Vacuum-field Rabi splitting in the presence of inhomogeneous broadening: Resolution of a homogeneous linewidth in an inhomogeneously broadened system

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(Received 28 August 1995)

We study the effect of inhomogeneous broadening of the electronic state on vacuum-field Rabi splitting. The broadening has no effect on the size of the splitting and, in general, does not lead to an inhomogeneous broadening of the split states. From a spectroscopic point of view, these results have interesting consequences, since they allow the extraction of a homogeneous line in an inhomogeneously broadened system.

PACS number(s): 42.50.Ct, 71.36.+c

When two discrete oscillators are coupled together strongly enough they develop two normal modes and exhibit an anticrossing behavior: They are said to be in the strong-coupling regime. This is a general denomination in physics, and both oscillators can be very different in nature. This regime has also been observed in quantum optics between an electronic oscillator and a single-photon mode in an optical cavity, first in atomic physics [1] and lately in solid-state physics [2]. This effect is called vacuum-field Rabi splitting (VRS) in atomic physics or cavity-polariton splitting in microcavity solid-state physics. Although inhomogeneous broadening of the atomic or electronic oscillator is not a source of concern in atomic physics because of the very low atom density, it has been mentioned several times in solid-state physics [3–5]. In this paper we want to address the question of the vacuum-field Rabi splitting (or strong-coupling) regime of an inhomogeneously broadened electronic state. The issue is this: If we let Ω be the one-atom VRS, is the VRS for an inhomogeneous set of n electronic oscillators equal to the collective VRS of n atoms, inhomogeneously broadened, or equal to the incoherent superposition of the one-oscillator VRS [Fig. 1(a)]? This last option was proposed to explain the very small VRS (0.3 meV instead of 3 meV) observed in some semiconductor microcavities [4]. We will show that none of these hypotheses are entirely correct and that the answer is in exact analogy with the case of the VRS in an inhomogeneous vacuum field, as already described by Haroche [6]. The inhomogeneous broadening has no effect on the peak separation of the splitting and, in general, does not lead to an inhomogeneous broadening of the split states. This has interesting consequences from a spectroscopic point of view, as it allows the extraction of a homogeneous line in an inhomogeneously broadened system by the means of cw measurements. The result will be derived both from an atomic physics quantum model and a linear dispersion model.

Let us start with the well-described one- or collective n-atom VRS: The total Hamiltonian H_T of the atom plus photon system is the sum of (i) a two-level atomic oscillator, of energy $\hbar\omega_{\rm at}$ which is described by a spin-$\frac{1}{2}$-type Hamiltonian; (ii) a single-photon mode, of energy $\hbar\omega_{\rm ph}$, (harmonic oscillator, with an infinite series of $|m\rangle$ states with $m=0,1\ldots$ photons); (iii) an interaction Hamiltonian that mixes photon and atom operators [7]

$$H_T=\tfrac{1}{2}\hbar\omega_{\rm at}(c^\dagger c-cc^\dagger)+\tfrac{1}{2}\hbar\omega_{\rm ph}(a^\dagger a+aa^\dagger)$$
$$-\hbar g(ac^\dagger+a^\dagger c)\quad\text{with}\quad \hbar g=d\sqrt{\hbar\omega_{\rm ph}/\epsilon_0 V_{\rm cav}},$$

where c, a and $c^\dagger$, $a^\dagger$ are the atom and photon annihilation and creation operators, $V_{\rm cav}$ the effective volume of the cavity, and d the electric-dipole matrix element. At resonance ($\omega_{\rm ph}\approx\omega_{\rm at}$) and in the absence of interaction ($g=0$), $|e\rangle|m\rangle$ is degenerate with the $|g\rangle|m+1\rangle$ state. Turning on the light-matter interaction ($g\neq0$) lifts the degeneracy. At exact resonance, the eigenstates are a symmetric and antisymmetric combination of the uncoupled states, with energy separation $E_{|+\rangle|0\rangle}-E_{|-\rangle|0\rangle}=\hbar\Omega_{\rm Rabi}$:

$$|\pm,0\rangle_{\delta=0}=\frac{1}{\sqrt{2}}\,(|e\rangle|m\rangle\pm|g\rangle|m+1\rangle),$$

$$\hbar\Omega_{\rm Rabi}=2d\sqrt{\hbar\omega_{\rm ph}/\epsilon_0 V_{\rm cav}}\sqrt{1+m}.$$

