Microscopic Theory of Motional Narrowing of Microcavity Polaritons in a Disordered Potential

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The influence of static disorder on the optical properties of microcavity-embedded quantum wells is studied by means of a microscopic model. Both the exciton-photon coupling and the exciton-disorder interaction are treated nonperturbatively. Despite the nonconservation of the in-plane momentum due to disorder, the Rabi splitting is still present. Moreover, the lower polariton is subject to motional narrowing of the spectral response, while for the upper one this effect is less important. This result shows very good agreement with recently observed spectra in high-quality samples. [S0031-9007(97)03351-6]

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The optical response of Wannier excitons in narrow quantum wells (QWs) usually shows a significant broadening of the exciton line due to the presence of impurities and, mainly, to the imperfections of the QW interfaces. In addition to inhomogeneous broadening, other features such as asymmetry of the exciton line and Stokes shift in the photoluminescence originate as a consequence of the disorder present in the system. Existing models which describe these properties are based on the solution of the Schrödinger equation for the exciton center-of-mass motion in the presence of a disordered potential [1–5]. The optical response is then calculated as usual by means of the Fermi golden rule.

When a QW is embedded in a planar semiconductor microcavity (MC), the influence of in-plane disorder on the optical properties becomes crucial. In fact, the conservation of the exciton in-plane momentum is essential for the existence of the MC polariton modes and, in particular, of the normal mode splitting (Rabi splitting) in the strong coupling regime. However, it was shown [6,7] that a simplified model in which the effect of disorder is included only through an inhomogeneous exciton level distribution, without lifting the in-plane momentum conservation, still describes the main features of the optical response. The question thus arises whether lifting the in-plane momentum conservation produces relevant changes in the optical spectra and, in particular, in the Rabi splitting.

In this Letter, we propose a microscopic model of disordered QW embedded in a MC. The coupled equations of the exciton and cavity photon systems are solved numerically and the optical spectra for normal incidence are calculated. The treatment is nonperturbative and includes multiple scattering to all orders, both for the exciton-photon interaction [8], and for the exciton-disorder interaction which involves different **k** vectors. In this context, we stress the importance of including the full polariton dispersion and the interbranch scattering. The calculations first show that the Rabi splitting still exists for sizable values of the disorder parameter, indicating that the **k**-conserving exciton-photon interaction prevails over the **k**-nonconserving exciton-disorder interaction. Moreover, the obtained polariton inhomogeneous broadening is smaller than what expected from the **k**-conserving models [6,7]. The shape of the optical spectra is strongly asymmetric at resonance, showing an upper polariton peak broader and less pronounced than the lower one. We explain these results in terms of *motional narrowing* of the polariton modes. The term motional narrowing indicates the averaging of the fluctuations when a quantum particle with a finite size moves along a disordered potential with a correlation length of comparable size [9]. Recently, Whittaker *et al.* [10] have reported reflectivity measurements on a high quality MC sample displaying both these features. They propose a scaling theory which explains the subaverage broadening in terms of motional narrowing. Their theory, however, while relying on the image of microcavity polaritons, disregards the interbranch scattering and the strong nonparabolicity of the polariton dispersion. The model fails to predict the asymmetry of the polariton spectrum at resonance. Our results, on the contrary, explain very satisfactorily the observed polariton broadenings in terms of motional narrowing, thus showing the importance of a nonperturbative treatment. We also provide a qualitative argument which shows that only the upper polariton is affected by the interbranch scattering and its linewidth is therefore larger than the one of the lower polariton.

Let us consider a planar Fabry-Pérot resonator with perfectly reflecting mirrors and thickness L_c. The coupling to the outside radiation will be introduced later. We

