Optical bistability of semiconductor microcavities in the strong-coupling regime

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(Received 20 February 1996; revised manuscript received 28 May 1996)

We propose a mechanism for achieving optical bistability in semiconductor heterostructures. In contrast to the well-known refractive shift of the Fabry-Perot mode in macroscopic resonators, we consider the bleaching of the exciton Rabi splitting in strong-coupling microcavities and prove that this nonlinear process leads to an optically bistable behavior. Through a model based on the transfer-matrix formalism, we predict the dependence of this effect on specific material parameters and on the nature of the cavity. Numerical calculations are carried out for quantum well embedded cavities and the possibility of room-temperature bistable operation is also discussed. [S1050-2947(96)01610-1]

PACS number(s): 42.65.Pc, 71.35.−y, 42.50.−p, 78.66.−w

I. INTRODUCTION

Semiconductor microcavities have attracted a continuously growing interest, mainly related to the possibility of controlling spontaneous emission and of studying fundamental aspects of light-matter interaction [1–11]. In particular, the ''strong-coupling'' regime, in which the exciton and the cavity photon are in such close interaction to give rise to two coherent mixed modes, often referred to as ''cavity polaritons,'' has received a lot of attention. The Rabi splitting of the modes and the oscillating behavior of their emission have been detected in a number of different systems and in a wide range of temperatures. [3,4,12–17].

In view of applications for optoelectronic devices, the analysis of the nonlinear optical properties offers a fundamental and intriguing field of microcavity physics, which is still to be fully explored. Very recently, a crossover from the strong-coupling to the weak-coupling regime has been observed at low temperatures in quantum well (QW) microcavities both in resonance [18], and out of resonance with the exciton transition [19]. In [19] the sample was excited well above the continuum edge, so that free electron-hole pairs were generated. Under low intensity excitation, the cavity emission showed a familiar Rabi splitting, which was, however, bleached at high intensities. A similar behavior was observed by Norris *et al.* [18] in the reflectance spectrum. A spectrally broad resonant laser pulse was used to cover the Rabi split peaks. The physical origin of the bleaching is the same in both experiments. It is in fact a consequence of the saturation of the exciton oscillator strength with carriers (bound or free) density, due to screening and phase-space filling effects. However, owing to the very different lifetimes of free electron-hole pairs (nanosecond scale) and cavity-coupled excitons (less than 1 ps), the use of an ultrafast pulsed source became necessary in [18], while in [19] a low power cw laser was sufficient. Nevertheless, it is evident that for optoelectronic applications exciton resonant, or nearly resonant, operation is a much more interesting situation; therefore, in this work we will limit ourselves to this condition.

Our purpose is to show, with a fairly simple theoretical model, that the Rabi splitting bleaching leads to optically bistable behavior. We will also perform an accurate analysis of possible ways to alter and control the performances of this ideal bistable device.

We wish to observe that the mechanism proposed here is completely different from the dispersive optical bistability which has been extensively studied and demonstrated in thick semiconductor Fabry-Perot (FP) resonators [20–23]. In fact, conventional FP bistability relies on the variation of the FP refractive index with radiation intensity, which causes a shift in the energy of the FP optical mode (owing to the change of optical thickness) eventually giving rise to positive feedback. In the process the radiation is very weakly coupled to the exciton, and the FP mode, which is required to be off resonance to maximize the performances, retains a purely photonic character. On the contrary, in a perfectly tuned microcavity, the electromagnetic wave gives birth to mixed exciton-photon states, referred to as cavity polaritons, and we will show how bistability can result from a modification in the coupling strength which changes the energy of the polariton modes. Different from thick FP bistability, this is clearly a purely resonant phenomenon, whose magnitude depends on the rate of variation of the exciton susceptibility alone, and not of the total dielectric constant. As will be apparent in the following, the proposed mechanism presents advantages related to the cavity polariton dynamics, which is faster and more controllable than the exciton one, and to the larger exciton generation rate attainable with resonant operation.

The paper is organized as follows: in Sec. II we give a description of our theoretical model, results are presented and discussed in Sec. III, conclusions and perspectives are examined in Sec. IV.