The case of several identical, indistinguishable atomic oscillators is described using Dicke states [8] for the collective excitation. The collective excited states are

$$|G\rangle=|g\cdots g\rangle,$$

$$|E_1\rangle=\frac{1}{\sqrt{n}}\,C^\dagger|g\cdots g\rangle=\frac{1}{\sqrt{n}}\sum_{i=1}^{n}|g\cdots e_i\cdots g\rangle|0\rangle,$$

$$|E_2\rangle=\sqrt{2/n(n+1)}(C^\dagger)^2|g\cdots g\rangle$$

$$\vdots$$

where $C^\dagger$ is the collective creation operator, $C^\dagger=\Sigma c_i^\dagger$. The degeneracy of the manifolds increases very quickly with order but the first singly excited manifold consists of only two states, in exact analogy with the one-atom case. The result is the same as for the one-atom case, replacing $|g\rangle$ with $|G\rangle$ and $|e\rangle$ with $|E_1\rangle$; the energy separation is

$$\hbar\Omega_{\rm Rabi}^{m=1}=2\sqrt{n}\hbar g=\sqrt{n}\hbar\Omega_{\rm Rabi}^{n=m=1}.$$

It can be shown that the case of coupling to an inhomogeneous vacuum field [9] results in a quadratic averaging of the Rabi splitting but *not* of the spectral line position; i.e., the

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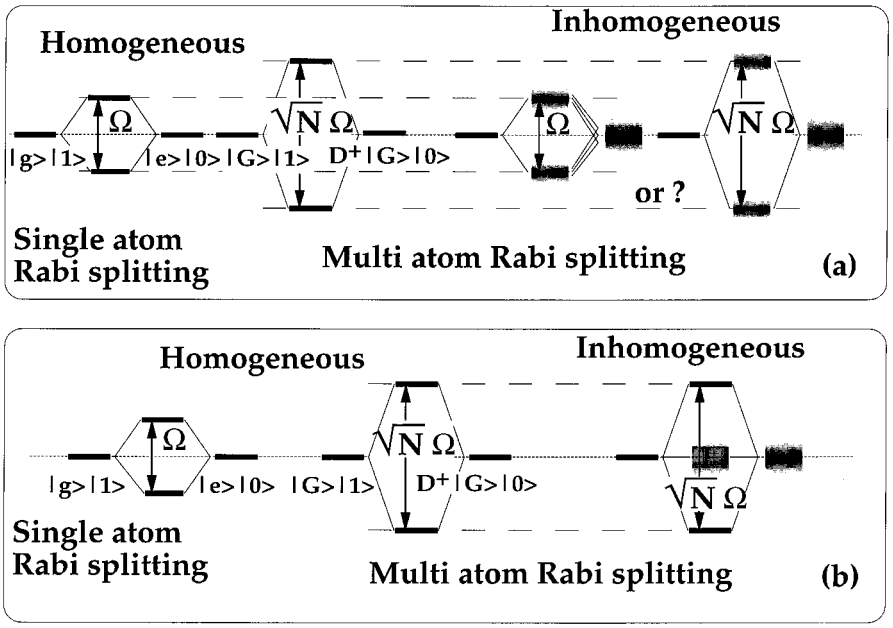

FIG. 1. Energy diagrams of the coupled photon-electronic states of the multiatom Rabi splitting. (a) The case of homogeneous broadening and possible effects on an inhomogeneous broadening as proposed by several authors. (b) Effect of inhomogeneous broadening from both quantum electrodynamics and the classical model.

linewidth of the split line is determined by the homogeneous linewidth and not by the inhomogeneous broadening of Ω [6].

