisms are considered, scattering by acoustic phonons and radiative recombination. All other scattering mechanisms such as exciton-exciton, exciton-free carrier scattering, and scattering with optical phonons have been neglected. The range of validity of the model is therefore limited to low excitation densities and low temperatures. In particular, the neglect of exciton ionization forces us to consider temperatures lower than 100 K. Transition rates of MCP scattering by acoustic phonons $W_{i,\mathbf{k}\rightarrow j,\mathbf{k}'}$, where $\mathbf{k}$ and $\mathbf{k}'$ are the in-plane MCP wave vectors and i,j are the branch indexes have been calculated by using the deformation-potential interaction and the Fermi golden rule. The scattering rate is subdivided as $W_{i,\mathbf{k}\rightarrow j,\mathbf{k}'}=W_{\rm abs}+W_{\rm em}$, where $W_{\rm abs}$ involves absorption of three-dimensional phonons of wave vector $\mathbf{q},q_z$, and $W_{\rm em}$ is the emission of phonons. Fermi golden rule for the absorption term gives

$$W_{\rm abs}=\frac{2\pi}{\hbar}\sum_{q_z}|X^*_{j,\mathbf{k}'}\langle\mathbf{k}'|\langle 0_{\mathbf{q},q_z}|H_{\text{exc-ph}}|1_{\mathbf{q},q_z}\rangle|\mathbf{k}\rangle X_{i,\mathbf{k}}|^2 \times n_{E_{\rm ph}}\delta[E_{j,\mathbf{k}'}-E_{i,\mathbf{k}}-E_{\rm ph}(\mathbf{q},q_z)]. \quad (1)$$

Here $\mathbf{q}=\mathbf{k}'-\mathbf{k}$ is the exchanged in plane momentum, $H_{\text{exc-ph}}$ is the deformation-potential interaction, $|n_{\mathbf{q},q_z}\rangle$ is a phonon number state, $|\mathbf{k}\rangle$ is the exciton state of wave vector $\mathbf{k}$, and $E_{i,\mathbf{k}}$ is the polariton energy, branch i, wave vector $\mathbf{k}$. Here $n_{E_{\rm ph}}$ is the Bose occupation number for phonons, and $E_{\rm ph}$ is the phonon energy. The $X_{i,\mathbf{k}}$ are the Hopfield coefficients whose square modulus gives the exciton content in the polariton state of branch i, wave vector $\mathbf{k}$.[7] We remark that the matrix element of the electron-phonon interaction between two MCP states is thus scaled by the exciton content in both the initial and final polariton modes. This scaling significantly lowers the scattering rates in the photonlike regions of the polariton dispersion, while it is, substantially, unity in the excitonlike part of the lower branch dispersion. A further significant lowering of the rates is due to the reduction of the scattering phase space in the photonlike regions of polariton dispersion.[9] The expression for $W_{\rm em}$ is completely analogous to that of $W_{\rm abs}$, provided the Bose occupation factor n in Eq. (2) is replaced by $n+1$ and $E_{\rm ph}$ by $-E_{\rm ph}$ in the Dirac δ function.

In the numerical calculations of the scattering rates W we used the GaAs bulk values of Ref. 12 for the deformation potentials, and electron and hole masses taken from detailed envelope function calculations.[10] We kept the isotropic term of the interaction only, which is related to the longitudinal-acoustic phonons. These assumptions have been thoroughly tested in the bare-exciton case (open cavity), and found to give values that compare well with experimental results.[13] In particular, the slope of linear increase of the phonon-scattering rates of $\mathbf{k}=0$ excitons with temperature[14] is well reproduced.

