Linewidth narrowing of polaritons

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Polaritons are formed when the electromagnetic field couples to excitons in a semiconductor. In their high-quality semiconductor quantum microcavities, Fisher *et al.*[5] observe an unexpected decrease in the polariton linewidth (the full width at half maximum) near resonance. This paper uses Green's-function methods to show how the exciton linewidth and the cavity losses combine together, and that they can give a polariton linewidth reduced below that of either of its components. The quantum well disorder is described as a spatially varying potential due to both alloy fluctuations and well width variation, which causes the exciton component to become broadened. [S0163-1829(96)07931-3]

I. INTRODUCTION

The interaction between the electromagnetic field in semiconductor microcavities and quantum well excitons is a topic of active research.[1–4] Polaritons are formed when the electromagnetic field couples to excitons in a semiconductor producing a coherent superposition of the photon and exciton modes. In a quantum structure, where a microcavity is coupled to a quantum well exciton, these polaritons are localized by the well layer in the growth direction, but unconfined in the in-plane direction. If the exciton is tuned through the cavity frequency, resonant coupling effects cause anticrossing or level repulsion between the polaritons to occur.

The energies of the polaritons depend mainly on the cavity mode and exciton energies. An uncoupled cavity mode has some spectral width because of its losses. An uncoupled exciton is broadened because it is affected by losses due to recombination and by imperfections and other disorder in the quantum well. When the reflectivity spectrum of the system is measured, each polariton has a dip around its central frequency, with a particular line shape and linewidth. The naive expectation might well be that the linewidth of the polaritons would be just that of the weighted averages of the independent cavity and exciton linewidths, where the weights are the probabilities of the cavity mode and exciton components.

In contrast to this, the observations by Fisher *et al.*[5] in their high-quality semiconductor structures show a marked decrease in the spectral full width at half maximum (FWHM) of the polaritons near resonance–the polariton linewidths are seen to be less than the average values. Well disorder is by far the largest contributor to the width of the exciton in these results because of the alloy fluctuations in the well material and interface fluctuations at the well edges.

These results motivated us to develop a simple theoretical model that combines (in Sec. V) the effects of the losses from the microcavity (Sec. III) and the disorder in the quantum well (Sec. IV) in a systematic way, while properly allowing for the superposition of cavity and exciton that makes up each polariton. The theory can not only predict ''less than average'' linewidths like those seen in the experiments of Fisher *et al.*, but makes the more dramatic prediction that the polariton linewidth can in fact be less than that of either the cavity or exciton. Other theories that consider the broadening of polaritons are found in Refs. 6 and 7. The first of these includes Gaussian disorder and dispersion, but does not derive a method of combining the cavity and disorder broadening. The second considers a different system to that in this paper, with isolated excitons and no cavity mode dispersion.

The theory in this paper starts with an idealized polariton system, to which we add cavity losses and then well disorder. A Green's function method is used, where the polariton linewidth is calculated by first adding a simple loss interaction. Following this, we add the exciton scattering interaction caused by a random potential that varies with position in the quantum well. The disorder term is based on that used by Glutsch and Bechstedt,[8] and here we incorporate their exciton theory into one which describes the polariton spectrum. The theory presented here is intended to demonstrate the physics clearly, and so is more approximate than is strictly necessary. However, the character of the predictions are not significantly affected.

II. AN IDEAL POLARITON

A polariton consists of an exciton coupled to a cavity mode. A full model would include a description of the different types of excitons, and their excited states, as well as all the cavity modes and the couplings between these. However, in practice we know that excitons will only couple to a cavity mode if they have matching wave vectors. Also, for low excitation strengths only the lowest-energy exciton is significant. Further, maintaining the low level of excitation means that the exciton can be adequately described as a boson. This is because the low density of excitons means that any phase space filling[9] or exciton-exciton scattering processes are unlikely to occur. This means we can consider an ''ideal polariton,'' consisting of an idealized quantum well exciton, an idealized cavity mode, and the exciton to cavity mode coupling term between the two. To express the exact coupling term in a simple form, we use the[10] coulomb gauge, the dipole approximation, and the rotating wave approximation.

The resulting model consists of two harmonic oscillator boson modes, one for the exciton, and one for the cavity mode. These are coupled together by the cavity-exciton interaction. In the Schrödinger picture it is

$$\overline{H_{P0}}=\sum_k \hbar\Omega(k)\overline{e^\dagger}(k)\overline{e(k)}+\hbar\omega(k)\overline{a^\dagger}(k)\overline{a(k)}+\hbar A(k) \times[\overline{e^\dagger}(k)\overline{a(k)}+\overline{e(k)}\overline{a^\dagger}(k)], \quad (2.1)$$

where $\overline{e^\dagger}(k)$, $\overline{e(k)}$ are the exciton mode operators and $\overline{a^\dagger}(k)$, $\overline{a(k)}$ are the operators for the field in the cavity mode. The parameter $A(k)$ is the coupling between the cavity mode and the exciton. It is in units of frequency, and is equal to half the splitting between the pair of polaritons at resonance, that is, when $\Omega(k)=\omega(k)$. The variable k corresponds to the in-plane wave vector of the modes.

