Influence of Structural Disorder and Light Coupling on the Excitonic Response of Semiconductor Microcavities

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The interplay between structural disorder, Coulomb interaction, and light-coupling effects is investigated for high-quality quantum wells in a semiconductor microcavity. The independently measured experimental susceptibility of a single quantum well in linear dispersion theory yields excellent agreement with a series of measured reflectivity spectra for a variety of microcavity and distributed Bragg structures, showing that the disorder-averaged response of the exciton in its quantum well determines the optical response. [S0031-9007(98)06153-5]

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Static structural disorder on mesoscopic length scales is an unavoidable feature of semiconductor microstructures. Besides the impurities, lattice imperfections, etc., which are also present in volume (bulk) crystals, quantum-well systems additionally exhibit interface imperfections due to the formation of steps during the layer-by-layer growth or fluctuations of the composition of neighboring atomic layers. Because of its strongly quantum confinement dependent energetic properties, the free exciton optical response is a very sensitive measure of structural disorder properties. A very interesting question in this context arises regarding the interplay between structural disorder, the attractive Coulomb interaction of the electron-hole pairs, and the radiative coupling effects on the optical response of real structures. The microscopic modeling of all these effects in a fully self-consistent theory is one of the major challenges of current approaches which will keep theorists busy for some time to come. However, important insights into this complex problem can be gained already by well-controlled experimental studies where aspects of the disorder problem are isolated.

Recent studies of Whittaker *et al.* [1] and Savona *et al.* [2] showed that the excitonic response of semiconductor microcavities depends very sensitively on structural disorder [3,4]. Because of the strong nonperturbative light-matter interaction, the microcavity reflection spectrum exhibits pronounced normal-mode coupling (NMC), as first observed by Weisbuch *et al.* [5]. Measurements of the energy dependence of the two resulting reflectivity dips show characteristic modifications of this so-called cavity polariton response. Related light-coupling effects dominate the response of quantum-well Bragg and anti-Bragg structures [6–8]. A very fascinating question in this context is whether light-coupling effects are indeed able to modify the influence of structural disorder on the excitonic quasiparticles within their quantum wells.

In this Letter we present an experimental test using extremely high-quality samples to study to interplay between light-coupling and disorder effects. Even though the concept is rather straightforward, the detailed analysis requires substantial accuracy and very good sample characteristics. For this purpose we grew a series of quantum-well structures with and without Bragg mirrors. We then *measured* the optical response of all these structures such that we knew the energy dependent reflectivity of our quantum well (QW) in a high-quality microcavity. Clearly, since this is an experiment, all microscopic effects are included consistently. We then measured the optical response of all these structures that we knew the energy dependent reflectivity of each microcavity containing one or more QW's. Next, we measured the optical transmission of our reference QW's and used these results to extract the *disorder averaged* excitonic response. Using this effective *measured* susceptibility in linear dispersion theory (LDT) [5, 9–15], we computed the microcavity spectrum and compared it with the experimental results of the complete microcavity system. If the LDT results are able to reproduce the experimental spectra of the microcavity system, this demonstrates that polaritonic effects in the exciton-disorder light coupling are negligible in contrast to the claims in Refs. [1] and [2].

LDT constructs the reflectivity and transmission of light incident on a multilayer structure by working from the boundary conditions at each interface required by Maxwell's equations [16]. Such an approach requires a knowledge of the refractive index n and the absorption coefficient α or the corresponding optical susceptibility of each layer. The QW absorption coefficient α_{QW} used in our LDT is shown in Fig. 1(a). The *e-hh* exciton has a FWHM linewidth of 0.56 meV, and it was measured with ≈ 0.9 Å bandpass (0.17 meV) on a sample called DBR13, which consists of 30 QWs. The GAAs barriers have a thickness such that the $\lambda/2$ Bragg condition occurs somewhere on the sample. The absorption coefficient α_{QW} was calculated from experimental transmission data $\alpha_{\mathrm{QW}} = \frac{1}{30} L \ln(I_0/I_t)$, where $L = 85$ Å is the thickness

