Motional narrowing of inhomogeneously broadened excitons in a semiconductor microcavity: Semiclassical treatment

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The narrowing of the exciton features in the optical spectra of quantum microcavities usually referred to as a motional narrowing is described within a simple semiclassical approach taking into account the exciton inhomogeneous broadening and assuming wave-vector conservation in the plane of the cavity. Good agreement with existing experimental data is achieved. The narrowing of exciton-photon modes in calculated spectra, however, cannot be called ''motional'' since no in-plane motion or quantization of excitons was assumed in the model. [S0163-1829(98)04704-3]

The narrowing of a spectral line in a disordered system due to some averaging process is usually referred to as ''motional narrowing.'' More specifically, for a quantum particle motional narrowing arises due to its necessarily finite mass which prevents its full localization in a fluctuation potential.[1] The distribution function of a quantum particle in the presence of potential fluctuations is therefore narrower than that of an ideal classical particle. To observe the ''motional narrowing'' effect, strictly speaking, one should be able to change the mass of the particle keeping the disorder potential constant. Recent experimental and theoretical works[2,3] discussed the possibility of direct observation of the exciton-polariton motional narrowing in semiconductor microcavities.

Semiconductor microcavities with embedded quantum wells (QW's) intensively studied in the 1990's (Refs. 4–6) represent an excellent model systems for the study of exciton photon coupling in a two-dimensional system. In the strong coupling regime, anticrossing of the exciton and confined photon modes result in the appearance of two polariton branches split by about 5–8 meV. The cavity photon mode has a nearly parabolic in-plane dispersion and can be described by an effective mass which is usually a few times smaller than the exciton mass. The idea of Whittaker *et al.*[2] was that tuning of the cavity photon mode to the exciton resonance results in the decrease of the effective masses of mixed exciton-polariton states which causes the additional narrowing of spectral lines detected experimentally. Thus, the narrowing of exciton-polariton modes at the anticrossing point has been interpreted as a manifestation of ''motional narrowing.''

The concept of motional narrowing requires the nonconservation of the exciton-polariton wave vector in the plane of the QW which is a necessary condition for the localization of quantum particles. Savona *et al.*[3] developed a microscopic model taking into account the in-plane scattering of excitons which yielded a good agreement with experiment. An asymmetric narrowing of cavity polariton modes has been attributed in Ref. 3 to the wave-vector nonconservation in the system and considered as a manifestation of motional narrowing.

Solving the excitonic problem in k space one should necessarily consider the multiple exciton-polariton scattering to all orders to account for the exact eigenstates of the system.[3] Solving the same problem in real space one does not need *a priori* the in-plane scattering but can just suppose some energy distribution of exciton resonances. Thus, the physical interpretation of the narrowing of cavity modes as a motional narrowing seems to me still questionable. Actually, the wave-vector non-conservation leads to the resonant Rayleigh scattering of light in a plane of the QW.[7–10] There have been indications that the fraction of photons scattered in the plane is not more than 0.1% of the total number of reflected photons for planar semiconductor structures.[11–13] The contribution of scattered photons to the cw reflection spectrum is negligible in this case and cannot cause substantial modifications to the resonant spectral features. Evidently, the narrowing of cavity polariton modes is a consequence of exciton inhomogeneous broadening. However, I do not see clear evidence that it is due to the motional narrowing and cannot be simply caused by a frequency distribution of extended exciton states conserving the wave vector. The latter possibility is considered theoretically in the present paper.

A semiclassical, macroscopic approach to the problem of light coupling with inhomogeneously broadened exciton state in a QW has been formulated in Ref. 13. The approach consists in the solution of Maxwell equations for light incident on the QW taking into account exciton inhomogeneous broadening in the nonlocal dielectric susceptibility of the QW and assuming in-plane wave-vector conservation. Here I use an extended version of this model. Its key points are as follows.

In the framework of linear nonlocal response theory,[14] Maxwell equations for a light wave normally incident on a single quantum well in the vicinity of the exciton resonance frequency can be written in the form

$$\nabla\times\nabla\times\mathbf{E}=\frac{\omega^2}{c^2}\mathbf{D}, \tag{1}$$

where

$$\mathbf{D}(z)=\epsilon_\infty\mathbf{E}(z)+4\pi\mathbf{P}_{\text{exc}}(z), \tag{2}$$

$$\mathbf{P}_{\rm exc}(z)=\int \chi(z,z')\mathbf{E}(z')dz', \tag{3}$$

and the nonlocal susceptibility is expressed as

$$\chi(\omega,z,z')=\chi(\omega)\Phi(z)\Phi(z'), \tag{4}$$

where $\Phi(z)$ is the exciton envelope function taken with equal electron and hole coordinates, and z is the normal to QW plane direction. Taking into consideration only the ground exciton state, one can write in the absence of inhomogeneous broadening

$$\chi(\omega-\omega_0)=\frac{\epsilon_\infty\omega_{\rm LT}\pi a_B^3\omega_0^2/c^2}{\omega_0-\omega-i\gamma}, \tag{5}$$

where ω_0 is the exciton resonance frequency, γ is its nonradiative homogeneous broadening, $\omega_{\rm LT}$ is the exciton longitudinal-transverse splitting in the bulk material, and a_B is the bulk exciton Bohr radius.