Rate equations for the MCP population $n_{i,\mathbf{k}}$ at a given in-plane $\mathbf{k}$ vector for branch i are introduced by using the scattering rates $W_{i,\mathbf{k}\rightarrow j,\mathbf{k}'}$ and the total radiative recombination rates $\Gamma_{\mathbf{k},i}$:

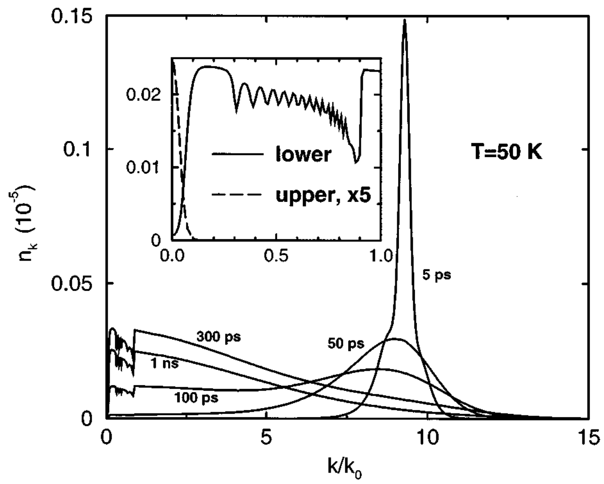

FIG. 2. The population of the lower polaritons at different time delays after excitation, at $\delta=0$, and at $T=50$ K. The inset shows (enlarged) the radiative region, and the upper branch population, at a delay of 1 ns.

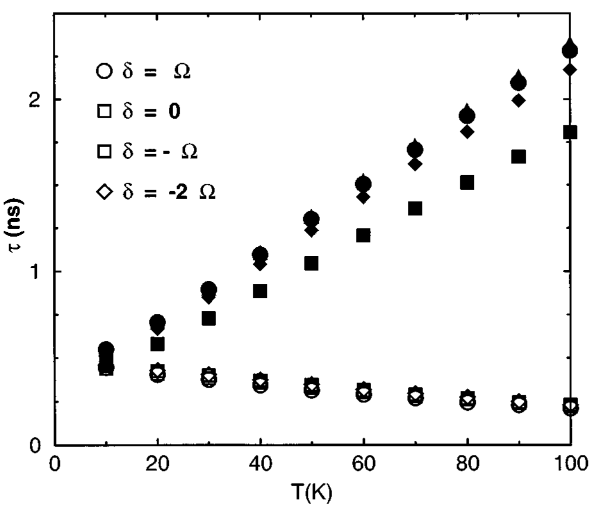

FIG. 3. Rise time τ_R (empty symbols) and decay time τ_D (filled symbols) for the semiconductor microcavity in the case of four different detunings δ indicated in the figure.

$$\dot{n}_{i,\,\mathbf{k}}=\sum_{j,\mathbf{k}'} W_{j,\mathbf{k}'\to i,\mathbf{k}} n_{j,\mathbf{k}'}(n_{i,\mathbf{k}}+1) - \sum_{j,\mathbf{k}'} W_{i,\mathbf{k}\to j,\mathbf{k}'} n_{i,\mathbf{k}}(n_{j,\mathbf{k}'}+1) - \Gamma_{i,\mathbf{k}} n_{\mathbf{k}}. \qquad (2)$$

The $(n_{\mathbf{i},\mathbf{k}}+1)$ factors result from the bosonic character of excitons, and represent stimulated emission and absorption processes, which are negligible in the range of densities considered. We also assume an isotropic excitation of the system and obtain isotropic populations at any later time.

Finally, the observed PL signal is easily computed, being proportional to the total number of photons emitted per unit time and per unit solid angle. In a real experiment, integration over a small detection window of a few degrees is carried out. We therefore also integrate the rate of emitted photons around normal incidence over a small detection window of $\Delta\theta=5°$. We have

$$I_{\mathrm{low}}(\mathrm{t})=\int_0^{\Delta k} \Gamma_{\mathrm{low},\,\mathbf{k}} n_{\mathrm{low},\,\mathbf{k}}(t)\, d\mathbf{k}/k_0^2. \qquad (3)$$

Here $\Delta k = k_0 \sin\Delta\theta$, and an analogous equation holds for the upper branch signal.

