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Microcavity polariton linewidths in the weak-disorder regime

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Polariton linewidths have been measured in a series of high-quality microcavities with different excitonic inhomogeneous broadening in the weak-disorder regime. We show experimentally that the influence of the disorder on the polariton linewidths is canceled when the polariton energies are far in the tail of the excitonic absorption. The measured linewidths are quantitatively compared with an estimation using the measured excitonic absorption spectrum of the bare quantum wells, and good agreement is found.

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The role of static disorder in the linewidth of cavity polaritons has been intensively discussed in recent years, experimentally and theoretically. In 1996, Whittaker *et al.*¹ reported reflectivity spectra measured on a GaAs semiconductor microcavity (MC) containing three InGaAs quantum wells (QW's). They observed that when the cavity mode is in resonance with the bare heavy-hole (HH) exciton transition the linewidth of the lower polariton (LP) is smaller than expected by averaging the bare exciton and cavity linewidths. Moreover, the lower-polariton linewidth is narrower than that of the upper polariton. They explained the subaverage broadening with a scaling theory based on a motional narrowing argument: When a quantum particle of finite size moves in a disordered potential with correlation length smaller in size, the spectral linewidth is reduced compared to that of a classical particle due to spatial averaging over the disorder. However, their model, which neglected interbranch scattering and nonparabolicity of the polariton dispersion, failed in explaining the different linewidths of the lower and upper branches near resonance. In 1997, Savona *et al.*² proposed a one-dimensional microscopic model of disordered quantum wells embedded in a microcavity where exciton-photon coupling and exciton-disorder interaction are treated nonperturbatively. This model was able to reproduce both the subaverage broadening due to motional narrowing, and the larger linewidth in the upper polariton due to additional interbranch scattering. In 1998, Ell *et al.*³ showed that all these features were reproduced by using the measured optical absorption of the bare QW together with linear dispersion theory. They concluded that the disorder-averaged excitonic response in the bare QW fully determines the optical properties of the well embedded in a microcavity, and in particular that the asymmetric line shape of an inhomogeneously broadened exciton absorption⁴ is responsible for the narrower LP linewidth compared to that of the upper polariton. At the same time Whittaker⁵ proposed a unified picture where numerical calculations using a microscopic two-dimensional model were in good agreement with a simpler model describing the polariton to exciton scattering in terms of an absorption picture. In this model, the polariton linewidth is given by the photon lifetime due to escape through the mirrors and by the loss of photons due to absorption into

exciton states. In other words, the photonic fraction of the upper and lower polaritons is resonant with states in the tails of the exciton absorption that are in the weak-coupling regime and simply act as a source of absorption in the cavity. Therefore, in this model the difference between the widths of the two polariton branches is a consequence of the asymmetry of the inhomogeneously broadened exciton line shape, with larger absorption on the high-energy side.⁴ This absorption model is thus a simplified version of the model of Ell *et al.*,³ but in essence also agrees with the suggestion of Savona *et al.*² that the large width of the upper branch is caused by scattering of polaritons into higher-momentum exciton states. It is emphasized by Whittaker⁵ that this simple model is justified because motional narrowing effectively eliminates the contribution to the linewidth due to disorder-induced scattering between low-momentum polaritons in the same branch, and the only important contribution is the polariton to exciton scattering. Thus a very intriguing feature of this absorption model is the prediction that the disorder contribution to the polariton linewidth should *disappear* in structures with small but finite exciton inhomogeneous broadening, when polariton to exciton scattering becomes impossible because the polariton energies are too far in the tails of the excitonic absorption (see Fig. 3 in Ref. 5). More precisely, the disorder contribution to the polariton linewidth should disappear, especially in the lower polariton, when the exciton inhomogeneous broadening is much narrower than the Rabi splitting in the microcavity (weak-disorder limit). However, this point was not experimentally verified in these previous works, also due to the significant exciton inhomogeneous broadening in the samples investigated. Recent achievements in growth of high-quality microcavities^{6,7} now open the possibility of experimentally verifying this prediction.