This two-mode system can be diagonalized, and then be written in terms of new polariton operators, which describe the two new polariton modes that can be used in place of the original exciton and cavity modes. To diagonalize, we derive the Heisenberg equations for the system. These Heisenberg operators are generated from the Schrödinger operators by the Hamiltonian H_{P0}. The new operators are

$$\hat{e}(k;t)=\exp(i\overline{H_{P0}}t/\hbar)\overline{e(k)}\exp(-i\overline{H_{P0}}t/\hbar),$$
$$\hat{a}(k;t)=\exp(i\overline{H_{P0}}t/\hbar)\overline{a(k)}\exp(-i\overline{H_{P0}}t/\hbar), \quad (2.2)$$

and the equations of motion are

$$\frac{d}{dt}\hat{a}(k;t)=-i\omega(k)\hat{a}(k;t)-iA(k)\hat{e}(k;t),$$
$$\frac{d}{dt}\hat{e}(k;t)=-iA(k)\hat{a}(k;t)-i\Omega(k)\hat{e}(k;t). \quad (2.3)$$

This model could be easily extended to include the effects of cavity losses, but for clarity we will add the losses as an interaction in the next section. This makes it possible to describe both interactions, the cavity losses and the well disorder on an equal footing, and using the same method. It also means that the mechanism for combining the two broadening effects results naturally from the theory. The solution of the equations of motion is just a simple eigenvalue problem. The two eigenvalues (or energies) of the eigenmodes (the polaritons) are

$$2\epsilon_{0\pm}(k)=\Omega(k)+\omega(k)\pm\sqrt{[\Omega(k)-\omega(k)]^2+4A(k)^2}. \quad (2.4)$$

These two polaritons are just the dressed states of the coupled cavity-exciton mode system. They are distinguished by a label $\pm$ denoting the upper $(+)$ or lower $(-)$ polariton branch, and their wave number k. If we introduce the operators $\overline{p_\pm^\dagger}(k)$, $\overline{p_\pm}(k)$ to describe the polaritons, the eigenvectors of the system describe how the cavity and exciton operators combine to form the polariton operators. The polariton operators for the coupled cavity-exciton system are $\overline{p_\pm^\dagger}(k)$, $\overline{p_\pm}(k)$. The cavity and exciton modes have coupled together to form two new polariton modes. The polariton operators in the interaction picture generated by the ideal H_{P0} Hamiltonian are related to the exciton and cavity operators by the following expressions:

$$\hat{e}(k;t)=d_+(k)\hat{p}_+(k;t)+d_-(k)\hat{p}_-(k;t),$$
$$\hat{a}(k;t)=c_+(k)\hat{p}_+(k;t)+c_-(k)\hat{p}_-(k;t), \quad (2.5)$$

where

$$d_\pm(k)=\mp c_\mp(k)=\pm\frac{1}{\sqrt{2}}\sqrt{1\pm\sin[\alpha(k)]},$$
$$\tan[\alpha(k)]=\frac{\Omega(k)-\omega(k)}{2A(k)}. \quad (2.6)$$

The interaction picture operators for the ideal polariton can be related to equivalent Schrödinger picture operators:

$$\hat{p}_\pm(k;t)=\exp(i\overline{H_{P0}}t/\hbar)\overline{p_\pm}(k)\exp(-i\overline{H_{P0}}t/\hbar) =\hat{p}_\pm(k;0)\exp[-i\epsilon_{0\pm}(k)t], \quad (2.7)$$

and so the Schrödinger picture Hamiltonian in terms of the polariton operators is

$$\overline{H_{P0}}=\hbar\sum_k [\epsilon_{0+}(k)\overline{p_+^\dagger}(k)\overline{p_+}(k)+\epsilon_{0-}(k)\overline{p_-^\dagger}(k)\overline{p_-}(k)]. \quad (2.8)$$

The Green's-functions for a polariton in the idealized system is denoted $G_{0\pm}(k;t-t')$ or $G_{0\pm}(k;\lambda)$ when fourier-transformed into the frequency domain. If $\hat{\mathbf{T}}$ is the time-ordering operator, the Green's-function is defined as

$$\langle\hat{\mathbf{T}}\hat{p}_\pm(k;t)\hat{p}_\pm^\dagger(k';t')\rangle=\delta_{kk'}G_{0\pm}(k;t-t'). \quad (2.9)$$

III. LOSSES IN THE OPTICAL CAVITY

A model of the real polariton must include the effect of the cavity losses. In this section we expand the effect of the cavity losses as a Taylor series in ideal polariton Green's functions $G_{0\pm}$. In Green's-function calculations, the losses are typically inserted by simply adding an imaginary part to the mode frequency. This contrasts with more systematic approaches using reservoir techniques,[11] but these do not apply directly to Green's function methods. However, similar methods can be used by adding a suitable interaction term and applying approximations equivalent to those used in the reservoir techniques.

However, here we introduce an idealized scattering model which leads to the same predictions of a Lorentzian line shape and exponential decay. We do this by simply adding an imaginary part to the frequency, which we treat as an interaction. This enables us to easily combine the loss and quantum well disorder interactions together in Sec. V.