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neglect higher order modes of the Fabry-Pérot, assuming that their energy separation from the fundamental mode is much larger than the exciton-radiation coupling. Under this assumption, an exciton state with a given in-plane wave vector $\mathbf{k}$ is coupled to the fundamental Fabry-Pérot mode with the same $\mathbf{k}$. Consequently, the polariton Hamiltonian, which describes the coupled exciton-photon system, writes

$$H_{\text{pol}} = \sum_{\mathbf{k}} [\hbar v \sqrt{k^2 + k_z^2}\, a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \hbar \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} + C_{\mathbf{k}} (a_{\mathbf{k}}^\dagger b_{\mathbf{k}} + a_{\mathbf{k}} b_{\mathbf{k}}^\dagger)] + \sum_{\mathbf{k},\mathbf{k}'} V(\mathbf{k} - \mathbf{k}') b_{\mathbf{k}'}^\dagger b_{\mathbf{k}}, \quad (1)$$

where $k_z = 2\pi/L_c$, $a_{\mathbf{k}}$ and $b_{\mathbf{k}}$ are the cavity photon and exciton Bose operators, respectively, and $C_{\mathbf{k}}$ is the polariton coupling constant [8]. The last term in (1) describes the in-plane disorder in terms of scattering between exciton states at different wave vectors. The coefficient $V(\mathbf{k} - \mathbf{k}')$ is the Fourier transform of the disordered two-dimensional potential $V(\mathbf{R})$, which is an effective potential acting on the exciton center-of-mass motion. This formulation implies the assumption that the disorder does not affect the exciton internal degrees of freedom [1], which is valid provided the fluctuation amplitude of the energy gap along the QW plane is smaller than the exciton confinement energy. This requirement is always met by good quality samples.

The coefficient $V(\mathbf{k})$ characterizes the disorder and, in general, depends on the specific realization of the disordered system. However, for most purposes, it is possible to use a statistical description of the disordered potential which has the advantage of accounting for the important parameters characterizing the disorder, without including its microscopical details. We assume, as in Ref. [4], that all the n-point correlation functions of the exciton potential $V(\mathbf{R})$ factorize as products of two-point correlation functions. The second order correlation is written as

$$g(\mathbf{R} - \mathbf{R}') = \langle V(\mathbf{R}) V(\mathbf{R}') \rangle, \quad (2)$$

where the brackets mean configuration average. Then the potential $V(\mathbf{R})$ can be modeled as

$$V(\mathbf{R}) = \sum_{\mathbf{k}} c(\mathbf{k}) [\tilde{g}(\mathbf{k})]^{\frac{1}{2}} \exp(-i\mathbf{k} \cdot \mathbf{R}), \quad (3)$$

where $\tilde{g}(\mathbf{k})$ is the Fourier transform of the correlation function (2) and $c(\mathbf{k})$ is a random function with complex values on the unitary circle, which is delta correlated, namely, $\langle c^*(\mathbf{k}) c(\mathbf{k}') \rangle = \delta_{\mathbf{k},\mathbf{k}'}$. Moreover, since $V(\mathbf{R})$ is a real quantity, the condition $c(\mathbf{k}) = c^*(-\mathbf{k})$ must be obeyed. The function $c(\mathbf{k})$ provides the random character of the potential $V(\mathbf{R})$, while the correlation function $g(\mathbf{R})$ accounts for the significant parameters such as the fluctuation amplitude and the correlation length. We assume a Gaussian correlation $\tilde{g}(\mathbf{k}) = v_0^2 e^{-\frac{1}{2}\lambda_c^2 k^2}$, where λ_c and v_0 are the correlation length and the average amplitude of the fluctuations of $V(\mathbf{R})$, respectively. The two parameters λ_c and v_0 can be related to measurable quantities by introducing the potential density

$$P(\omega) = \frac{\pi}{L^d} \int \delta(\hbar\omega - V(\mathbf{R}))\, d\mathbf{R}, \quad (4)$$

where L represents a quantization length for the exciton center-of-mass motion and d is the dimensionality of the system ($d = 2$ in the QW case). It has been shown [3,4] that in the limit of infinite exciton mass the potential density $P(\omega)$ corresponds to the exciton density of states (in the absence of a MC) and gives the line shape of the excitonic transition. In this case, the variance of the distribution (4), which we denote by σ, defines the exciton inhomogeneous broadening. In the case of finite exciton mass, the exciton line turns out to be narrower than σ [3], which is a more rigorous definition of the motional narrowing effect. The quantity σ can however be taken as a figure of merit of the amount of disorder present in the system. With the assumption of Gaussian correlation, from (3) one obtains [4]

$$\sigma^2 = g(0) = v_0^2 \left(\frac{L}{\lambda_c} \frac{1}{\sqrt{2\pi}} \right)^d. \quad (5)$$

