Motional Narrowing in Semiconductor Microcavities

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(Received 20 February 1996; revised manuscript received 10 June 1996)

The linewidths of exciton features in spectra from semiconductor quantum wells normally correspond directly to the width of the probability distribution of the disorder potential causing the broadening. This is because the mass of the exciton is large, so its motion in the potential is essentially classical. We show that in a microcavity, where polariton mixing leads to much smaller masses, quantum mechanical effects are expected to cause significant spatial averaging over the disorder potential and hence motional narrowing of the spectral lines. Our prediction is verified by experimental measurements of linewidths near resonance in a high quality microcavity structure. [S0031-9007(96)01794-2]

PACS numbers: 71.36.+c, 71.35.Cc, 72.15.Rn

The term "motional narrowing" is applied to many situations in which the width of a spectral line in a disordered system is reduced by some averaging process. An important example arises from transitions involving a quantum particle moving in a disorder potential. A classical particle can be fully localized, giving a transition line shape which simply reflects the probability distribution of the potential. By contrast, the states of a quantum particle must have a finite extent, with the consequence that spatial averaging over the disorder potential causes a reduction in the linewidth [1]. Analogous processes occur in many areas of physics, particularly NMR and ESR [2], and molecular transitions in high pressure gases [3].

In this Letter we present the first experimental demonstration of motional narrowing due to the center of mass motion of excitons in a semiconductor. Though narrowing has been predicted theoretically [4], the large exciton mass, $M_e \sim m_0$, in a typical semiconductor means that the magnitude is small. However, in microcavity structures such as the one we describe, polariton effects are important and the exciton-polariton has a much smaller effective mass, $M_p \sim 10^{-5} m_0$. As a result, the polariton is much less localized by the disorder, and significant motional narrowing occurs. Most importantly, by tuning through the cavity resonance, we are able to vary the mass and see corresponding changes in linewidth.

Before discussing polaritons in a microcavity, we describe the treatment of the effects of disorder on a normal quantum well exciton. There are numerous microscopic mechanisms which can cause disorder in a quantum well—interface roughness, alloy fluctuations, etc. Whatever the mechanism, the disorder introduces a potential, V_{dis}, which depends on the electron and hole coordinates. Provided the disorder is weak compared to the exciton binding energy, an adiabatic approximation can be used to decouple the exciton internal and center of mass coordinates. The center of mass then moves like a particle of mass M_e in an effective potential $V_e(R) = \langle e(R)|V_{\mathrm{dis}}|e(R)\rangle$, where $|e(R)\rangle$ represents an exciton with the center of mass coordinate R. This decoupling in itself causes some spectral narrowing, since the effective potential is an average of V_{dis} over length scales typically of the order of the exciton Bohr radius, $a_B \sim 100$ Å [5].

There are two important length scales for a particle moving in a disordered potential: One is λ_c, the correlation length of the potential, the other is λ_{qm}, the scale associated with the quantum mechanical state of the particle. λ_c depends on the properties of the microscopic disorder in a particular sample, but, because of the averaging in the potential definition, it has a lower bound of the same order as a_B, a bound which is typically reached. We estimate λ_{qm} by noting that the optically active exciton states have energies of order Γ, the observed linewidth. The corresponding length scale is then $\lambda_{\mathrm{qm}} \sim \hbar/\sqrt{2M_e\Gamma}$. If we take reasonable values for GaAs of $M_e = 0.25 m_0$, $\Gamma = 5$ meV, we find $\lambda_{\mathrm{qm}} \sim 50$ Å. Hence, for a normal quantum well exciton, λ_{qm} is of the same order as λ_c, and quantum mechanical effects are not very important. There is little motional narrowing, and the exciton line has approximately the same width as the probability distribution of the effective potential [4].

Having established that there is little motional narrowing in normal quantum wells, we now discuss the behavior of polaritons in semiconductor microcavities. Our microcavity is a Fabry-Pérot structure consisting of a GaAs cavity contained between two GaAs/AlGaAs Bragg reflecting mirrors. The cavity supports confined electromagnetic modes at energies for which its width is an integer multiple of half the wavelength. In our structure, the width of the cavity is chosen such that the second order confined mode is resonant with the excitonic transition in three InGaAs quantum wells located near its center. More details of the structure and the growth procedure are given in Ref. [6].

Polaritons are the mixed eigenmodes which are formed when the exciton and cavity states couple together close

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to resonance. Each polariton mode, $|p\rangle$, is a linear combination of an exciton $|e\rangle$ and a cavity photon $|l\rangle$: $|p\rangle = c_e|e\rangle + c_l|l\rangle$. Exactly on the resonance, both polaritons consist of equal mixtures of the exciton and photon states, so $c_e = \pm c_l = 1/\sqrt{2}$. The symmetric and antisymmetric modes are then split by a finite energy, known as the vacuum Rabi splitting [7]. When the system is detuned from resonance, the mixture is not equal, with one mode predominantly exciton in character ($c_e \to 1$, $c_l \to 0$), the other predominantly photon ($c_e \to 0$, $c_l \to 1$).