This model of the losses correctly predicts the exponential decay of the correlations in the cavity mode. However, it does not include the noiselike effects caused by the loss coupling individual quanta out of the cavity mode at random times. Despite this, it can still give the correct answers because the cavity mode-exciton mode coupling is linear, which contrasts with the situation in nonlinear systems where this does not hold (for example see Refs. 12 and 13).

The Hamiltonian in the Schrödinger picture of a polariton system that includes the cavity losses is

$$\overline{H_{PL}}=\overline{H_{P0}}+\hbar\overline{V_{PL}},$$

$$\overline{V_{PL}}=\sum_k \overline{V_L}(k)=\sum_k -i\gamma(k)\overline{a^\dagger}(k)\overline{a}(k). \quad (3.1)$$

As in the previous section, we use the interaction picture generated by the ideal $\overline{H_{P0}}$ Hamiltonian. The interaction Hamiltonian is

$$\hat{V}_{PL}(t)=\sum_k \hat{V}_L(k;t)=\sum_k -i\gamma(k)\hat{a}^\dagger(k;t)\hat{a}(k;t). \quad (3.2)$$

The form of the cavity loss term can be easily rewritten in terms of the polariton operators using Eq. (2.5). Before proceeding further, we will restrict this calculation to the case where the two polaritons are widely separated as compared to the linewidth caused by the losses. This is not a necessary approximation, but it does make the following calculations much simpler. Since this theory is motivated by a desire to explain the general principle of how the broadening interactions combine together, this simplicity is an advantage. First we will convert the exact cavity loss term into the polariton operators. The result is

$$\begin{aligned}\hat{V}_L(k;t)&=-i\gamma(k)\hat{a}^\dagger(k;t)\hat{a}(k;t)\\&=-i\gamma(k)[c_+^*(k)\hat{p}_+^\dagger(k;t)+c_-^*(k)\hat{p}_-^\dagger(k;t)]\\&\quad\times[c_+(k)\hat{p}_+(k;t)+c_-(k)\hat{p}_-(k;t)]. \quad (3.3)\end{aligned}$$

For widely separated polaritons, the time varying exponential terms due to the difference in the energies of the two polaritons will oscillate rapidly [note Eq. (2.7)]. For times much longer than the period of this rapid oscillation, the oscillations will average to zero. The remaining loss term is just

$$\begin{aligned}\hat{V}_L(k;t)\approx&-i\gamma(k)[c_+^*(k)c_+(k)\hat{p}_+^\dagger(k;t)\hat{p}_+(k;t)\\&+c_-^*(k)c_-(k)\hat{p}_-^\dagger(k;t)\hat{p}_-(k;t)]. \quad (3.4)\end{aligned}$$

The convenience of this approximation is now easy to see. The exciton interaction due to the cavity losses has no terms coupling the upper and lower polaritons together. Consequently, the effect of the loss decomposes into two independent parts — one for the upper polariton, and another for the lower polariton.

We can now write the interaction Hamiltonian for either of the polaritons as

$$\hat{V}_{L\pm}(t)=-i\sum_k c_\pm^*(k)c_\pm(k)\gamma(k)\hat{p}_\pm^\dagger(k;t)\hat{p}_\pm(k';t). \quad (3.5)$$

We now represent the Green's functions for the polariton affected by the cavity losses $G_{L\pm}$ (the ''lossy polariton'') in terms of the Green's functions of the unaffected polariton $G_{0\pm}$. To do this we expand the Green's functions for the lossy polariton $G_{L\pm}$ in a Taylor series in powers of the matrix elements of the loss interaction $\hat{V}_{L\pm}$. The Taylor series for this Green's function is

$$G_{L\pm}(k;t-t')=\sum_{m=0}^{\infty} G_{L\pm}^{(m)}(k;t-t'), \quad (3.6)$$

$$G_{L\pm}^{(m)}(k;t-t')=\left\langle \hat{\mathbf{T}}\hat{p}_\pm(k;t)\prod_{j=1}^{m}\left[-i\sum_{k_j,t_j}\hat{p}_\pm^\dagger(k_j;t)c_\pm^*(k_j)c_\pm(k_j)\gamma(k_j)\hat{p}_\pm(k_j';t)\right]\hat{p}_\pm^\dagger(k;t')\right\rangle. \quad (3.7)$$

Once the expectation value of this is taken, Wick's theorem can be applied to expand the $2(m+1)$th order moments over the vacuum state into a sum of products of all unique pairings of second-order moments. Note that for Wick's theorem to apply, the quantum expectation value needs to be taken over the vacuum state. In this case, the $\hat{p}_\pm(t),\hat{p}_\pm^\dagger(t')$ operators can be viewed as creating the polariton we are attempting to describe out of this vacuum at t', then destroying it again at the later time t. The application of Wick's theorem leaves us with a single fully connected term of each order. The disconnected terms are reassigned to become part of the vacuum contribution for lower-order terms.