II. THEORY

The density of electron-hole pairs in a semiconductor has a great importance in determining excitonic properties. It is well known, in fact, that dielectric screening and phase-space filling effects act efficiently in reducing the oscillator strength f when the electron-hole density n is sufficiently

large [24]. Experiments and simple theoretical considerations [25,26] indicate a saturation behavior of the type

$$f=\frac{f_0}{1+n/n_s}, \tag{1}$$

n_s being the saturation density at which f is half its value at zero density f_0. Both free and bound e-h pairs can contribute, with relative relevance depending on the sample temperature [27]. We consider the case of resonant excitation at low temperature, when practically no free carriers are generated because no LO phonon absorption is allowed. Room temperature operation will, however, be discussed qualitatively later on.

For the sake of simplicity, we choose as our system a standard $Al_{0.1}Ga_{0.9}As$ microcavity with 20 pairs AlAs/$Al_{0.2}Ga_{0.8}As$ Bragg reflectors. A single 200 Å GaAs QW is placed at the center of the cavity. The Fabry-Perot resonance is exactly tuned on the main heavy-hole exciton transition. The case of bulk cavities, although being equivalent in the main features, present additional complexities which make a detailed calculation more cumbersome. They will be described later on.

We can now write down a rate equation for the QW exciton population [28,29], which, however, must be divided into two components in order to keep into account the difference between $k_\parallel \simeq 0$ excitons, which are strongly coupled with the cavity mode, and larger $k_\parallel$ excitons, which behave like the usual QW excitons and decay primarily in the plane of the cavity [18]. Our equations then read

$$\frac{\partial N_0}{\partial t}=-\frac{N_0}{\tau_0}+\frac{2d}{\hbar}I_0\mathrm{Im}(\chi)-\frac{N_0}{\tau_m}+\frac{N_k}{\tau_p}, \tag{2a}$$

$$\frac{\partial N_k}{\partial t}=-\frac{N_k}{\tau_k}+\frac{N_0}{\tau_m}-\frac{N_k}{\tau_p}, \tag{2b}$$

where N_0 and N_k are the densities of coupled and uncoupled excitons, respectively, $(n=N_0+N_k)$, I_0 is the intensity of the electromagnetic field at the QW position, and d the QW thickness. The radiative lifetime τ_0 of $k_\parallel \simeq 0$ excitons is determined principally by the cavity properties and is usually of several hundreds of femtoseconds [15–17], much shorter than the radiative lifetime τ_k of uncoupled excitons (some ten picoseconds [30]). The last two terms in the above equations take into account momentum scattering processes resulting mainly from interface roughness (characteristic times of the order of 1 ps depending on the inhomogeneous broadening) [18] and from phonon interaction (times varying over a hundred picosecond range depending on the lattice temperature and the exciton energy) [31,32]. These processes broaden the initial $k_\parallel \simeq 0$ exciton distribution and are modeled inserting the two lifetimes τ_m and τ_p which represent the transfer rates from cavity-coupled excitons to uncoupled excitons and vice versa. Clearly, the value of $\tau_{m,p}$ is also determined by the width of the cavity mode which fixes the $k_\parallel$ phase space for coupled excitons. In our case it seems reasonable to assume $\tau_m \simeq 1$ ps and τ_p several tens of picoseconds. As it will become apparent later on, a detailed knowledge of $\tau_{m,p}$, however, is not crucial in our calculations. Finally, we assume for the exciton contribution to the susceptibility a standard Lorentzian oscillator form

$$\chi=\frac{f\omega_0^2}{\omega_0^2-\omega^2-i\Gamma\omega}, \tag{3}$$

where ω_0 is the exciton resonance frequency and Γ the broadening factor. Neglecting in first approximation exciton-exciton scattering and considering that the band gap variation with exciton density is compensated by the quenching of the exciton binding energy, we can take into account the effect of the density only on the oscillator strength adopting Eq. (1).