In this article we report a detailed investigation of the polariton linewidths in a series of semiconductor microcavities with variable exciton inhomogeneous broadening, in the weak-disorder regime. We have used molecular beam epitaxy grown GaAs single QW's embedded in AlGaAs microcavities, with three different well widths of 25 nm, 15 nm, and 10 nm. With decreasing well width, the exciton becomes more sensitive to the interface and alloy disorder due to the larger wave function overlap with the interface region. The

FIG. 1. Absorption profile, as deduced from the photoluminescence spectrum at 20 K corrected by a Boltzmann population factor, for a 25 nm (bottom), a 15 nm (middle), and a 10 nm (top) GaAs/Al_{0.3}Ga_{0.7}As single quantum well. In the insets, four-wave mixing (FWM) traces at 5 K are given with the corresponding fits of the homogeneous broadening.

optical properties of bare GaAs/Al_{0.3}Ga_{0.7}As QW's of the same nominal thicknesses and grown under similar conditions are shown in Fig. 1. The excitonic absorption spectrum is deduced from the photoluminescence spectrum, measured at 20 K at resonant excitation and low intensity, divided by the Boltzmann population factor, assuming an exciton population in thermal equilibrium at the lattice temperature as described in Ref. 8. This procedure is already a good approximation at 20 K in the samples investigated as we have experimentally verified by comparison with higher temperatures. The corresponding half widths at half maximum (HWHM's) of the HH excitonic absorption are indicated. All the linewidths will be given in the following as HWHM's. In order to estimate the exciton inhomogeneous broadening we have compared these linewidths with measurements of the homogeneous broadening by degenerate four-wave mixing (FWM). Details of the experimental setup can be found in Ref. 9. In the inset, the time-integrated FWM for collinearly polarized pulses is shown versus their relative delay time at 5 K close to the low-density regime ($\sim 10^9$ excitons/cm²). For the 25 nm wide QW (bottom plot) the decay is monoexponential and the FWM is a free-polarization decay, indicating the absence of disorder probed by the exciton.⁹ The corresponding homogeneous broadening agrees well with the absorption linewidth at 20 K, when we take into account

2.1 $\mu\text{eV/K}$ linewidth broadening by acoustic-phonon absorption.⁹ Thus, the exciton absorption in the 25 nm QW is predominantly homogeneously broadened. In the 15 nm wide QW (middle plot) signatures of exciton inhomogeneous broadening appear in the absorption line shape, which is broader and slightly asymmetric.⁴ The FWM is now a photon echo with a delayed maximum, and its decay was fitted as proposed by Erland *et al.*¹⁰ An acoustic-phonon coefficient of $\sim 2 \mu\text{eV/K}$ was also measured in this QW, resulting in a homogeneous broadening contribution of $\sim 80 \mu\text{eV}$ at 20 K. Thus the total linewidth of the 15 nm wide QW is an interplay of comparable homogeneous and inhomogeneous broadenings. Finally, the absorption profile of the 10 nm QW (top plot) is strongly inhomogeneously broadened, with a negligible contribution due to the homogeneous broadening as deduced by FWM, and largely asymmetric.

We have recently reported on the optical properties of a microcavity formed by embedding a 25 nm GaAs single QW inside an AlGaAs λ cavity.^{6,11} An AlAs/Al_{0.15}Ga_{0.85}As Bragg reflector of 25 (16) periods was grown at the bottom (top) of the cavity. A special design of the Al_{0.3}Ga_{0.7}As barriers and the spacer layer was shown to be successful in reducing excess free-carrier densities in the microcavity compared to more conventional designs.¹¹ We obtained a Rabi splitting to linewidth ratio of 19 at 11 K, one of the best reported until now to our knowledge for single QW microcavities. With the same design, we have grown two new microcavities containing, respectively, a 15 nm and a 10 nm wide single GaAs QW, which are investigated in this work in comparison with the MC containing the 25 nm QW. The energy of the polaritons versus the energy distance between the cavity resonance and the bare HH exciton (detuning) is shown in Fig. 2 for these two microcavities at low temperature (5 K). A good fit to the data is obtained by solving a three-coupled-oscillator model including the light-hole (LH) exciton.¹¹ The coupling to the LH exciton results in the appearance of three polariton modes, the lower, middle, and upper polaritons. From the fits we deduced 3.8 meV HH Rabi splitting (2.4 meV LH Rabi splitting) for the MC containing the 15 nm QW, and 4.15 meV HH Rabi splitting (2.8 meV LH Rabi splitting) for the MC containing the 10 nm QW. Thus, in all the microcavities investigated the HH exciton inhomogeneous broadening is smaller than one-half the HH Rabi splitting, i.e., the structures are in the weak-disorder regime.⁵ Note that for smaller well widths the stronger quantization of the LH exciton leads to a smaller mixing of this state in the LP and middle polariton (MP) resonances around zero detuning.