The single remaining connected term has $m+1$ terms like $\hat{p}_\pm\hat{p}_\pm^\dagger$ and these become $m+1$ factors of $G_{0\pm}$ from Eq. (2.9). Similarly, we can see from Eq. (3.7) that every one of the m factors of $-i\gamma$ is associated with one like $|c_\pm|^2$. This means that the final result is identical to the expression obtained for just a cavity mode with losses — except that the decay rate of the polariton is a factor of $|c_\pm|^2$ smaller. After transforming the terms into frequency space, we find the Green's function Taylor series for the lossy polariton spectrum is

$$G_{L\pm}^{(m)}(k,\lambda)=[-i|c_\pm(k)|^2\gamma(k)]^m G_{0\pm}(k;\lambda)^{m+1}. \quad (3.8)$$

The spectral function contains the amount and type of broadening added to the polariton by the cavity losses. The Lehman representation[14] expresses this lossy Green's function in powers of the unperturbed Green's function:

$$G_{L\pm}(k;\lambda)=\sum_{m=0}^{\infty} S_{L\pm}^{(m)}(k)G_{0\pm}(k;\lambda)^{m+1}. \quad (3.9)$$

The calculated expression for the lossy polariton can be converted into this form, and gives the spectral moments

$$S_{L\pm}^{(m)}(k)=[-i|c_\pm(k)|^2\gamma(k)]^{m+1}, \quad (3.10)$$

which are just those for a Lorentzian of width $\sigma_{L\pm}=|c_{\pm}(k)|^2\gamma(k)$. The spectral function can therefore be written

$$S_{L\pm}(k,\lambda)=\frac{\sigma_{L\pm}}{\pi}\frac{1}{\lambda^2+\sigma_{L\pm}^2}. \tag{3.11}$$

As expected, this has shown that the losses have a Lorentzian spectral function, which gives rise to a Lorentzian cavity mode spectrum. For an uncoupled cavity mode we can easily recover the standard result in the limit as $c_{\pm}(k)\to 1$. How the decay rate $\gamma(k)$ varies with in-plane wave vector k is not considered here, but has been treated by Citrin.[15] In the system considered here, the simple model of the loss has given us the same answers as a more rigorous treatment would have, as was noted above.

IV. DISORDER IN THE QUANTUM WELL

A model of the real polariton must also include the effect of the well disorder. In this section we expand the effect of the well disorder as a Taylor series in the Green's functions of the lossy polariton $G_{L\pm}$. Following the approach of Glutsch and Bechstedt,[8] we use a Gaussian model of the disorder, where the matrix elements that couple excitons of different in-plane momenta have a random Gaussian distribution. This type of disorder term can result from a model of the quantum well which has small Gaussian fluctuations in its width and depth. We assume that the scale of the disorder is small enough so that the matrix elements are δ correlated in space.

In this section the final approximation we will make is to assume a dispersionless polariton. Calculations including dispersion could be done numerically, but here we are interested mainly in the fraction of the disorder that affects the polarition, and in how the broadening due to disorder combines with the cavity loss linewidth. We leave such calculations for subsequent work.

The Hamiltonian in the Schrödinger picture of a polariton system that includes the well disorder is

$$\overline{H_{PE}}=\overline{H_{PL}}+\hbar\overline{V_{PE}},\quad \overline{V_{PE}}=\sum_k\sum_{k'}\overline{V_E}(k-k'). \tag{4.1}$$

Using the interaction picture generated by the ideal $\overline{H_{P0}}$ Hamiltonian, the interaction Hamiltonian is

$$\hat{V}_{PE}(t)=\sum_k\sum_{k'}\hat{V}_E(k-k';t)$$
$$=\sum_k\sum_{k'}\tilde{V}_E(k-k')\hat{e}^{\dagger}(k;t)\hat{e}(k;t). \tag{4.2}$$

The well disorder is assumed to cause a Gaussian variation to the exciton energies, so the properties of $\tilde{V}_E(k-k')$ are controlled by its variance. An average over all space, or over all possible configurations of the random potential, is denoted $\langle\cdots\rangle_{\text{space}}$. The variance is

$$\langle\tilde{V}_E(k_1-k_2)\tilde{V}_E(k_3-k_4)\rangle_{\text{space}}$$
$$=\tilde{\psi}_E(k_1-k_2)\,\delta(k_1-k_2,k_3-k_4). \tag{4.3}$$

The well disorder term can be easily rewritten in terms of the polariton operators using Eq. (2.5). This step assumes that the loss interaction has not affected the composition of the polariton — that is, that the coefficients $d_{\pm}(k),c_{\pm}(k)$ derived in the section on the ideal polariton remain valid for the polariton with cavity losses added. It is consistent with the widely separated polariton or rotating wave type approximation used when adding the losses. An important consequence is that it allows us to add the effects of disorder to the lossy polariton in the same way as we have added the effects of cavity loss to the ideal polariton — without requiring new coefficients $c'_{\pm},d'_{\pm}$ for the fraction of cavity mode and exciton mode in the lossy polariton to be calculated.