Our aim is to calculate the linear optical response of the system. To this purpose, we work out the cavity photon propagator $D(\mathbf{k}, \mathbf{k}', \omega)$, which is the Fourier transform of the probability per unit time of a photon being scattered from $\mathbf{k}$ to $\mathbf{k}'$. It obeys the following Dyson equation:

$$D(\mathbf{k}, \mathbf{k}', \omega) = D^{(0)}(\mathbf{k}, \omega) \delta_{\mathbf{k},\mathbf{k}'} + \sum_{\mathbf{k}''} D^{(0)}(\mathbf{k}, \omega) \times C_{\mathbf{k}}^* G(\mathbf{k}, \mathbf{k}'', \omega) C_{\mathbf{k}''} D(\mathbf{k}'', \mathbf{k}', \omega), \quad (6)$$

where $D^{(0)}(\mathbf{k}, \omega) = (\hbar v \sqrt{k^2 + k_z^2} - \hbar\omega + i\epsilon)^{-1}$ is the free photon propagator and $G(\mathbf{k}, \mathbf{k}', \omega)$ is the exciton propagator in the presence of disorder which, in turn, obeys the following Dyson equation:

$$G(\mathbf{k}, \mathbf{k}', \omega) = G^{(0)}(\mathbf{k}, \omega) \delta_{\mathbf{k},\mathbf{k}'} + \sum_{\mathbf{k}''} G^{(0)}(\mathbf{k}, \omega) V(\mathbf{k} - \mathbf{k}'') G(\mathbf{k}'', \mathbf{k}', \omega). \quad (7)$$

In the above equation, $G^{(0)}(\mathbf{k}, \omega) = (\omega_{\mathbf{k}} - \omega + i\epsilon)^{-1}$ is the free exciton propagator.

The numerical solution of Eqs. (6) and (7) in a two-dimensional $\mathbf{k}$ space requires a huge computational effort. Since we want to include multiple scattering to all orders into the calculations, a perturbative approach is not adequate. We thus restrict ourselves to the problem in one-dimensional $\mathbf{k}$ space. The analogous problem for a bare QW has been solved both in one [4] and two [5] dimensions, and only quantitative differences between the 1D and 2D absorption spectra emerge from the calculations. We choose $\lambda_c = 140$ Å. This assumption is justified by the fact that every short-range fluctuation of the energy gap is averaged out by the finite space extent of the exciton envelope function [1,3]. Thus, only fluctuations larger than the Bohr radius a_B contribute to the exciton center-of-mass potential $V(\mathbf{R})$. The Dyson equations are solved over an interval of 2.5×10^6 cm^{-1} around $k = 0$ by taking 400 sample points. This corresponds to a quantization length in real space $L = 10^4$ nm.

The coupling to the external radiation is introduced by means of the quasimode formalism [6]. The coupling between the cavity mode and the external radiation field, which takes place through the cavity mirrors, is applied as a first order perturbation over H_{pol}. The forward scattering amplitude is finally obtained in terms of the cavity photon propagator as

$$S_k(\omega) = 1 - i\gamma_c(k)D(k,k,\omega), \tag{8}$$

whose square modulus gives the MC transmission spectrum at incident wave vector k. In this expression, $\gamma_c(k)$ is the cavity mode broadening which enters through the quasimode formalism. In our calculations we perform configuration averages of the square modulus of (8) in order to reproduce the macroscopic spectra [4,5].

The transmission spectra of polaritons in a MC-embedded QW are calculated for different values of the disorder parameter σ and normal incidence. The value used for the cavity mode broadening is $\gamma_c(0) = 0.7$ meV and the polariton coupling is chosen such that the system is in the strong coupling regime. The bare exciton energy $\hbar\omega_0 = 1.5$ eV is resonant with the cavity mode at $k = 0$ and the exciton mass is $M = 0.25 m_e$. In Fig. 1 we plot the linewidths of the two polariton peaks as a function of σ. It appears that the two polariton lines become broader as σ increases. Moreover, the pronounced asymmetry of the transmission spectrum is clearly seen in the inset of Fig, 1. In the absence of motional narrowing, the polariton lineshape is given by the convolution of a Lorentzian of width $\gamma_c(0)/2$ and a Gaussian of width $\sigma/2$