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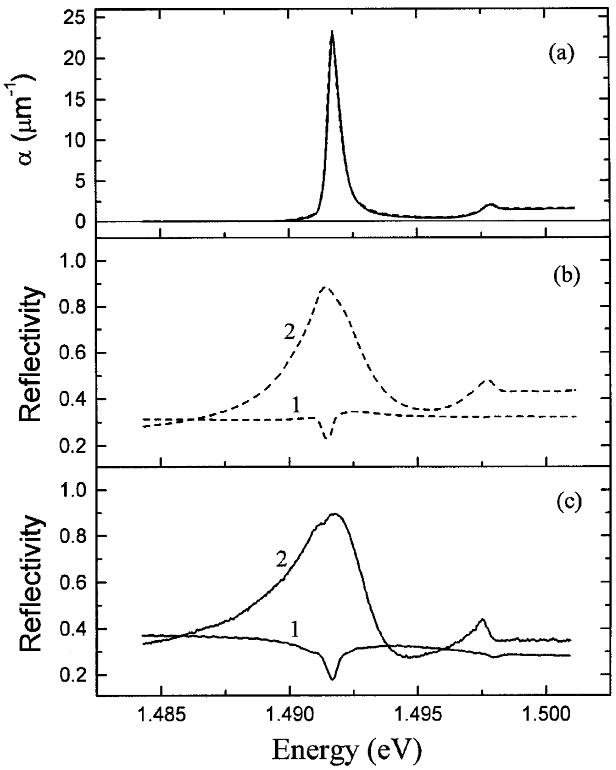

FIG. 1. (a) Experimental (solid line) and calculated (dashed line) absorption coefficient as a function of energy. The lines are hard to distinguish. (b) Calculated and (c) measured reflectivity at Bragg resonance (2) and at $0.85\lambda/2$ spacing (1) as a function of energy.

of the QW, I_0 is the measured incident light intensity, and I_t is the measured transmitted light intensity. To avoid pronounced constructive radiative coupling effects, the measurement was performed for QW spacing of $0.85\lambda/2$, i.e., far from the Bragg condition.

In order to test the approach to use the experimental susceptibility for the optical response of a heterostructure, we use the measured data to compute the corresponding reflection in the Bragg resonance, where the light coupling leads to a pronounced resonance broadening [Fig. 1(b)]. Figure 1(c) shows the corresponding measured reflectivity. Note that the linewidth of the $R(E)$ is dramatically broader at the Bragg $(\lambda/2)$ spacing which is the linear cw spectroscopy equivalent of the increased radiative damping rate reported in [7] and related to the splitting seen in [17]. The comparison indicates that the measured reflectivity spectra are well reproduced, putting the absorption measurement in a transfer matrix calculation.

In the next step we now use the extracted susceptibility to compute the response of our microcavity structures. The solid curves in Figs. 2(a) and 2(b) show LDT calculations of the linewidths of the two branches for two of our own samples, referred to as NMC63 and NMC65, which were grown by molecular beam epitaxy. NMC63 consists of a $3\lambda/2$ GaAs spacer with two InGaAs QWs, one at each antinode, and NMC65 contains a λ GaAs spacer with one InGaAs QW at the antinode. The QWs are identical to those of DBR13. NMC63 has 28/33 $(R = 99.6)$ quarter-wave layers of GaAs and AlAs in the top/bottom mirror, while NMC65 has 16/21 $(R = 95.6)$.