In the case of an inhomogeneously broadened system, one has to consider a set of n distinguishable electronic oscillators; this implies that each $|g\cdots e_i\cdots g\rangle$ state has to be considered separately. The total Hamiltonian becomes

$$H_T=\sum_{i=1}^{n}\tfrac{1}{2}E_i(c_i^\dagger c_i-c_1c_1^\dagger)+\tfrac{1}{2}E_0(a^\dagger a+aa^\dagger)-\sum_{i=1}^{n}\hbar g_1(ac_1^\dagger + a^\dagger c_i),$$

with E_0 the uncoupled photon mode energy and E_i the uncoupled electronic oscillator energy, ordered so that $E_i<E_{i+1}\ \ \forall i=1,\ldots,n-1$. We consider only the case of the first manifold, which is $(n+1)$-fold degenerated. It is instructive to start with nongeneral cases.

(a) At resonance $[E_0=E_i=E_j\ \ \forall(i,j)]$ and with $g=g_i=g_j\ \ \forall(i,j)$; the eigenenergies are the solutions of

$$D_n(\lambda)=(E_0-\lambda)^{n-1}[(E_0-\lambda)^2-n\hbar^2g^2]=0,$$

which gives two split eigenstates:

$$|\pm,0\rangle=\frac{1}{\sqrt{2}}\left(|g\cdots g\rangle|1\rangle\pm\frac{1}{\sqrt{n}}\sum_{i=1}^{n}|g\cdots e_i\cdots g\rangle|0\rangle\right)$$

$$=\frac{1}{\sqrt{2}}\left(|g\cdots g\rangle|1\rangle\pm\frac{1}{\sqrt{n}}C^+|g\cdots g\rangle|0\rangle\right)$$

with energy separation

$$\hbar\Omega_n=2\sqrt{n}\hbar g=\sqrt{n}\hbar\Omega_1$$

and $(n-1)$-fold degenerate states at $E=E_0$. Note also that (i) the *nature* of the central states is very different from the two split states, as they are only a linear combination of $|g\cdots e_i\cdots g\rangle|0\rangle$ states; i.e., there is no mixing with the $|G\rangle|1\rangle$ state; (ii) the split states are well separated from the $n-1$ remaining states and are formally the same as in the homogeneous case. Inhomogeneous broadening is therefore not expected. (This point will be discussed again in the linear dispersion model.)

(b) Still at resonance and if $E_0=E_i=E_j$ but with inhomogeneous coupling $(g_i\neq g_j)$, the eigenvalue equation becomes

$$D_n(\lambda)=(E_0-\lambda)^{n-1}\left[(E_0-\lambda)^2-\hbar^2\sum_{i=1}^{n}g_i^2\right]=0.$$

With the solution

$$|\pm,0\rangle=\frac{1}{\sqrt{2}}\left(|g\cdots g\rangle|1\rangle \pm\sum_{i=1}^{n}\frac{g_i}{\sqrt{\sum_{j=1}^{n}g_j^2}}|g\cdots e_1\cdots g\rangle|0\rangle\right),$$

$$\bar{\Omega}=2\hbar\left(\sum_{i=1}^{n}g_i^2\right)^{1/2}$$

and $(n-1)$-fold degenerate states at $E=E_0$. $\bar{\Omega}$ is the quadratic averaging of the individual coupling constant, as in the case of the inhomogeneous vacuum-field Rabi splitting.