Before proceeding we restrict this calculation to the case where the two polaritons are widely separated as compared to their linewidth caused by the disorder. First we will convert the exact exciton disorder term into the polariton operators. The result is

$$\hat{V}_E(k-k';t)=\tilde{V}_E(k-k')\hat{e}^{\dagger}(k;t)\hat{e}(k';t)=\tilde{V}_E(k-k')$$
$$\times[d_+^*(k)\hat{p}_+^{\dagger}(k;t)+d_-^*(k')\hat{p}_-^{\dagger}(k;t)]$$
$$\times[d_+(k)\hat{p}_+(k;t)+d_-(k')\hat{p}_-(k;t)]. \tag{4.4}$$

For widely separated polaritons, the time-varying exponential terms due to the difference in the energies of the two polaritons will oscillate rapidly [note Eq. (2.7)]. For times much longer than the period of this rapid oscillation, the oscillations will average to zero. The remaining disorder term is just

$$\hat{V}_E(k-k';t)\approx\tilde{V}_E(k-k')\{d_+^*(k)d_+(k')\hat{p}_+^{\dagger}(k;0)\hat{p}_+(k';0)$$
$$\times\exp[-i[\epsilon_{+0}(k')-\epsilon_{+0}(k)]t]$$
$$+d_-^*(k)d_-(k')\hat{p}_-^{\dagger}(k;0)\hat{p}_-(k;0)$$
$$\times\exp[-i[\epsilon_{-0}(k')-\epsilon_{-0}(k)]t]\}. \tag{4.5}$$

Just as in the case of the cavity loss interaction, the convenience of this approximation is easy to see. The exciton interaction due to the well disorder has no terms coupling the upper and lower polaritons together. Consequently, the effect of the disorder decomposes into two independent parts — one for the upper polariton, and another for the lower polariton.

We can now write the interaction Hamiltonian for either of the polaritons as

$$\hat{V}_{E\pm}(t)=\sum_k\sum_{k'}\hat{p}_{\pm}^{\dagger}(k;t)[d_{\pm}^*(k)d_{\pm}(k')\tilde{V}_E(k-k')]$$
$$\times\hat{p}_{\pm}(k';t). \tag{4.6}$$

The Green's functions for the whole system, including the well disorder and the cavity losses is denoted $G_{\pm}(k_1,k_2;t-t')$. Again following Ref. 8 we neglect parts of the Green's function that have elements that represent transitions that end with a different wave number k' than their starting k. We justify this by noting that for reflectivity

measurements, the incoming light has a fixed k. Since the measured intensity is dominated by elastically scattered light, this has the same k.

We now represent the Green's functions $G_{\pm}$ for the polariton affected by the well disorder and losses (the ''real polariton'') in terms of the Green's functions of the lossy polariton $G_{L\pm}$. To do this we expand $G_{\pm}$ in a Taylor series in powers of the matrix elements of the disorder interaction $\hat{V}_{E\pm}$. The Taylor series for this Green's function, to be averaged over all possible spatial configurations of the disorder, is

$$G_{\pm}(k_0;t-t')=\sum_{m=0}^{\infty} G_{L\pm}^{(m)}(k_0;t-t'), \tag{4.7}$$

$$G_{\pm}^{(m)}(k_0;t-t')$$

$$=\left\langle \hat{\mathbf{T}}\hat{p}_{\pm}(k_0;t)\left\langle \prod_{j=1}^{m}\left[-i\times\sum_{k_j,k_j',t_j}\hat{p}_{\pm}^{\dagger}(k;t)d_{\pm}^{*}(k_j)d_{\pm}(k_j')\right.\right.\right.$$

$$\left.\left.\left.\times\tilde{V}_E(k_j-k_j')\hat{p}_{\pm}(k';t)\right]\right\rangle_{\text{space}}\hat{p}_{\pm}^{\dagger}(k_0;t')\right\rangle. \tag{4.8}$$

Once the expectation value of this is taken, Wick's theorem can be applied to expand the $(m+1)$th order moments into a sum of products of all unique pairings of second-order moments. Note that for Wick's theorem to apply, the quantum expectation value needs to be taken over the vacuum state. In this case, the added $\hat{p}_{\pm}(k_0;t),\hat{p}_{\pm}^{\dagger}(k_0;t')$ operators can be viewed as creating the polariton we are attempting to describe at t', and then destroying it at the later time t.

Any term in the expansion of the Green's function has only one unique term that is fully connected after Wick's theorem has been applied to the operator expression. The disconnected terms are reassigned to become part of the vacuum contribution for lower-order terms. This leaves the evaluation of the spatial average for the remaining term. In general, for a mth order disorder term, the result is a sum of $(m-1)!!$ products of second-order terms. When m is odd, the unpaired disorder term means that the average, and hence the entire term, is zero.

This is similar to the expression obtained for an exciton interacting just with the disorder.[8] The difference is that each second order disorder term has a factor like $|d_{\pm}|^4$ associated with it, so the width of the polariton caused by the disorder is $|d_{\pm}|^2$ smaller than the width of the exciton. After transforming the terms into frequency space, and in the *dispersionless* limit, the terms in the Taylor series for the Green's function are

$$G_{\pm}^{(m)}(k_0;\lambda)=(m-1)!!\left[|d_{\pm}(k_0)|^4\psi_E(0)\right]^{m/2}$$
$$\times G_{L\pm}(k_0;\lambda)^{(m+1)}. \tag{4.9}$$

The spectral function contains the amount and type of broadening added to the polariton by the disorder in the quantum well. The Lehman representation[14] expresses this disordered Green's function in powers of the Green's function for the lossy polariton:

$$G_{\pm}(k_0;\lambda)=\sum_{m=0}^{\infty} S_{E\pm}^{(m)}(k_0)G_{L\pm}(k_0;\lambda)^{m+1}. \tag{4.10}$$