Supposing that due to inhomogeneous broadening the exciton resonance frequency is described by some distribution function $f(\nu-\omega_0)$ and neglecting the possible dependence of the exciton envelope function on energy one should substitute the resonant dielectric susceptibility (5) by the function

$$\tilde{\chi}(\omega)=\int d\nu\chi(\omega-\nu)f(\nu-\omega_0). \tag{6}$$

The amplitude reflection ant transmission coefficients for light normally incident on the QW can be written as

$$r=\frac{i\alpha\tilde{\chi}}{1-i\alpha\tilde{\chi}},\quad t=1+r, \tag{7}$$

respectively, where $\alpha=\Gamma_0c^2/\varepsilon_\infty\omega_{\rm LT}\pi\omega_0^2a_B^3$. Γ_0 is the exciton radiative rate,

$$\Gamma_0=\frac{k\omega_{\rm LT}\pi a_B^3}{2}\left[\int \Phi(z)\cos kz dz\right]^2=\frac{\pi}{\sqrt{\epsilon_\infty}}\frac{e^2}{mc}\frac{f_{xy}}{S}, \tag{8}$$

in terms of the oscillator strength per unit area f_{xy}/S. k is the wave vector of light in the media.

In Ref. 13 we only considered symmetric distribution functions f (Gaussian, Lorentzian, etc.). In the present work the distribution function is chosen in a form which allows asymmetry; namely,

$$f(x)=\frac{\Delta_1+\Delta_2}{2\sqrt{\pi}\Delta_1\Delta_2}\exp\{-x^2/[(\Delta_1^2-\Delta_2^2)\Theta(x)+\Delta_2^2]\}, \tag{9}$$

where $\Theta(x)$ is the heavyside function.

The convenient way to obtain the complex eigenfrequencies of cavity polaritons within the present approach is to solve the dispersion equation for cavity polaritons written in terms of amplitude reflection and transmission coefficients.[15,16] In the case of a symmetric λ cavity with an embedded QW in the center it has the form

$$r_B(t+r)\exp(ikL_c)=1, \tag{10}$$

where r_B is the amplitude reflection coefficient of the Bragg mirror and L_c is the cavity length. Substituting the coefficients r and t from Eq. (7) and using the approximate formula suggested in Ref. 17,

$$r_B=\sqrt{R}\exp\left[i\frac{n_cL_{DBR}}{c}(\omega-\omega_c)\right], \tag{11}$$

with n_c being the refractive index in the cavity, L_{DBR} the characteristic length of the mirror, and ω_c the stop-band center, one can finally derive the dispersion equation in the following form:

$$(\omega_c-\omega-i\gamma_c)=V^2\int d\nu\frac{f(\nu-\omega_0)}{\nu-\omega-i\gamma}, \tag{12}$$

where $V^2=(1+\sqrt{R})c\Gamma_0/\sqrt{R}n_c(L_c+L_{DBR})$, $\gamma_c=[(1-\sqrt{R})/\sqrt{R}](cn_c(L_c+L_{DBR}))$. In the case of no inhomogeneous broadening, the distribution function f reduces to the δ function, and Eq. (12) reduces to the conventional two-coupled oscillator problem.

The present model is similar to the approach developed by Savona and Weisbuch[18] which also assumed wave-vector conservation in the plane and frequency distribution of the exciton resonance, which was described by some distributed coupling constant in their case. An essential difference between the two models consists in the chosen form of the distribution function for the exciton resonance frequency (or coupling constant): in Ref. 18 the symmetric Gaussian distribution was assumed while here we use a more general asymmetric function (9).

In the numerical calculations a GaAs cavity contained between two GaAs/GaAlAs Bragg reflecting mirrors with three embedded closely lying InGaAs QW's were considered. The chosen structure parameters corresponded to the sample of Whittaker *et al.*[2,19] For all calculated spectra, $\Gamma_0=$ 0.026 meV, $\gamma=0.3$ meV, $\Delta_1=2$ meV, $\Delta_2=1$ meV was assumed. The reflectivity of microcavity structures with embedded quantum wells was calculated using the transfer matrix technique as described in Refs. 15,16.