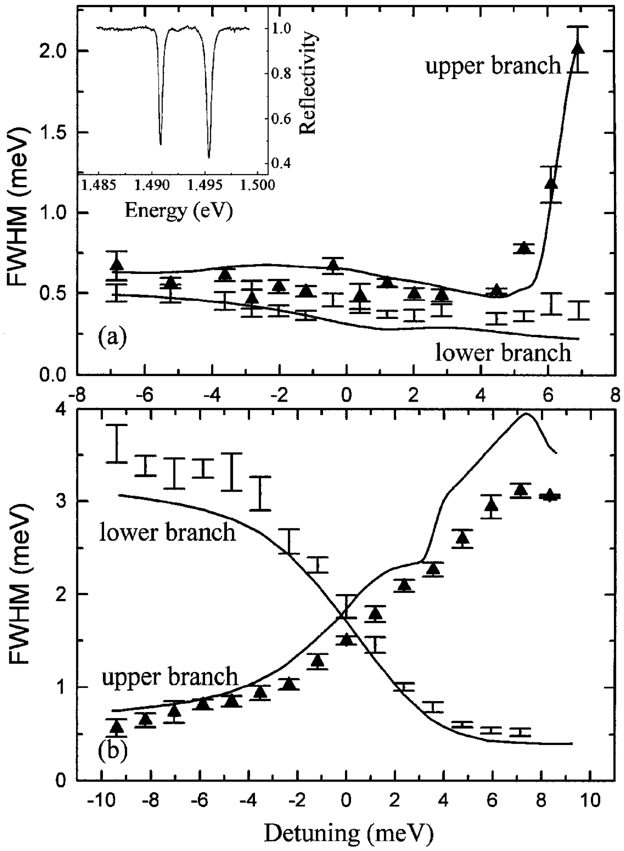

FIG. 2. Comparison of measured (symbols) and calculated NMC linewidths (solid line) for samples NMC63 (a) and NMC65 (b) as a function of the detuning. The inset in (a) shows the reflexivity spectra as a function of energy for approximately equal dips.

Experimental measurements of linewidths were done with sample cooled to 10 K. Different detunings were achieved by scanning the cavity line through resonance with the almost fixed exciton absorption ine by moving about 2 mm over the surface of the samples, away from the sample growth center. The spectra were measured in reflectivity in the linear regime by probing a 30 μm diameter on the sample, with a beam half-cone angle of 4.4°, using a spatially filtered, Gaussian, broadband light-emitting diode (LED) probe centered at ≈ 850 nm $(\approx 1.46$ eV). A small probe spot size and incident angle is needed in order to minimize averaging over different detunings in a single measurement. The reflectivity was measured using a 1.25 meter scanning Spex spectrometer with a 0.75 Å $(\approx 0.13$ meV) bandpass, and a Hamamatsu R943-02 photomultiplier tube with a lock-in amplifier detection technique. Note in the LDT calculation of these lines, each reflectivity spectrum was calculated by averaging over the set of thicknesses encompassed by a 30 μm spot in order to mimic experimental conditions. A small amount (0.00038 $\mu \mathrm{m}^{-1}$ for NMC63 and 0.001 $\mu \mathrm{m}^{-1}$ for NMC65) of constant background absorption was added to all mirror layers and the spacer, resulting in bare-cavity FWHM linewidths of 0.5 and 3.7 meV, in agreement with measurements. These do not qualitatively change the results but do improve the agreement of measurement and theory. The broadening δ_{spect} due to the spectrometer bandpass has been taken into account by $\delta_{\mathrm{plotted}} = (\delta^2_{\mathrm{measured}} - \delta^2_{\mathrm{spect}})^{1/2}$. Experimental detuning was calculated by $\hbar\omega_{\mathrm{ex}} - \hbar\omega_c$, where $\hbar\omega_c$ was determined by multiplying the wavelength rate the cavitylike mode tunes with position when far from the resonance condition.

The comparisons in Figs. 2(a) and 2(b) show excellent agreement of the LDT calculations with the full experimental results. Similar agreements have been found with a third sample studied, NMC64, which has the same spacer as NMC63, but 38/43 top/bottom mirror layers. Note that NMC63, 64, 65, as well as DBR13 were grown under the same growth conditions and in rapid succession.