The calculated expression for the polariton under the influence of the quantum well disorder can be converted into this form. For even values of m the calculated expression for the polariton can be converted into this form, and gives the spectral moments

$$S_{E\pm}^{(m)}(k_0)=(m-1)!!\left[|d_{\pm}(k_0)|^4\psi_E(0)\right]^{m/2}. \tag{4.11}$$

This is just that for a Gaussian spectral function with variance $\sigma_{E\pm}^2=|d_{\pm}(k_0)|^4\psi_E(0)$. The spectrum can therefore be written

$$S_{E\pm}(k_0;\lambda)=\frac{1}{\sqrt{2\pi\sigma_{E\pm}^2}}\exp[-\lambda^2/2\sigma_{E\pm}^2]. \tag{4.12}$$

Although the exciton is nearly dispersionless, the cavity mode is not. As a result the polariton dispersion is not negligible, and largely depends on the fraction of cavity mode in it. One way of visualizing the effects of dispersion is to think about the spatial extent or the localization of the wave functions. This approach is useful in seeing how much of the disorder potential the polariton will feel. The total wave function of the exciton consists of an electron-hole (relative motion) part and a center of mass (combined motion) part. The electron-hole wave function is localized down to the exciton Bohr radius of about 10 nm. The center-of-mass wave function is localized to about 1 nm, because the exciton has a small dispersion, and correspondingly large effective mass. This means that the exciton ''size'' is dominated by the electron-hole size of about 10 nm. In contrast, the cavity mode has a large dispersion and correspondingly small effective mass, and so is very delocalized. This means that in a polariton the delocalization due to the photon will almost always dominate that due to the electron-hole wave function. Consequently, the size of the polariton depends mainly on the fraction of photon in it.

Including dispersion in the theory adds significant complications. The well disorder is effectively δ correlated with respect to the size of the polariton, so the dispersion occurs in the ''white noise'' limit discussed by Halperin,[16] who used an approach based on the Schrödinger equation in one dimension. The spectrum of a dispersive particle in a random potential undergoes what has been called ''motional narrowing,'' or center-of-mass averaging. This can be visualized in terms of the localization of a particle — a delocalized particle averages over more of the fluctuating potential and so it sees a smaller variation in average potential. Different-sized polaritons will undergo different amounts of averaging, and will therefore see different levels of disorder fluctuations.

Existing calculations (such as Refs. 16 and 17) present numerical results for the disorder broadening that are asymmetric with a nontrivial functional form. The spectral functions calculated are narrower than that predicted purely on the basis of the variance of the well disorder — this is due to the delocalization (or ''motional narrowing'' discussed above. For a polariton, this narrowing will vary as its degree of localization changes according to its composition. The

spectral function also has a wide tail in the higher-energy half, which has a Boltzmann character, and contrasts with the Gaussianlike nature of the rest of the spectral function. The size of this tail increases for larger disorder strengths, but decreases for larger k.

Another effect caused by the dispersion is due to the scattering of the polariton into states of different in-plane momentum (different k). These have a different energy, so the $d_{\pm}(k)$ values are different. The sum over all k values when working out the dispersive equivalent of Eq. (4.9) is complicated not only by the change of the Green's functions with k, but by the change in $d_{\pm}(k)$ also.

If we ignore the cavity loss term, Halperin's calculation can be extended to the case of polaritons[6] in a two-dimensional quantum well. The calculation shows how the amount of broadening of the polariton due to the disorder changes with the composition of the polariton. The result differs somewhat from the $|d_{\pm}|^2$ we have calculated from the dispersionless case. The difficulties of treating the effects of delocalization (or dispersion) using this Green's-function method have so far prevented us from including it in this theory directly. In any case, the delocalization adds extra narrowing, and will not destroy the narrowing predicted by these calculations — and does not affect the way the two kinds of broadening mechanism are combined in the next section.

V. THE POLARITON SPECTRUM

This section shows how the two causes of broadening combine together. The previous two sections showed individually what fractions of the cavity loss and quantum well disorder will contribute to to give the total polariton width. The mechanism for combining them is shown here to be a convolution. Together with the fractions of the two broadenings, this gives rise to ''less than average'' linewidths for the polaritons. The models of the loss and well disorder interactions are chosen to correspond to a polariton in a quantum well microcavity, but this convolution result is general, and holds for other combinations or types of interaction.

Consider a polariton in an lossy cavity, but with an ideal quantum well, only the cavity mode contributes linewidth. The losses will cause the cavity mode, and hence the polariton, to be broadened into a Lorentzian peak with a FWHM of $2|c_{\pm}(k)|^2\gamma(k)$. Alternatively, consider a polariton with a perfect cavity but with some quantum well disorder. The well disorder will cause the exciton, and hence the polariton, to be broadened into a Gaussian peak with a FWHM of $2\sqrt{2\ln(2)}|d_{\pm}(k)|^2\sqrt{\psi_E}$. In either case, one polariton will get narrower, and the other wider as the system is tuned through resonance. At resonance, where $|d_{\pm}(k)|=|c_{\pm}(k)|=1/\sqrt{2}$, the width will be one half of the maximum value.