In a standard experiment with pulsed excitation the input-output characteristic will usually show a hysteresis loop owing to the finite exciton lifetime, which makes it impossible for the population to instantaneously follow the variation of the pulse field. This does not necessarily imply a true bistability (the existence of two possible stable configurations of population and field for the same incident intensity) since it originates from the system being in a nonequilibrium condition. Therefore, we consider the case of quasistatic operation, in which the exciting pulse is somewhat longer than all the lifetimes of the exciton population, and put the partial derivatives in Eqs. (2) equal to zero. After some straightforward algebra we obtain

$$N_0+N_k=\frac{2d}{\hbar}I_0\mathrm{Im}(\chi)\frac{\dfrac{1}{\tau_p}+\dfrac{1}{\tau_k}+\dfrac{1}{\tau_m}}{\dfrac{1}{\tau_0\tau_p}+\dfrac{1}{\tau_k\tau_0}+\dfrac{1}{\tau_k\tau_m}}, \tag{4}$$

which can be solved in conjunction with Eqs. (1) and (3) to get the exciton population corresponding to a given intensity I_0 of the intracavity field.

In order to compute the optical response we now adopt a transfer-matrix approach in which every section of the microcavity structure is described by a 2×2 matrix defined as [33]

$$m_j=\begin{bmatrix} \cos\left(\dfrac{\omega}{c}n_j l_j\right) & -\dfrac{i}{n_j}\sin\left(\dfrac{\omega}{c}n_j l_j\right) \\ -in_j\sin\left(\dfrac{\omega}{c}n_j l_j\right) & \cos\left(\dfrac{\omega}{c}n_j l_j\right) \end{bmatrix}, \tag{5}$$

where $n_j=\sqrt{\varepsilon_j}$ is the refractive index, l_j the width of the jth section, and c is the velocity of light. For the QW we use $\varepsilon=\varepsilon_\infty+4\pi\chi$, χ being given by Eq. (3). The transfer matrix M for the whole cavity is then given by the product of the single matrices m_j, including Bragg mirror layers, cavity region, and QW region. From it the reflectance $R=|r|^2$ and the transmission $T=n_{sub}|t|^2$, where n_{sub} is the index of refraction of the substrate, are calculated in the usual way [33]

$$r=\frac{M_{11}+n_{sub}M_{12}-M_{21}-n_{sub}M_{22}}{M_{21}+n_{sub}M_{22}+M_{11}+n_{sub}M_{12}}, \tag{6a}$$

$$t=\frac{2}{M_{11}+n_{sub}M_{12}+M_{21}+n_{sub}M_{22}}. \tag{6b}$$

Clearly these two quantities depend on the exciton population through the dielectric constant of the QW layer, which in turn depends on the intensity of the intracavity field. Therefore, an additional relation is needed to link I_0 with the incident field I_{in} and with the reflected and transmitted waves. The electric field at the center of the cavity is related to the incoming field and the transmittance by the transfer matrix a corresponding to the half microcavity structure

$$a = m_{QW2} m_{c2} m_{RBR}, \tag{7}$$

where m_{QW2} is the transfer matrix of the half quantum well, m_{c2} is that of the half cavity, and m_{RBR} is the transfer matrix of the right Bragg reflector. Thus we obtain the following equation for the total transmittance:

$$T = I_t / I_{in} = I_0 / [I_{in} |a_{11} + a_{12}|^2]. \tag{8}$$

Our problem is then fully solved by finding solutions for the fields and the exciton population satisfying contemporarily Eqs. (4), (6), and (8). When more than one set of solutions for a given incident intensity exists, a multistable regime occurs, and the history of the system determines in which of these states the microcavity is.

We also wish to analyze the case of bulk cavities, where the active material extends over the whole cavity length, so that one has to consider the dependence of the electromagnetic field on the position. Furthermore, one should also include the wave-vector dependence in the susceptibility function χ. In principle we can proceed analogously to what was done before by inserting an explicit z-coordinate dependence in the intracavity intensity I_0 and in the exciton density n, and discretizing the cavity matrix in the product of several matrices each corresponding to a layer sufficiently thin to consider the field amplitude and the exciton population nearly constant. Equation (8) is then substituted by a full set of equations. The solution of such a system can be very complicated, and spatial dispersion makes it even more difficult owing to the need of additional boundary conditions. However, if one is interested only in the main features, the problem can be simplified by neglecting spatial dispersion and the modification in the spatial profile of the field induced by the carriers density. It can then be proved that the active material can be represented by a single centered QW, provided the oscillator strength is substituted by the effective one