(c) In a general case ($E_i\neq E_j$ and $g_i\neq g_j$), equations and solutions become

$$D_n(\lambda)=(E_n-\lambda)D_{n-1}(\lambda)-\hbar^2g_n^2\prod_{k=1}^{n-1}(E_k-\lambda)$$

$$=\prod_{i=1}^{n}(E_i-\lambda)\left((E_0-\lambda)-\sum_{j=1}^{n}\frac{\hbar^2g_i^2}{(E_j-\lambda)}\right)=0,$$

and the eigenvalues equation is

$$\sum_{j=1}^{n} \frac{\hbar^2 g_j^2}{(E_j-\lambda)} - (E_0-\lambda) = 0;$$

near resonance, $E_i = E_0 + \epsilon_i$, $\epsilon \ll E, \hbar g_i$, $i = 1, \ldots, n$:

$$E_{\pm} = E_0 \mp \hbar \left(\sum_{i=1}^{n} g_i^2 \right)^{1/2} - \frac{1}{2} \frac{\sum_{i=1}^{n} \epsilon_i g_i^2}{\sum_{i=1}^{n} g_i^2} + O(\epsilon^2).$$

The n-1 remaining states, of energy λ_i are spread within the energy bandwidth of the n initially uncoupled states; more precisely,

$$\lambda_i \in [E_i, E_{i+1}] \quad \forall i = 1, \ldots, n-1.$$

The answer to Fig. 1(a) is summarized in the diagram of the energy levels of the coupled photon-electronic states in Fig. 1(b),

Cases (a) or (b) correspond to the coupling of a Fabry-Pérot (FP) mode with a much larger mode size [10] than the electronic oscillator coherence area or localization length. This would describe pure spatial inhomogeneities and $n \approx$(localization area or coherence length)/(FP mode size). Case (c) corresponds to a FP mode coupled to a spectrally broadened electronic oscillator. It follows that (i) the splitting is unaffected compared to the equivalent coherent one, and the coupled eigenstates are identical. This result is independent of the origin (spatially or spectrally) of the inhomogeneous broadening. (ii) The coupled modes have a coherence area (or a spatial extension) of the order of the average of the electronic oscillator and the FP mode sizes [11], and, therefore an incoherent summation over each electronic state [5] would only be valid for experimental setup, where the probe beam is much larger than the electronic coherence area *and* the FP mode size [12]. Moreover, although a reduction of the Rabi splitting in semiconductor microcavities could be due to coupling to bound excitons, which have a weaker oscillator strength, (but a narrow linewidth), it cannot account for a band of localized excitons because the oscillator strength per unit surface for the whole excitonic band remains constant, independently of its localized or delocalized nature [13].

Analogous results can be obtained from a classical model. Zhu *et al.* [14] demonstrated that linear dispersion theory can be used to describe vacuum-field Rabi splitting. The cavity is modeled by the standard Airy description of a FP cavity and the two-level atomic system by a Lorentz oscillator dispersive dielectric constant [14]

$$\epsilon(\nu) = n(\nu)^2 = \epsilon_\infty + \frac{N f_0 e^2}{m \epsilon_0} \frac{1}{\nu_0^2 - \nu^2 - i\gamma\nu},$$

where f_0 is the oscillator strength, e (m) the charge (mass) of the electron, N the oscillator density, ν_0 the resonance frequency, and γ the oscillator homogeneous linewidth. This model neglects the multiple reflections at the oscillator-cavity interface and the location of the oscillators with respect to the node of the optical field in the cavity. This approximation is valid for microcavities in atomic physics and can easily be extended to semiconductor systems [15]. The