Figure 1 shows the calculated reflection spectra for different detunings between the exciton resonance and the cavity mode. As in the experiment, the exciton resonance was tuned through the cavity mode. The positions of the bare exciton and cavity photon resonances are shown by dashed lines. The splitting between polariton branches and energies of resonances are in good agreement with the experimental data (Fig. 1 of Ref. 2).

Figure 2 shows the calculated and experimental (data taken from Ref. 2) linewidth [full width at the half-maximum (FWHM)] as a function of the detuning for upper and lower polariton branches. The detuning is defined as the difference between the bare cavity mode and exciton energies. One can see that the calculation exhibits both tendencies interpreted in Refs. 2,3 as manifestations of motional narrowing, namely, the sum of the line widths of the lower and upper branches is clearly much smaller at the anticrossing than in off-resonant conditions, and the behavior of the lower and upper branches is strongly asymmetric. While the calculation does not show such an excellent fit to the data as the model of Savona *et al.* does, within the present approach it seems evident that the experimental data can be reproduced using a

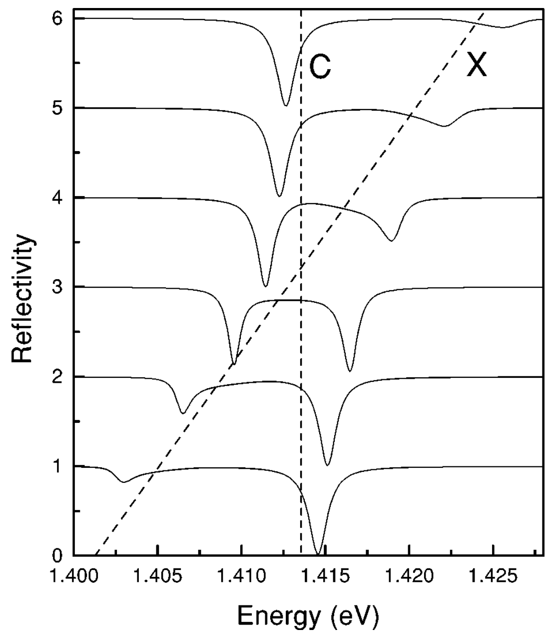

FIG. 1. Reflectivity spectra of the microcavity for different detunings between exciton and cavity modes. Dashed lines show the positions of bare exciton and cavity photon modes.

somewhat more complicated distribution function f and taking into account the variation of the homogeneous broadening as one tunes the temperature.

To obtain the narrowing of lines at the resonance one does not even need an asymmetric distribution function f. The dashed lines in Fig. 2 show the calculated linewidths for $\Delta_1 = \Delta_2 = 1.5$ meV. The sum of FWHM's for two polariton branches at zero detuning is 2.14 meV, while at the detuning

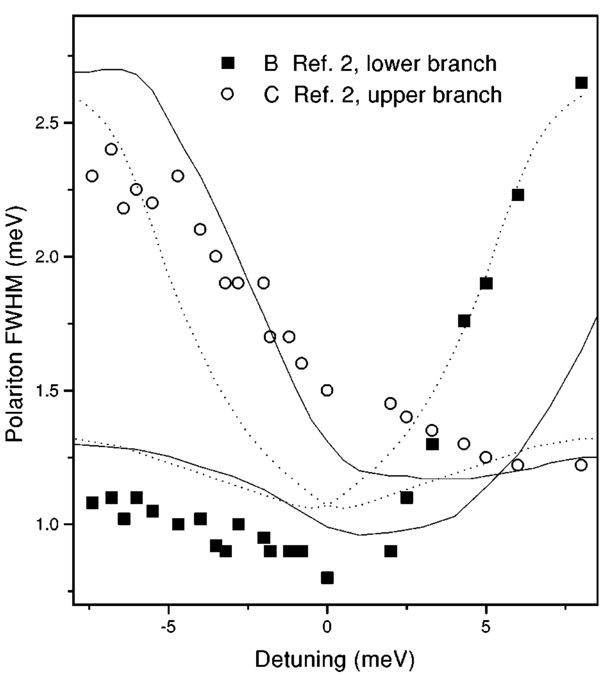

FIG. 2. Experimental (points) and calculated (lines) widths of upper and lower exciton-polariton resonances (FWHM) as a function of the detuning between cavity and exciton resonances. Solid lines were calculated with $\Delta_1 = 2$ meV, $\Delta_2 = 1$ meV; dashed lines were calculated with $\Delta_1 = \Delta_2 = 1.5$ meV.