$$f_{eff} = \frac{1}{d} \int f(z) g(z) dz, \tag{9}$$

where $f(z)$ is the actual oscillator strength as obtained from Eqs. (1), (4), and $g(z)$ describes the z dependence of the electric field in the cavity

$$g(z) = (m^{11}_{RBR} + m^{12}_{RBR}) \cos\left[\frac{\omega}{c} n(L/2 - z)\right] - \frac{i}{n}(m^{21}_{RBR} + m^{22}_{RBR}) \sin\left[\frac{\omega}{c} n(L/2 - z)\right], \tag{10}$$

n being the refractive index of the cavity material and L the cavity thickness. Thus the greater active thickness compensates for the reduced oscillator strength of the bulk exciton, and since the saturation intensities are not much different, one can expect a behavior similar to that of QW cavities.

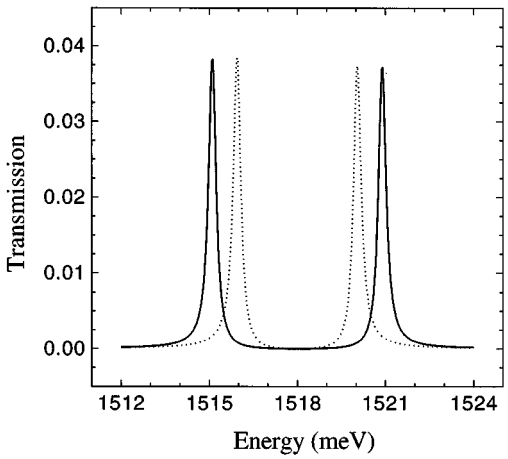

FIG. 1. Transmission spectra of a quantum microcavity with two values of the exciton population: $n = 0$ (solid line) and $n = n_s$ (dotted line). Exciton parameters are $\omega_0 = 1518$ meV, $4\pi f = 3.975 \times 10^{-3}$, and $\Gamma = 0.5$ meV.

III. RESULTS

In order to achieve a bistable condition, the considered material must experience a sudden and deep change in its optical properties, sufficient to give rise to a positive feedback mechanism, when the intensity of the incident light is increased. In the case of resonant excitation in a microcavity sample, such a change can be seen as a consequence of the Rabi splitting bleaching induced by the exciton-photon coupling. As one can see from Fig. 1, the reduction of the Rabi splitting with the exciton population results in a large alteration of the transmission coefficient for frequencies near those of the two split peaks. To probe the bistable behavior, however, we need to check the relation between the electromagnetic field and the exciton population. To this purpose it is useful to introduce a saturation intensity I_s into Eq. (4), giving rise to an exciton population equal to n_s. All intensity variables can be scaled to I_s, thereby leaving all quantitative aspects to the determination of I_s for each particular system. In standard experiments on macroscopic FP bistability with GaAs QW's, a value of I_s equivalent to 600 W/cm^2 is fairly typical under quasistatic excitation [25]. The main difference in our proposed device originates from the carrier lifetime. Instead of slow QW excitons or even free e-h pairs ''cavity excitons,'' with the effective lifetime

$$\tau_{eff} = \frac{\frac{1}{\tau_p} + \frac{1}{\tau_k} + \frac{1}{\tau_m}}{\frac{1}{\tau_0 \tau_p} + \frac{1}{\tau_k \tau_0} + \frac{1}{\tau_k \tau_m}}, \tag{11}$$

control the dynamics. The values of τ_{eff} depend on the QW and microcavity characteristics and can be as low as ~ 1 ps, which means an increase in saturation intensity, at most of two, three orders of magnitude; this can be partially compensated by $\mathrm{Im}(\chi)$ when the chosen operation frequency is