Now consider the polariton affected by both losses and well disorder. The Green's function of the ideal polariton is $G_{0\pm}(k;\lambda)$. When the cavity losses are added to this system, this is broadened out by the spectral function of the cavity loss. The Green's function for this lossy polariton is given by Sec. III, and is

$$G_{L\pm}(k;\lambda)=\int d\Lambda S_{L\pm}(k;\Lambda)G_{0\pm}(k;\Lambda+\lambda). \quad (5.1)$$

In Sec. IV we calculated the effect of the well disorder on the lossy polariton. The Green's function for the resulting ''real'' polariton is then just the Green's function for the lossy polariton broadened out by the spectral function of the well disorder. With the same method as above, this is

$$G_{\pm}(k;\lambda)=\int d\Lambda' S_{E\pm}(k;\Lambda')G_{L\pm}(k;\Lambda'+\lambda). \quad (5.2)$$

By combining these two equations together, the real polariton Green's function $G_{\pm}$ can be expressed in terms of the ideal polariton Green's function $G_{0\pm}$ and the two spectral density functions for the cavity losses $S_{L\pm}$ and the well disorder $S_{E\pm}$. This is

$$G_{\pm}(k;\lambda)=\int d\Delta S_{\pm}(k;\Delta)G_{0\pm}(k;\Delta+\lambda),S_{\pm}(k;\Delta)$$

$$=\int d\Lambda' S_{E\pm}(k;\Lambda')S_{L\pm}(k;\Delta-\Lambda'). \quad (5.3)$$

An additional step could add the effect of homogeneous broadening of the exciton if it were necessary. The spectral function would be calculated in a similar way as for the effect of the cavity losses, and would add an extra convolution into the calculation of the polariton spectral function. However, here we consider the case where the homogeneous broadening of the exciton is negligible compared to its inhomogeneous broadening — a valid approximation for real systems.

Far away from resonance the polaritons will correspond almost completely to uncoupled exciton and cavity modes. As a result, of the two resonances that would be observed, one would correspond to that of the cavity mode and have Lorentzian shape with FWHM 2γ, and the other would correspond to the exciton with a Gaussian line shape with FWHM $2.35\sqrt{\psi_E}$.

As the exciton frequency is increased, the lower polariton gains some component from the cavity mode. As a result, its spectral peak loses some of its Gaussian nature, and gains a Lorentzian component. The converse happens to the upper polariton. Figure 1 shows how the line shape of the polariton varies as it changes from being all cavity mode (the narrow, higher peak) into being all exciton (the lowest peak). Between these two extremes, the curve is a mixture of Lorentzian and Gaussian shapes.

To get a qualitative picture of this result, imagine a system where the polariton was affected by both cavity losses and well disorder, but where the well disorder caused a homogeneous broadening. In this kind of polariton, the width of the polariton would be due to the convolution of two Lorentzians, and would be

$$2|c_{\pm}(k)|^2\gamma(k)+|d_{\pm}(k)|^2\sqrt{\psi_E}. \quad (5.4)$$

We can see that the resulting polariton width is just the weighted average of the two linewidths. Exactly at resonance both polaritons will have the same width, the average of the two linewidths of the uncoupled cavity mode and uncoupled exciton.

Now imagine a system where again the polariton was affected both by cavity losses, and well disorder, but where the

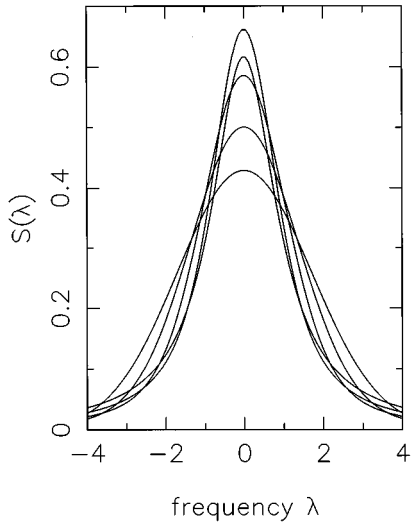

FIG. 1. The polariton line shape at five different detunings. The tall Lorentzian peak corresponds to when the polariton is cavity mode only, and the broad Gaussian to exciton only. The parameters are $\gamma=1.00$ and $\psi=1.70$, so that the disorder FWHM is twice the loss FWHM. The components vary according to the fractions defined by the dispersionless model, but the graph is still similar in character to that for a dispersive case.

cavity losses were inhomogeneous. In this kind of polariton, the width of the polariton would be due to the convolution of two Gaussians, and by adding the variances we would get a width of

$$2\sqrt{2\ln(2)}[|c_{\pm}(k)|^4\gamma(k)^2+|d_{\pm}(k)|^4\psi_E]^{1/2}. \qquad (5.5)$$

At resonance we can see that the resulting polariton width is less than the average of the two linewidths. For equal cavity and exciton widths, the polariton width would be just $1/\sqrt{2}$ of the average of the two widths.