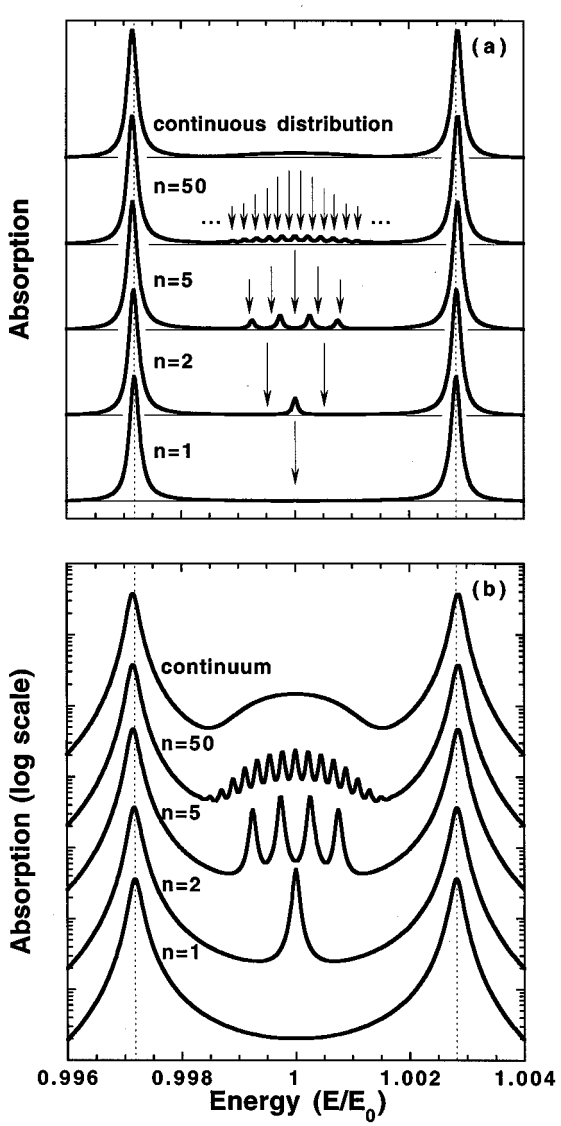

FIG. 2. Absorption spectrum of vacuum-field Rabi splitting for a set of 1, 2, 5, and 50 independent electronic oscillators and a continuous Gaussian distribution, where the total integrated absorption is kept constant; (a) linear and (b) logarithmic scales. Parameters are $\gamma_c/E_0 = 3.2\ 10^{-4}$, $\gamma/E_0 = 1\ 10^{-4}$, and $\sigma/E_0 = 1\ 10^{-3}$. Arrows indicate energies of the uncoupled electronic oscillators.

splitting given in [14] is the *transmission* line splitting. It is more appropriate to use the *absorption* line splitting, which is

$$\Omega = \sqrt{\Omega_{\max}^2 - (\gamma_c^2 + \gamma^2)/2} \quad \text{and} \quad \Omega_{\max} \propto \sqrt{N f_0 / L_c}$$

with the linewidth $\Delta_{\pm} = (\gamma_c + \gamma)/2$, where L_c is the cavity length and γ_c the FP mode linewidth. The simple physical picture is that in order to form a FP resonance, the round-trip phase shift has to be an integer multiple of 2π. Because of the form of the real part of the Lorentz oscillator refractive index, the round-trip phase shift vs photon energy becomes N-shaped, up to a point where the phase shift conditions are fulfilled three times. This gives rise to the doublet structure because the central solution, which also corresponds to a maximum of absorption, does not create a FP resonance.