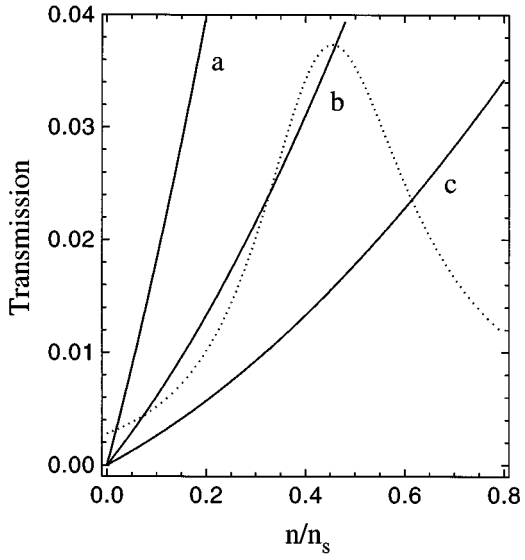

FIG. 2. Transmission at $\omega = 1520.4$ meV of the considered microcavity as a function of the exciton population calculated from formula (6b) (dotted line) and from relation (8) (solid lines). Curve (a) corresponds to an incident intensity $I_{in} = 0.005 I_s$, curve (b) to $I_{in} = 0.015 I_s$, curve (c) to $I_{in} = 0.035 I_s$, where I_s is the saturation intensity as defined in the text. Other parameters as in Fig. 1.

nearer to ω_0. We wish to remark however that the larger saturation intensity required is not an intrinsic drawback of the mechanism proposed; it is simply the expression of a faster dynamical behavior, which therefore requires fast pulsed sources (in [18] a 100 fs laser at some tens of W/cm^2 was found to be sufficient to quench the Rabi splitting). It must be noted in fact that the energy density I_s can be achieved at steady state in a shorter time, allowing the use of a faster pulsed laser with small power flux. Furthermore, as it will be clear in the following, the switch-on intensity for the bistable loop is found to be a much smaller fraction of

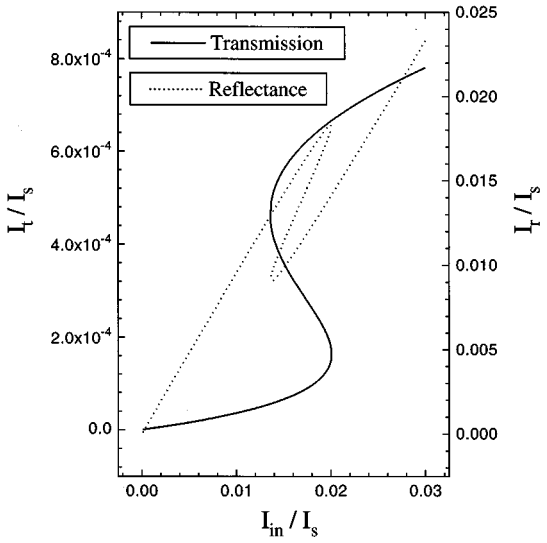

FIG. 3. Transmitted and reflected power of the considered microcavity at $\omega = 1520.4$ meV as a function of incident intensity, computed by simultaneously solving Eqs. (6b) and (8). Parameters as in Fig. 1.

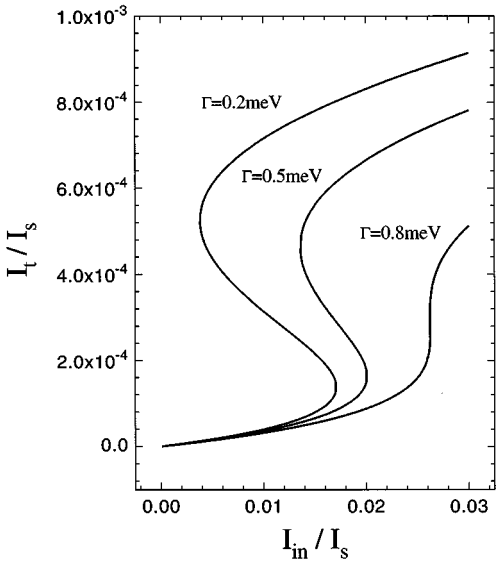

FIG. 4. Transmitted power of our microcavity as a function of the incident intensity for three values of the broadening Γ. Other parameters as in Fig. 3.

the saturation intensity than that of standard macroscopic FP devices. This is related to the possibility of operating in resonance with the polaritonic transitions and proves the overall better power requirements of our system.