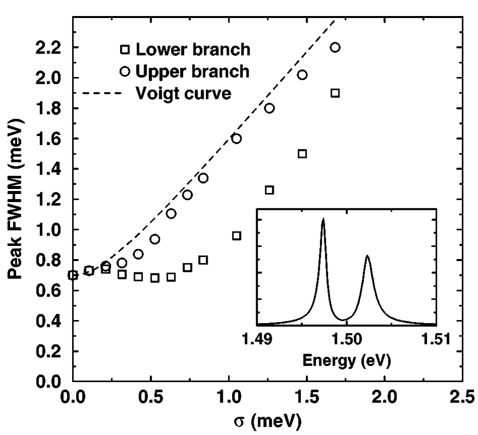

FIG. 1. Transmission peak linewidth calculated at resonance as a function of σ. Circles and squares indicate upper and lower polariton branches, respectively. The dashed line corresponds to the Voigt linewidth described in the text. The inset shows a typical transmission spectra for $\sigma = 0.7$ meV.

(Voigt profile) [11]. This linewidth is plotted in Fig. 1 as a function of σ. We see that both polariton branches exhibit a *subaverage broadening* behavior; namely, their linewidths lie below the Voigt linewidth. Another important characteristic is that the lower polariton peak is always narrower than the upper one, whose linewidth follows more closely the Voigt linewidth. Both these features can be attributed to a motional narrowing of the polariton modes.

In order to explain the difference in the broadenings of the two polaritons, we rewrite H_{pol} in terms of the polariton operators (which would be the eigenstates in absence of the disorder term) $B_{l,k} = W_l(k)a_k + X_l(k)b_k$, where $W_l(k)$ and $X_l(k)$ are the Hopfield coefficients which express the photon and exciton fraction in the polariton mode respectively. We have

$$H_{\mathrm{pol}} = \sum_{l,k} \hbar\Omega_{l,k} B^\dagger_{l,k} B_{l,k} + \sum_{l,k}\sum_{l',k'} V(k-k')X_l(k)X^*_{l'}(k')B^\dagger_{l,k}B_{l',k'}, \tag{9}$$

where the indices l and l' run over the two polariton branches. The lower polariton exciton fraction $X_1(k)$ varies from $1/\sqrt{2}$ to 1 in a region of the order of $\Delta k \sim (M_c\Omega_R/\hbar^2)^{\frac{1}{2}}$, where $M_c = \hbar k_z/v$ is the cavity mode mass at $k = 0$. In correspondence, the upper polariton quantity $X_2(k)$ varies from $1/\sqrt{2}$ to 0. From the polariton Hamiltonian we derive the corresponding eigenvalue equation for the center-of-mass motion of the two polaritons which, in k space, reads

$$\Omega_{l,k}\phi_l(k) + \sum_{l',k'} V_{l,l'}(k,k')\phi_{l'}(k') = \mathcal{E}\phi_l(k), \tag{10}$$

where the effective potential $V_{l,l'}(k,k') = V(k-k')X_l(k)X^*_{l'}(k')$ has been introduced. This potential depends separately on k and k' and the corresponding potential in real space is nonlocal. Let us disregard for the moment the cross terms in (10). We are left with two separate equations for the two branches. The equation for the lower branch, with $l, l' = 1$, is characterized by an effective potential $V_{1,1}(k,k') = V(k-k')X_1(k)X^*_1(k')$ which has essentially the same k space extension as the exciton potential $V(k-k')$, given by λ_c^{-1}. Within this region, the lower polariton dispersion is mostly excitonlike, because $\Delta k \lambda_c \ll 1$. Thus, the lower polariton is subject to the same motional narrowing effect as the bare exciton and the narrowing with respect to the Voigt linewidth follows. In the case of the upper polariton, the extension in k space of $V_{2,2}(k,k')$ significantly reduced by the factor $X_2(k')$ with respect to $V(k-k')$. The new correlation length is consequently given by $\lambda'_c = 2\pi/\Delta k \gg \lambda_c$. Moreover, the polariton mass along its dispersion is always close to the $k = 0$ value $2M_c$. It can be verified that the rescaling of the correlation length and of the mass in the equation of motion compensate and that, again, the same motional narrowing is expected. The different linewidths obtained in the numerical calculations indicate that the cross terms in Eq. (10) play a crucial role. The cross term with $l = 1$ and $l' = 2$ is negligible because $\Delta k \lambda_c \ll 1$ and the above considerations for the lower polariton are still valid. For the same reason, the other cross term actually appears to be even more important than the diagonal term with $l, l' = 2$. We thus argue that this term causes the additional broadening obtained in the numerical calculation. The above derivation differs significantly from that in Ref. [10] in the fact that the k dependence of the coefficients $X_l(k)$, the exact polariton dispersion, and the coupling between the two branches are fully taken into account. All these contributions are fundamental for the nonperturbative approach which includes multiple scattering to all orders.