In the inset, the reflectivity spectrum measured with white light near normal incidence ($\sim 1^\circ$) is shown for both MC's at zero detuning. The white-light source was focused on the sample with a spot diameter of 70 μm resulting in a small energy broadening of the cavity mode from the wedge gradient and the incident wave vector spread.¹¹ The spectra were detected using a spectrometer and an intensified multi-channel analyzer, with a total spectral resolution close to a Lorentzian line shape of 65 μeV HWHM. It can be seen from the insets of Fig. 2 that the LP linewidth is narrower

FIG. 2. Polariton energies versus detuning for the 10 nm (upper) and the 15 nm (lower) GaAs quantum well microcavity at 5 K. The solid lines are fits to the data. The insets show the reflectivity spectrum of the microcavity at zero detuning.

than that of the MP and its value is subaverage, in agreement with previous observations on the effect of disorder on the polariton linewidths.¹

In order to investigate in detail the role of the disorder in the polariton linewidths, we have measured the detuning dependence of the LP and MP linewidths, as shown in Fig. 3. The measured linewidths, well fitted with Lorentzian profiles, have been corrected by subtracting the spectral resolution. The closed squares are the LP linewidths while the open circles are the MP linewidths. The solid (dotted) lines are the cavity linewidths, due to the finite transmission of the Bragg mirrors, multiplied by the calculated photonic content in the lower (middle) polariton. The cavity linewidth was calculated taking into account the change of the linewidth with energy position in our specially designed structure with uniform Bragg mirrors and a wedged spacer layer,¹¹ and agrees well with the measured LP linewidth at large negative detunings. Uncertainties in the fit of the cavity linewidths are less than 10%. Comparable LP broadenings are measured at large negative detuning in all the samples, consistent with the fact that all the samples have nominally equal Bragg mirror reflectivities. It can be clearly seen from Fig. 3 that close to zero detuning the 15 and 10 nm QW MC's show a LP linewidth narrower than that of the MP. Moreover, from the comparison with the calculated photonic part the LP linewidth is subaverage and given *only by the cavity contribution*. In the 25 nm QW MC, instead, the LP and MP linewidths are almost equal at zero detuning, and the LP linewidth is larger than the cavity linewidth contribution.

In order to give a physical picture of the observed results,

FIG. 3. Linewidths versus detuning of the lower (closed squares) and middle (open circles) polaritons, for the different microcavity samples as indicated. The solid (dotted) lines are the calculated photonic linewidths of the lower (middle) polaritons.

we have plotted in Fig. 4 the polariton linewidths versus the distance $E_P - E_{HH}$ of the polariton energy from that of the HH bare exciton. According to the simple absorption model of Whittaker,⁵ the polariton linewidth is the sum of the cavity contribution and an excitonic part. This excitonic part γ_e of the linewidth should be related to the strength of the exciton absorption probed by the polariton resonances. We have quantitatively performed this comparison. The measured absorption spectra in Fig. 1 have been scaled in order to represent the absorption probability in the QW.¹² The area of the absorption probability $\alpha(\omega)$ in a QW is proportional to the oscillator strength per unit area¹³ f :

$$\int \alpha(\omega) d(\hbar\omega) = \frac{2\pi^2 \hbar e^2}{nm_0 c} f \quad (1)$$

with the refractive index n , the electron charge e and mass m_0 , and the speed of light c . The oscillator strength per unit area is proportional to the exciton radiative linewidth,¹³ which is related to the Rabi splitting $\hbar\Omega$ in the microcavity,¹⁴ and one gets

$$f = \frac{(\hbar\Omega)^2 n^2 m_0 L_{\text{eff}}}{8\pi e^2 \hbar^2} \quad (2)$$

with L_{eff} the effective length in the microcavity due to the penetration length into the Bragg mirrors. The absorption probability is a loss in the cavity that gives rise to a photon