Having done these two simple cases, we turn our attention to the more realistic case — the polariton affected by homogeneous cavity losses and by inhomogeneous well disorder. The Fourier convolution theorem gives an expression for the polariton spectral function $S_{\pm}$ as a fourier transform of

$$\widetilde{S}_{\pm}(k;\tau)=\widetilde{S}_{E\pm}(k;\tau)\widetilde{S}_{L\pm}(k;\tau)$$
$$=\frac{1}{2\pi}\exp[-|c_{\pm}(k)|\gamma(k)|\tau|-\tau^2|d_{\pm}(k)|^4\psi_E/2]. \qquad (5.6)$$

The combination of the Lorentzian cavity losses with the Gaussian disorder will give a result somewhere between the two limits of a double Lorentzian and double Gaussian discussed briefly above. Unfortunately there is no analytic expression for $S_{\pm}$, and so the analysis must be done numerically. However, it is clear that the convolution of the two different broadenings from the cavity losses and quantum well disorder will give a polariton linewidth reduced below the previously expected average value — a prediction of ''narrower than average'' polariton lines that is consistent with the observations of Fisher *et al.*[5] Figure 2 shows the

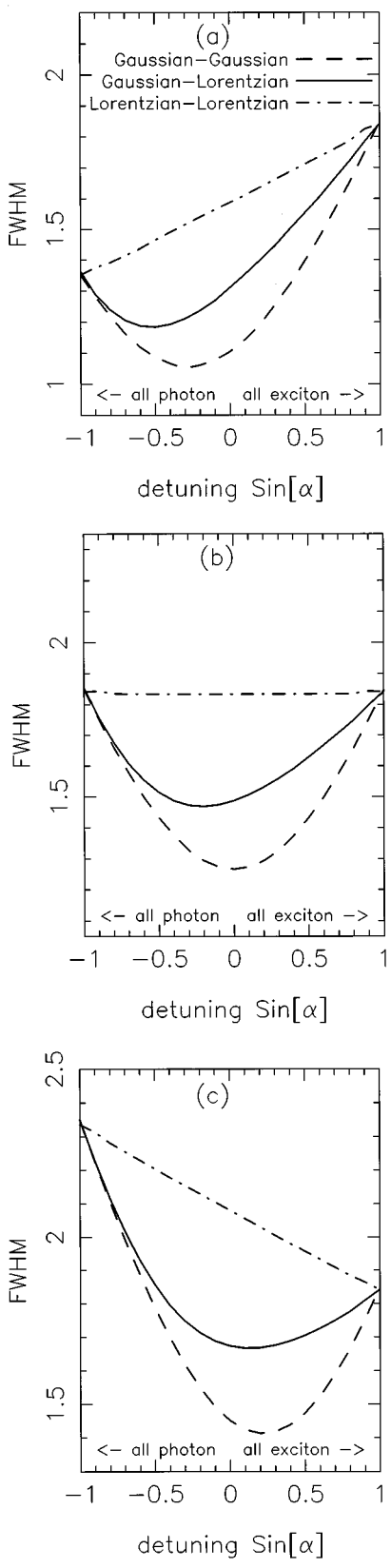

FIG. 2. FWHM for polariton spectra due to the effects of cavity loss and disorder. The double Lorentzian and double Gaussian FWHM's are also plotted for comparison to the real polariton. A range of loss and disorder strengths are covered.

three types of curve for different cavity loss and disorder strengths.

At this point it is also appropriate to make a short comment about the effect of dispersion discussed at the end of Sec. IV. The asymmetry caused by the Boltzmann tail will tend to broaden the polariton out slightly, more than would otherwise be the case. However, this effect is likely to be rather small, and it will not destroy the narrowing. The delocalization of the polariton will cause some extra narrowing not included in calculations in this paper.

Figure 2 demonstrates two important points. First, the Lorentzian cavity mode and Gaussian exciton linewidth curve does lie between the double Lorentzian and double Gaussian curves, and thus exhibits this ''narrower than average'' behavior. Also, the *minimum* linewidth of the polariton can be reduced below that of either of its two components. This is not very marked for widely differing exciton and cavity widths, but there is a more than 15% reduction in the case of evenly matched linewidths in Fig. 2(b).

VI. CONCLUSION

This theory shows how the spectral broadening due to different interactions combines together to give the total line shape and width of the coherent superposition of cavity mode and exciton that is a polariton in a microcavity quantum well. The interactions have different spectral functions which are *convolved* together to give the resulting polariton spectral function. The two broadening interactions included were the cavity losses, which cause homogeneous broadening of the cavity mode; and disorder in the quantum well, which causes an inhomogeneous, and approximately Gaussian, broadening in the exciton. The widths of the contributions from each interaction were reduced according to the fraction of cavity mode or exciton in the polariton. The results predict narrower than average polaritons like those seen by Fisher *et al.*[5] This behavior is most noticeable when the cavity loss and the disorder are of a similar size. In fact, in this regime there can be polarition linewidth *reduction*, a more significant effect than the observed narrower than average polaritons.

Although dispersion was not included, a consideration of its likely effects showed that the delocalization (or motional narrowing) it causes will enhance the narrowing effect and not degrade it. The convolution mechanism still occurs in a model that includes dispersion, and such a theory would just provide a more accurate spectral function from the disorder in the quantum well. To summarize, one of the causes of polariton linewidth narrowing is explained by the model presented here. The next step is to attempt a calculation that can combine both this and the dispersion into a single consistent calculation.

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