To further demonstrate the validity of our approach we used the same procedure to extract the actual reflectivity of the QW-embedded microcavity structure of Whittaker *et al.* [1]. It is shown in Fig. 3(a). In applying LDT to the data of [1], it was necessary to extract α_{QW} [18], shown in Fig. 3(b). The resultant extracted α_{QW}, when put into LDT for their sample with the top mirror etched away as well as 5.3% of the spacer, approximately reproduces their measured reflectivity dip, as shown in Fig. 3(c). The FWHM of the exciton absorption line is 2.65 meV, while the reflectivity dip has a FWHM of 3.1 meV. Cavity losses are taken into account by introducing an amount of mirror adsorption which results in a FWHM of the empty cavity line of 1.2 meV. Note that our LDT calculations agree with the data even better than the computations of Savona *et al.* [2], which involve in a one-dimensional microscopic model the scattering of excitons on a QW interface disorder potential leading to a motional narrowing effect.

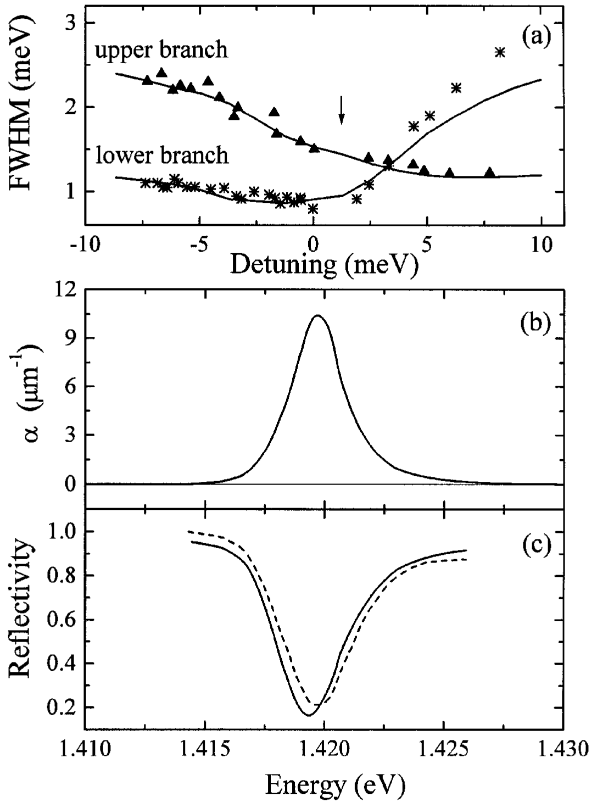

FIG. 3. (a) Comparison of measured and calculated NMC linewidths (stars: lower branch; triangles: upper branch) for the data of Ref. [1]. The arrow marks the detuning for equal reflectivity dips. (b) Reconstructed absorption coefficient and (c) comparisons of the measured (dashed line) reflectivity [1] and the calculated one (solid line) for removed top mirrors.

In studies of NMC linewidths, surprise has been expressed over two observations. The first surprise is that the upper branch linewidth δ_u is broader than the lower branch linewidth δ_ℓ. The second surprise is that on resonance δ_ℓ is sometimes less than the mean of the bare QW exciton linewidth δ_{ex} and the empty cavity linewidth δ_c, which would be the result expected for a homogeneously, symmetrically broadened oscillator. Linear dispersion theory provides an explanation of both of these puzzles by consideration of the bare QW exciton line shape.

The linewidth of the two branches was found to be very sensitive to small local changes in the excitonic absorption coefficient and refractive index in the immediate vicinity of the NMC peaks, denoted by $\hbar\omega_\ell$ and $\hbar\omega_u$. The absorption line, as can be seen in Figs. 1(a) and 3(b), is affected by disorder, resulting in an asymmetric line shape with higher absorption on the high energy side. Consequently, equality of the reflection dips does not occur at the resonance condition $\hbar\omega_c = \hbar\omega_{\mathrm{ex}}$; instead $\hbar\omega_c > \hbar\omega_{\mathrm{ex}}$ is required, so that the exciton tail absorptions are equal at $\hbar\omega_\ell$ and $\hbar\omega_u$. The linewidths δ_ℓ and δ_u are determined mostly by the locally different slopes of the QW refractive index, resulting in $\delta_u > \delta_\ell$ explaining the first surprise. In contrast, at resonance, the smaller low energy tail absorption is the main cause of the smaller linewidth of the lower branch.