FIG. 4. Measured (symbols) and calculated (lines) polariton linewidths versus the energy distance of the polariton from the bare HH exciton. The dotted lines are the estimated cavity contributions in the polariton linewidths.

decay rate $\alpha(\omega)c(nL_{\text{eff}})^{-1}$ additional to the transmission losses from the Bragg mirrors. Thus, finally, the measured absorption spectrum with an integral over the energy normalized to 1, $s(\omega)$, is related to γ_e by

$$\gamma_e = |c_l|^2 s(\omega) \frac{\pi(\hbar\Omega)^2}{8} \quad (3)$$

with $|c_l|^2$ being the photonic content of the polariton at the energy $\hbar\omega$. This formula is a valid approximation when $|c_l|^2$ is not too small; otherwise it predicts that the polariton linewidth tends to zero when the photonic content tends to zero, while the true value tends to the exciton linewidth.⁵

The solid lines in Fig. 4 are the calculated polariton linewidths $\gamma_p = |c_l|^2 \gamma_c + \gamma_e$ with γ_c the cavity linewidth (see Fig. 3) and γ_e calculated using Eq. (3) and the absorption spectra in Fig. 1. For comparison, the isolated cavity linewidth contribution $|c_l|^2 \gamma_c$ is plotted as dotted lines. The overall quantitative agreement between calculated and measured linewidths is quite good for $|E_p - E_{\text{HH}}| > 1$ meV,

where the absorption model holds, i.e., when the photonic content is not too small and when the polariton energies are resonant with excitonic states far enough in the tail of the absorption to be in the weak-coupling regime.⁵ In particular, it appears clearly that when the excitonic absorption profile is asymmetric the contribution to the polariton linewidth from the excitonic absorption is bigger for $E_p - E_{\text{HH}} > 0$, i.e., for the MP, than for $E_p - E_{\text{HH}} < 0$, i.e., for the LP, in agreement with the measurements. Moreover, in the LP the disorder-induced broadening is *absent* when the polariton energies are far in the tail of the excitonic absorption.

Some small discrepancies between calculation and measurements are also evidenced. A parameter not under control in this comparison is the density of excess free carriers in the samples, resulting in trion resonances in the low-energy tail of the excitonic absorption. In the upper part of Fig. 4 a trion resonance ~ 2 meV below the exciton seems to be present in the 10 nm bare QW, which is not observed in the micro-cavity sample. The opposite occurs in the 25 nm QW (lower figure), where a trion resonance is evidenced in the micro-cavity sample, approximately 1 meV below the HH exciton, also in agreement with photoluminescence measurements on this structure,¹¹ while it is less present in the bare QW sample. Additionally, in the 15 nm and 25 nm QW MC's, the MP linewidths appear to be slightly higher (by a few tens of μeV) than the calculated ones in the region where the photonic content is not very small. Even if this difference is at the limit of our accuracy, it is reasonable that when the disorder-induced broadening is very small the MP linewidths are affected by other scattering processes, such as phonon or free-carrier interactions, not included in the simple absorption model of Whittaker.⁵ In fact, the observed difference is on the order of what is calculated from additional homogeneous broadening of the upper polariton due to acoustic phonon scattering,¹⁵ which is inhibited in the lower polariton. As a general comment, note that the polariton linewidths shown in this work are very small, well below those reported in previous work investigating the role of disorder in cavity polaritons.^{3,5} Thus it is not surprising that small features such as trion/free carriers and phonon effects are affecting the measured linewidths in our samples.

In conclusion, we have shown experimentally that the influence of disorder on the polariton linewidth of microcavities in the weak-disorder regime can be canceled when the polariton energies are far in the tail of the excitonic absorption, in agreement with predictions in the literature.^{5,2} The measured linewidths have been *quantitatively* compared with those estimated from the measured excitonic absorption in the bare quantum wells, following the simple absorption model proposed by Whittaker.⁵ A generally good agreement is found.

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