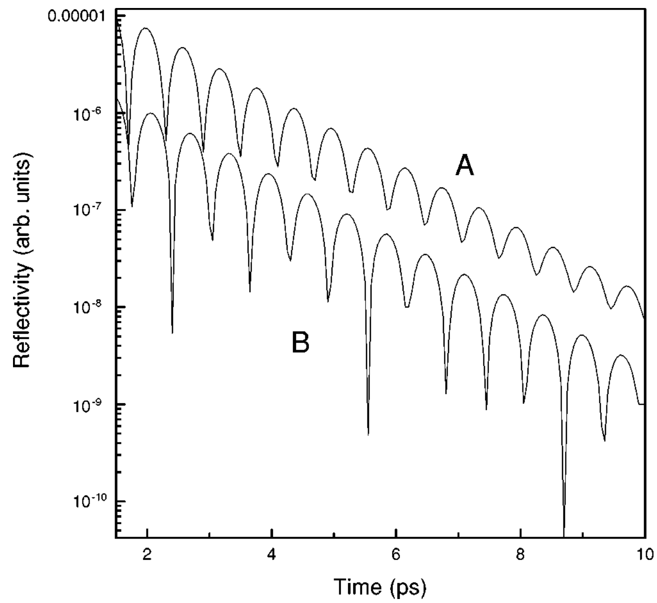

FIG. 3. Calculated time-resolved reflection spectra of the microcavity illuminated by a δ pulse of light at the anticrossing condition for $\Delta_1 = 2$ meV, $\Delta_2 = 1$ meV (A) and $\Delta_1 = \Delta_2 = 1.5$ meV (B).

of 7.5 meV this sum is 3.87 meV, i.e., much more. Evidently, the asymmetry of the behavior of the upper and lower modes can only be obtained in the case of the asymmetric frequency distribution of excitons.

The origin of the narrowing of the modes obtained within our model can be formulated as follows. If the vacuum-field Rabi splitting exceeds the original broadening of the exciton line, at the anticrossing condition the tails of the excitonic distribution which couple weakly with the light remain in between two split modes and do not effect the reflection. In the same terms one can explain the asymmetry between the upper and lower branches. In our case the lower branch is a subject of mixing between the photon mode and the lower part of the excitonic distribution which is sharper than the upper part. That is why the lower branch has a narrower linewidth at the anticrossing condition.

Important information about the inhomogeneous distribution of excitons can be obtained from the time-resolved spectra of microcavities. Oscillating time-resolved reflection spectra have been reported for microcavities with embedded QW's.[20,6] Theoretically, time-resolved reflection of the microcavity in the case of incident δ pulse is described by a function

$$G(\tau) = \int \frac{d\omega}{2\pi} \exp(-i\omega\tau) r_{\mathrm{cav}}(\omega), \tag{13}$$

where r_{cav} is the amplitude reflection coefficient from the structure. The experimentally detectable quantity is the integrated intensity of light which is proportional to $|G(\tau)|^2$. Previous works[18,13] showed that the period of Rabi oscillations (the beats between two cavity-polariton branches) strongly depends on the disorder. An effect of asymmetry of the frequency distribution of excitons is illustrated by Fig. 3, which shows the calculated time-resolved reflection spectra from the same structure as before at the anticrossing condition. Curve A was calculated with $\Delta_1 = 2$ meV, $\Delta_2 = 1$ meV, and curve B was calculated with $\Delta_1 = \Delta_2 = 1.5$ meV. Surprisingly, the period of the beats is shorter in the case of an asymmetric excitonic distribution than in the case of a symmetric excitonic distribution. Definitely we are still in the regime where the period of oscillations increases with the increase of inhomogeneous broadening. Thus, in the case of the asymmetric distribution the inhomogeneous broadening appears to be stronger even though the sum of Δ_1 and Δ_2 is the same for both curves.

In conclusion, the main features of the microcavity reflection spectra earlier attributed to the motional narrowing effect were described within a simple semiclassical model. The narrowing of both cavity polariton modes at the anticrossing and the asymmetry of the linewidth behavior at the anticrossing for upper and lower modes are clearly seen from the present calculation. The model assumed wave-vector conservation in the cavity plane, thus strictly speaking, it did not take into account the motional narrowing of exciton polaritons. On the other hand, the distribution of the exciton resonance frequency was assumed to be asymmetric, which implies the same kind of arguments as the microscopic theory of motional narrowing. Lower lying exciton states are subject to the quantum confinement effect in the disorder potential which increases their energy making the lower-frequency wing of the distribution of excitons sharper than the upper wing. This asymmetric distribution of excitons, however, has been supposed to be independent of the detuning between exciton and cavity modes which excludes effects connected with an effective change of the exciton mass due to its coupling with cavity photons. These kind of effects probably play an important role in Rayleigh scattering experiments, but not in the reflection.

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