To compute the optical constants we must use all the previously derived equations with the parameters relevant to the material under consideration. We report in Fig. 2 the transmission at 1520.4 meV as a function of the exciton population, computed from Eq. (6b); also shown is the transmission computed from Eq. (8) for three different incident intensities. The intersections of the two curves give the actual solutions of our system determining consistently all the electromagnetic field amplitudes. It appears clearly that in case (b), which corresponds to an incident intensity $I_{in} = 0.015 I_s$, three solutions exist, marking the existence of a bistable regime, while in the two other cases only one solution is present. The resulting input-output curves are plotted in Fig. 3, showing the appearance of a well defined hysteresis loop both in transmission and reflectance. We wish to stress that for bistability only a small fraction of the saturation intensity is needed, so that the increase in saturation intensity of the microcavity with respect to a standard FP macroscopic cavity is not a handicap.

In view of technological applications is now particularly relevant to analyze the effect of the damping parameter Γ which we have phenomenologically inserted in the susceptibility expression. Originally the exciton oscillator damping comes from both material defects and temperature broadening and, in general, an increase of the damping factor Γ causes the bistable performances to degrade. This can be seen in Fig. 4, where we plot the transmission as a function of the incident intensity for three different values of the damping parameter. Clearly, both the amplitude of the hysteresis loop and the switching jump of the output are rapidly reduced increasing Γ. Furthermore, the incident power at which the switching occurs is increased as well. The question of the contribution of homogeneous and inhomogeneous broadenings to exciton dephasing processes in QW micro-

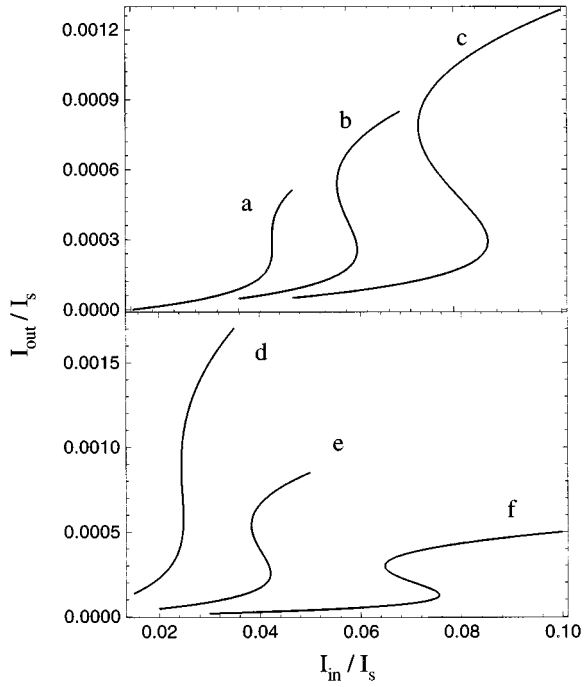

FIG. 5. Transmission input-output curves of our microcavity for three operating energies: (a) 1520.4 meV, (b) 1520.3 meV, (c) 1520.2 meV, and for three different Bragg mirrors configurations: (d) 18 periods of alternating $\lambda/4$ layers, (e) 20 periods, (f) 22 periods ($\omega = 1520.3$ meV). $\Gamma = 0.8$ meV and other parameters as in Fig. 1.

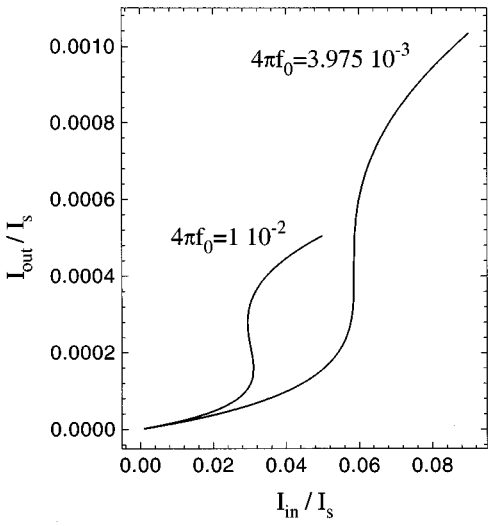

FIG. 6. Transmission input-output curves of the microcavity for two values of the exciton oscillator strength at 0.65 meV below the highest-energy transmission peak; $\Gamma = 1$ meV.

cavities is complicated by the strong coupling with the photonic mode and is still the object of intense investigation [34].