The cases treated here represent three different ratios of exciton/cavity linewidths: (i) $\delta_{\mathrm{ex}} \gg \delta_c$ [realized in Fig. 3(a)], (ii) $\delta_{\mathrm{ex}} \ll \delta_c$ [shown in Fig. 2(b)], and (iii) $\delta_{\mathrm{ex}} \approx \delta_c$ [the intermediate case which can be seen in Fig. 2(a)]. All three cases can be understood and explained by the LDT approach.

(i) The case of $\delta_{\mathrm{ex}} \gg \delta_c$, where δ_{ex} is strongly inhomogeneously broadened, is the most pronounced example of the second surprise. While we emphasize the importance of line shape asymmetries and spectrally local values of the susceptibility, Houdré *et al.* [19] has shown, for symmetrically inhomogeneously broadened lines and an NMC splitting much larger than δ_{ex}, that linewidths become $\delta_\ell \approx \delta_u \approx (\delta_{\rm ex}^{\rm hom} + \delta_c)/2$. This is the basic physics of the second surprise.

The article of Whittaker *et al.* [1] is unable to explain δ_u, but argues that motional narrowing applied to the microcavity polaritons results in significant motional narrowing which is unimportant for the QW alone. They claim that this is because the polariton effective mass is much lighter ($\approx 10^{-5} m_0$ compared with $0.1 m_0$) resulting in averaging over a much larger diameter (10^4 Å instead of an exciton Bohr radius). Although the effect of the microcavity is to bring out particular aspects of the QW, namely, the absorption and refractive index in the immediate vicinity of the two peaks, the agreement obtained by our approach here shows that there are not properties of the composite system unexpected from the properties of the QW and the empty microcavity put together self-consistently in LDT.

(ii) The case $\delta_{\rm ex} \ll \delta_c$ is realized by our NMC65 sample which has a lower mirror reflectivity. The linewidths versus detuning are very symmetric because the symmetric cavity oscillator linewidth is largest so that the exciton asymmetries are unimportant in the crossover at zero detuning.

(iii) The intermediate case is shown by our NMC63 sample, where cavity and excitonic properties contribute almost the same. As can be seen, the linewidths of the upper and lower branch are almost constant over a wide range of detuning because the cavity and exciton linewidths are almost equal; thus the linewidth is almost independent of their relative contributions. The large increase of the FWHM of the upper branch at $\hbar\omega_c > \hbar\omega_{\rm ex}$ emphasizes the importance of spectrally local absorption; at that detuning range the cavity resonance passes the hh-e ($2s$ and continuum) and ℓh-e $1s$ exciton resonances.

In conclusion, our analysis shows clearly that for currently available top-quality samples it is an excellent approximation to assume that the disorder-averaged excitonic response determines the optical properties. Based on this insight, it is shown here that the asymmetry in the bare QW absorption line shape when used in LDT explains the two most striking features of NMC linewidths: The linewidth of the lower NMC peak is narrower than the upper, and often narrower than the mean of the cavity and exciton linewidths. For InGaAs/GaAs QWs as presently grown, the success of LDT seems complete in that there is good agreement for both low and high In concentrations. In general there is no question that the correct approach is to treat disorder, Coulomb interaction effects, and light propagation on an equal footing, i.e., simultaneously. However, this is a very difficult problem whose solution is not required to analyze the properties of currently available structures. It remains for future studies to unravel the microscopic nature of the disorder and to see if samples can be grown where LDT fails.

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