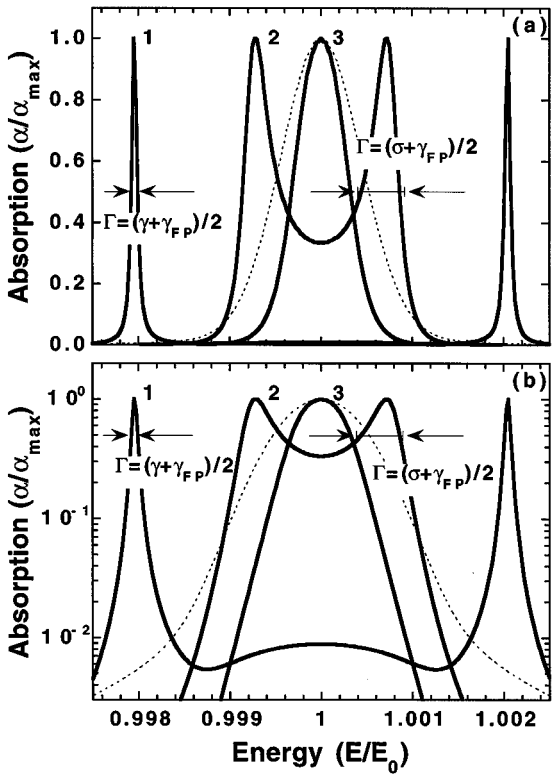

FIG. 3. Absorption spectrum of vacuum-field Rabi splitting (Ω) for an inhomogeneously broadened system. Dashed line: absorption spectrum of the uncoupled electronic oscillator (plasma dispersion function), $\sigma=\gamma_{\text{inhom.}}$, and $\gamma=\gamma_{\text{hom.}}$; (1) strong interaction energy ($\Omega\gg\sigma$), (2) moderate interaction energy ($\Omega\approx\sigma$), and (3) small interaction energy, weak-coupling regime; (a) linear and (b) logarithmic scales. Parameters are $\gamma_c/E_0=3\ 10^{-5}$, $\gamma/E_0=1\ 10^{-4}$, and $\sigma/E_0=1\ 10^{-3}$; the relative coupling strengths are 50(1), 5(2), and 1(3).

Extension of this method to a set of nonidentical oscillators is performed by replacing ϵ by

$$\epsilon(\nu)=n(\nu)^2=\epsilon_\infty+\begin{cases}\sum_i \epsilon_i(\nu)\\ \text{or}\\ \int_{-\infty}^{+\infty}\epsilon(\nu,\nu_0)g(\nu_0)d\nu_0,\end{cases}$$

where $g(\nu_0)$ is the spectral density of oscillators at the frequency ν_0.

Simulations are presented for a set of 1,2,5,50 independent electronic oscillators and a continuous Gaussian (full width at half maximum σ) distribution, while keeping the total integrated absorption constant [i.e., reducing $f_0(\nu_0)$ so that $\int_{\pm\infty}\alpha\ de=\text{const}$]. Results are shown in Figs. 2 and 3. The absorption spectrum exhibits two main lines and residual structures of lower optical activity at the resonance energy [16]. This is the classical analog of the QED model. The existence and the peak separation of the splitting in absorption is independent of the homogeneous or inhomogeneous nature of the electronic oscillator. This can easily be understood, considering that no specific distinction between inhomogeneous and homogeneous lines are made in this linear dispersion model. The refractive index n is only a function of the integrated absorption via the Kramers-Kroenig transformation, T_{KK} which is linear [i.e., it does not matter where the summation is made; $n=T_{\text{KK}}(\Sigma\alpha)=\Sigma T_{\text{KK}}(\alpha)$]. Figure 3 illustrates that for large enough VRS the linewidth is given by the *homogeneous* linewidth. As can be seen when increasing the interaction energy, the linewidth $\Delta_\pm$ of the Rabi split lines decreases from $(\sigma+\gamma_{\text{ph}})/2$ to $(\gamma+\gamma_{\text{ph}})/2$. This can be understood from a property of the plasma dispersion function [17] (i.e., the convolution of a Gaussian and a Lorentz function), stating that the central energy region has a Gaussian shape while out in the wings ($\nu-\nu_0\gg\sigma$) the function has a Lorentzian shape (see Fig. 2, log scale). As the linewidth is determined by the slope of the round-trip phase shift vs energy function, this explains the linewidth reduction. Alternatively, in Fig. 3, for a given coupling strength we would observe that the disappearance of a doublet structure occurs at very similar values when increasing the homogeneous or inhomogeneous linewidth.