Finally, in Fig. 2 we compare the results of the model with the experimental data presented in Ref. [10]. For this calculation, the parameter $\sigma = 1.4$ meV was used. This value has been chosen in order to reproduce the full width at half maximum of 3.1 meV, measured independently in Ref. [10] for the bare exciton transition. The cavity mode linewidth is $\gamma_c(0) = 0.8$ meV. The linewidths obtained by a simple linewidth averaging argument have been plotted for comparison. The present results are in very good agreement with the measurements by Whittaker *et al.* The main discrepancy appears for the lower polariton and for positive detuning. We argue that this discrepancy originates from having used a one-dimensional model for the derivation of the spectra. In fact, as showed by Glutsch *et al.* [5], the motional narrowing effects are more pronounced in 1D than in 2D systems. Another source of discrepancy comes from the way detuning is varied in the sample of Ref. [10]. Since the MC is not wedge shaped, the exciton energy is tuned by varying the temperature. This, however, implies an increase of the exciton homogeneous broadening for positive detuning (higher temperature), an effect which is not included in the model.

In conclusion, we have presented a model of MC polaritons in the presence of in-plane disorder in the QW. The numerical solution of the coupled exciton-photon equations reveals that polariton modes are subject to a motional narrowing effect analogous to the one acting on exciton states in bare QWs. It turns out that, in the strong coupling regime, the motional narrowing occurs mainly for the lower polariton branch. The upper polariton, on the other hand, is affected by multiple scatterings occurring between the two branches which cause additional broadening. The solution in one-dimensional k space allows a very clear explanation of recently measured data on high-quality samples.

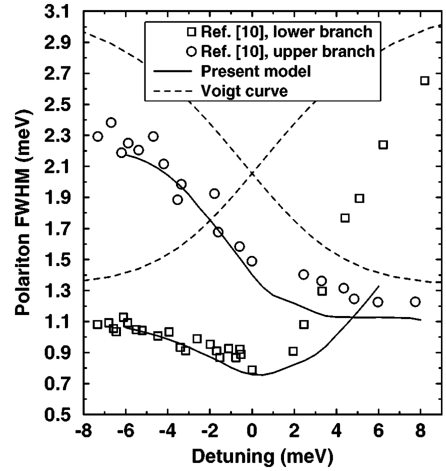

FIG. 2. Comparison between the calculated and measured [10] polariton linewidths as a function of the detuning. The dashed lines reproduce the results obtained from a simple linewidth averaging argument.

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[1] R. Zimmermann, Phys. Status Solidi (b) **173**, 129 (1992).
[2] R. Zimmermann, Nuovo Cimento D **17**, 1801 (1995).
[3] F. Grosse and R. Zimmermann, Superlattices Microstruct. **17**, 439 (1995).
[4] S. Glutsch and F. Bechstedt, Phys. Rev. B **50**, 7733 (1994).
[5] S. Glutsch, D. S. Chemla, and F. Bechstedt, Phys. Rev. B **54**, 11 592 (1996).
[6] V. Savona and C. Weisbuch, Phys. Rev. B **54**, 10 835 (1996).
[7] R. Houdré, R. P. Stanley, and M. Ilegems, Phys. Rev. A **53**, 2711 (1996).
[8] V. Savona, F. Tassone, C. Piermarocchi, A. Quattropani, and P. Schwendimann, Phys. Rev. B **53**, 13 051 (1996).
[9] B. L. Halperin, Phys. Rev. **139**, A104 (1965).
[10] D. M. Whittaker, P. Kinsler, T. A. Fisher, M. S. Skolnick, A. Armitage, A. M. Afshar, M. D. Sturge, and J. S. Roberts, Phys. Rev. Lett. **77**, 4792 (1996).
[11] P. Kinsler and D. M. Whittaker, Phys. Rev. B **54**, 4988 (1996).