III. PHOTOLUMINESCENCE DYNAMICS

As explained in the previous section, rate equations are numerically integrated choosing a sharply peaked distribution for the lower branch MCP at the energy of the exciton continuum (10 meV) as the initial condition. In the first part of the evolution, up to a few hundreds of picoseconds, both lower and upper branch MCP at small **k** are created after several steps of phonon emission, as shown in Fig. 2, for $T=50$ K and $\delta=0$. The PL signal correspondingly rises. For longer times, the polariton distribution reaches a quasistationary shape, which rigidly decreases in time (Fig. 2, inset). The PL signal therefore follows this decay.

The PL decay and rise times as functions of the temperatures and for the considered detunings are shown in Fig. 3. The PL decay time τ_D is found to be linearly increasing with temperature. This behavior of τ_D is the same one found for the bare QW exciton.[6,13,15] The same is true for the rise time.[13] The values obtained are also quantitatively close to those of a bare QW of 150 Å.[6] It is remarkable that both τ_D and τ_R are not much influenced by the cavity detuning δ. This fact is surprising when considering that both radiative rates and phonon-scattering rates in the strong-coupling region indeed are. We notice from Fig. 2 that, apart from small deviations in the radiative region, and close to $\mathbf{k}=0$, the lower branch distribution is Boltzmann-like. In particular, among the radiative states, the leaky modes are mostly populated, while the modes close to $\mathbf{k}=0$ are strongly depleted. Therefore, it is only the leaky modes that play the fundamental role in the overall PL dynamics. In the region of the leaky modes, the radiative recombination rates and the phonon-scattering rates are *not* significantly influenced by the cavity detuning δ. This justifies the result found in Fig. 3, where little influence on the cavity detuning, and in fact on the presence of the cavity itself, of the characteristic rise and decay times is found.

Although the overall PL dynamics is insensitive to the cavity confinement, the dynamics in the strong-coupling region, defined by $k<0.25k_0$, is necessarily much different from the bare QW case. First of all, two modes instead of only one appear in the strong-coupling case. Moreover, phonon scattering significantly slows down and radiative recombination rates are sizably enhanced. We do not enter into the details of the dynamics in this region, which may also be experimentally studied looking at the PL signal as a function of the emission angle, but just add a few remarks. It is shown in Fig. 2 that the polariton population in the lower branch is strongly depleted in this region. This suggests analogies with the polariton bottleneck of bulk polaritons.[16] However, the main difference between the two systems is that bulk polaritons do not have an intrinsic radiative recombination rate, so that practically only photonlike polaritons radiate at the sample surfaces. In particular, in the populated bottleneck region, low radiative rates are present.[16] Thus, it is the *relaxation* dynamics that governs the overall PL dynamics. On the other hand, in our case sizable radiative recombination rates in the leaky mode region are present, thus it is the leaky mode radiative recombination that governs the overall dynamics.

Finally, we comment on the effects of the large inhomogeneities of the exciton energies, which are present in currently grown samples. We notice that a naive picture of strong coupling does not apply to a strongly inhomogeneously broadened exciton. In this case, many different fractions of the exciton line interact with the same cavity mode. However, the interaction with the photon field is not of resonant type in the leaky mode region, and is presumably rather flat over the whole exciton line. The integrated decay rate over the whole exciton line remains unchanged. If this is the case, the conclusions of this paper on the small influence of the cavity confinement on the rise and decay times of the PL still hold in the case of the inhomogeneously broadened exciton.

IV. CONCLUSIONS

In this paper we have studied the dynamics of the microcavity polaritons in the strong-coupling regime. In the calculation we have included the detailed features of the DBR reflectivity, and therefore also the radiative recombination rates into the leaky modes. We have shown that these modes play a fundamental role in the dynamics. In particular, they produce an excitonlike behavior of the relaxation rates. Typical rise and decay times of the bare QW's are in fact obtained for any temperature and cavity detuning. The importance of the leaky modes is due to the large size of the solid angle of emission they span. Experiments on the time-resolved PL from strong-coupling microcavities support this result.[17] In particular, no sizable changes in the PL decay time can be inferred as the detuning is changed. Some analogies of the dynamics inside the small region of strong exciton-photon coupling with that of the bulk have also been mentioned. However, we noticed that the existence of radiative decay at large angles is peculiar to two-dimensional structures only.

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