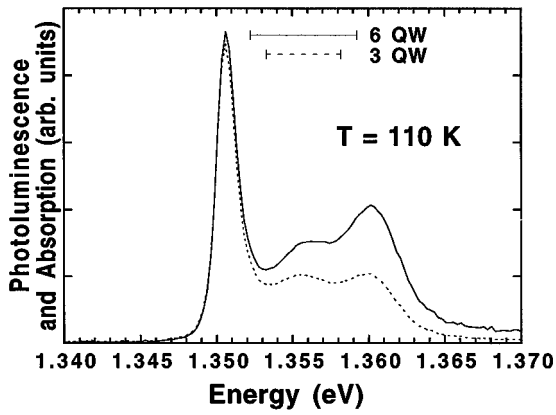

FIG. 4. Absorption spectrum, as deduced from a photoluminescence spectrum (dashed line), of a semiconductor microcavity consisting of two dielectric GaAs/$Al_{0.1}Ga_{0.9}$As Bragg mirrors separated by a 3λ/2 GaAs cavity with six (In,Ga)As imbedded quantum wells was grown. The six quantum wells (QW) were grown with two slightly different thicknesses. Horizontal, continuous (dashed) bar: calculated splitting for a homogeneous case of six (three) QW.

To demonstrate this effect, a semiconductor microcavity consisting of two dielectric GaAs/$Al_{0.1}$ $Ga_{0.9}$As Bragg mirrors separated by a 3λ/2 GaAs cavity with six (In,Ga)As imbedded quantum wells was grown. Similar samples are extensively described in previous communications [18]. The six quantum wells (QW) were (accidentally) grown with inhomogeneous thickness; i.e., two slightly different thicknesses were used, and the excitonic line of both sets of QW differs by a few meV, on the order of the excitonic linewidth of a single QW. Figure 4 shows an absorption spectrum taken close to the resonance condition. Several features in accordance with the theoretical predictions have been observed: (i) There exist three lines (Fig. 4). (ii) As a function of the detuning, the two sideband lines exhibit an anticrossing behavior, while the central line shows little change in energy position. (iii) The splitting of the two sideband lines is comparable to the expected splitting from a homogeneous case of six QW (horizontal bar in the figure) and not three (dashed horizontal bar). That the low-energy line is much

more intense than the other lines can be due to the fact that the measurement was performed at a slightly detuned resonance condition, the existence of cavity pulling [19,20], or finally temperature effects that tend to enhance the low-energy line [20]. A sharp decrease of the linewidth from $(\sigma+\gamma_{ph})/2$ to $(\gamma+\gamma_{ph})/2$ could not be observed in the samples that were investigated because the interaction energy is not large enough, compared to the inhomogeneous linewidth. Nevertheless, trends have been observed by T. A. Fisher and co-workers who reported [21] a linewidth narrower than $(\sigma+\gamma_{ph})/2$. Such an effect could be observed under high magnetic field [22] with GaAs QW at He temperature, where the homogeneous linewidth becomes very narrow (below 1 meV) and the magnetic field allows a continuous increase of the normal mode splitting.

In conclusion we have shown that the peak separation and the existence of the vacuum-field Rabi splitting is independent of the nature (inhomogeneous or homogeneous) of the broadening of the electronic state and that, in general, the linewidth of the Rabi split states is determined by the homogeneous linewidths of both oscillators. From the nature of the split eigenstates, it follows that the Rabi splitting occurs from a collective contribution of the whole inhomogeneous band of electronic state and *not* from a sharp selection of the state exactly resonant with the photon mode [3,4]. From a spectroscopy point of view, these results have interesting consequences as they allow the extraction of a homogeneous line in an inhomogeneously broadened system, by the means of cw measurements. A further critical question that remains to be addressed is how quickly inhomogeneous broadening will smear out the coherent effects expected in the strong coupling regime compared to the way it alters the splitting observed in the optical response or in photoluminescence.

We would like to acknowledge fruitful discussions with C. Weisbuch, V. Savona, F. Tassone, and L. C. Andreani. This work was supported by EPFL (Switzerland), the SMILES EEC Esprit program, and the Swiss national priority program for optics.

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$$|G\rangle|1\rangle(1/\sqrt{n})\sum_{i=1}^{n}|g\cdots e_i\cdots g\rangle|0\rangle,$$

$$\left\{ i=2,\ldots,n,[1/\sqrt{i(i-1)}]\left[\sum_{j=1}^{i}|g\cdots e_j\cdots g\rangle|0\rangle - (i-1)|g\cdots e_i\cdots g\rangle|0\rangle\right]\right\}.$$

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