The bistable performances of our microcavity system can be improved by directly modifying some structural parameters. In Fig. 5 we plot the transmission of the microcavity for three frequencies of the incident light in the sensitive region and for three kinds of Bragg mirrors with a different number of $\lambda/4$ layers. It can be noticed that, as the energy of the incident radiation is moved away from the transmission peak, a clearer hysteresis results, with a larger jump, but at a higher excitation intensity. Increasing the number of periods in the Bragg reflectors, and thus the mirror reflectivity, one gets at a given frequency a more evident loop but with a smaller output variation and at higher input powers because of the narrowing of the transmission peaks. A suitable combination of the above effects has to be found in order to optimize the required performance.

Up to now we have examined the case of low temperature. As the temperature increases the exciton ionization time becomes progressively shorter (approximately 300 fs at room temperature [26]) so that a rate equation for the free electron-hole pairs population has to be added to Eqs. (2) [28]. Since the exciton oscillator strength presents a saturating behavior like that of Eq. (1) also as a function of the free-carriers density [24], our results still qualitatively hold, although for free electron-hole pairs remarkable differences in saturation density and dynamical response times lead to a large variation (normally a reduction) of the saturation intensity. Actually qualitative modifications arise from the two terms which in the rate equations represent the dynamical process of the binding of free pairs into excitons (which depends on the square of the free-carriers density) and the free electron-hole pairs recombination, in turn described by a third-order polynomial [28]. Nevertheless, the most important effect of the temperature is to increase the homogeneous broadening, and thus the parameter Γ, owing to the increased phonon-scattering rates. As shown in Fig. 4, this has dramatic consequences on the bistable performances of the microcavity structure.

In order to improve the situation one can think of a greater effective oscillator strength for the excitonic transition, which can be obtained, for instance, by inserting more QW's in the cavity, by choosing another design of the QW or some different material. To illustrate this possibility, we plot in Fig. 6 two transmission input-output curves corresponding to different values of the exciton oscillator strength. We observe that a greater oscillator strength produces a more defined hysteresis loop at a lower incident power. We have also found that a Rabi splitting somewhat larger than the exciton broadening (about three or four times) is needed to get a bistable regime with reasonable powers and structural parameters. In this respect, bulk microcavities [13], which exhibit comparable splittings with very small inhomogeneous broadenings should be advantageous. Of course a dynamical hysteresis will be observed in any case.

A final but interesting aspect regards the dynamics of the switching process. Obviously, for the purpose of device applications fast operation is very important. In our system the rise time is limited in principle only by the time necessary for the excitons to form, which is at most half a Rabi oscillation period (some hundreds fs), while the switch off is governed by the recombination times of the carriers. Therefore, while at room temperature the latter should be greatly influenced by that of free electron-hole pairs (nanoseconds), at low temperature is determined by the excitons radiative lifetime, which varies from less than 1 ps for cavity-coupled excitons to several tens of picoseconds for the uncoupled ones. The momentum scattering rates control which of these

two radiative times is the most relevant. Switch off times may be somewhat improved choosing an appropriate structure in order to enhance the exciton decay rate. A notable improvement would result from decreasing the exciton radiative lifetime below the ionization one, in order to inhibit the creation of slowly recombining free carriers; however, since this also results in a considerable increase of the peaks linewidths, a practical compromise has to be found.

IV. CONCLUSIONS

We have proved with a fairly simple theoretical model that the bleaching of the exciton Rabi splitting in semiconductor microcavities leads to an optically bistable regime in the frequency region of the exciton resonance. This process is shown to be advantageous, for switching times and comparatively low intensity required, with respect to that of macroscopic FP cavities. Using a transfer-matrix approach for the electromagnetic field and a saturation behavior for the exciton oscillator strength, we have shown that the transmitted and reflected signal may present a static hysteresis loop as a function of the incident power. A careful analysis of the possible ways to control the bistable operation has been carried out in view of performance optimization, and the effect of the exciton broadening has been addressed. In particular, it has been shown that the increase of the damping parameter has dramatic consequences on the amplitude of the hysteresis loop and on the required incident powers.

ACKNOWLEDGMENTS

The authors are indebted to G. La Rocca, L. C. Andreani, and V. Agranovich for stimulating discussions. They are also grateful to V. Savona for a critical reading of the manuscript. Financial support from the European Community through Contract No. CHRX-CT94-0464 is gratefully acknowledged.

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