This behavior of the polariton modes is demonstrated experimentally in Fig. 1 which shows normal incidence reflectivity spectra from our microcavity structure at a magnetic field of 14 T. Only σ^+ circularly polarized spectra are shown in the figure; the σ^- results are similar, but the energies are shifted by the spin splitting of the exciton [6]. The band gap of the well material decreases with increasing temperature, so raising the temperature moves the exciton down through resonance with the photon. The resonance occurs at 85 K, where the two features have equal strength and are separated by a vacuum Rabi splitting of 7.3 meV. The strength of each of the reflectivity features is determined by the photon fraction of the corresponding polariton, $|c_l|^2$, so the exciton-like polariton mode is only strong close to resonance, where it mixes significantly with the photon.

It should be noted that the motional narrowing we observe does not intrinsically require a magnetic field—the main purpose of the field in our experiment is to shift the exciton energy so that it comes on resonance at a suitable temperature. We observe similar narrowing at zero magnetic field and fixed temperature by using an electric field to tune the exciton through resonance.

The cavity only confines the photon in the direction perpendicular to the mirrors. There is no confinement in the parallel plane, so the cavity photon is free to move in-plane. The perpendicular component of the photon wave vector is fixed by the mirrors to be $2\pi/L$, where L is the cavity width. Hence the energy of a photon with in-plane wave vector k is

$$E_l(k) = \frac{\hbar c}{n}\sqrt{\left(\frac{2\pi}{L}\right)^2 + k^2} \approx \frac{\hbar c}{n}\,\frac{2\pi}{L} + \frac{1}{2}\,\frac{\hbar c}{n}\,\frac{L}{2\pi}\,k^2,$$

with n the (average) cavity refractive index. Since, for small k, the in-plane dispersion takes this parabolic form, we can describe it using an effective mass, $M_l = nh/(Lc) \sim 10^{-5}m_0$.

The in-plane dispersion of the polariton depends on the dispersion of the photon and the extent of its mixing with the exciton. Polariton dispersions have been studied in detail by Houdré *et al.* using angle dependent measurements [8]. Their results agree well with a simple two-state treatment in which the form of the dispersion depends on the strength of the coupling and the extent of the detuning. Such a treatment is also applicable to experiments at high magnetic fields [9], since the large cyclotron energy means that each Landau level can be considered to interact independently with the photon. Figure 2 shows such theoretical dispersions (solid lines) for the two polariton branches when resonance occurs at $k = 0$. For sufficiently small k, the dispersions are parabolic, with an effective mass $M_p = (|c_e|^2/M_e + |c_l|^2/M_l)^{-1}$, as indicated by the dotted lines in the figure. The dependence

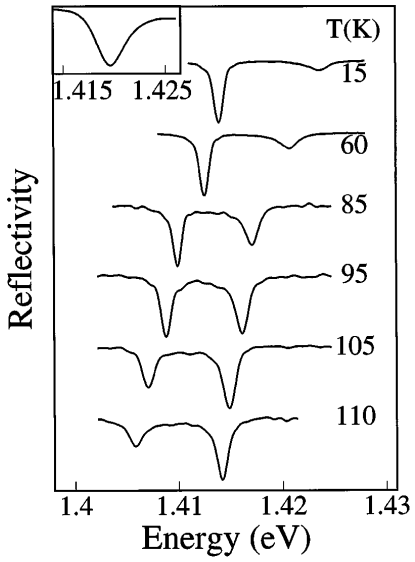

FIG. 1. Microcavity reflectivity spectra (σ^+ polarization) at a magnetic field of 14 T for a range of temperatures, demonstrating the anticrossing which occurs as the exciton is tuned through the cavity peak. The inset shows the bare exciton with the top mirror of the cavity removed, at a temperature of 22 K.

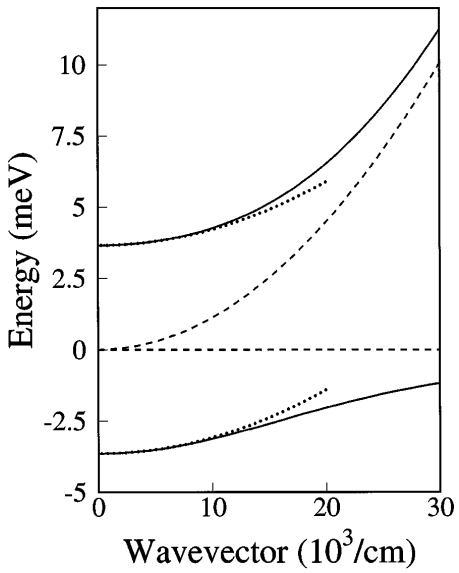

FIG. 2. Theoretical polariton dispersion (solid lines) for an on-resonance microcavity. The dashed lines indicate the uncoupled exciton (lower) and cavity photon (upper) dispersions. The dotted lines show the parabolic approximation, valid for small wave vectors.

of M_p on the coefficients c_e, c_l means that the polariton effective mass varies as the system is tuned through the resonance. Since M_l is very small compared to M_e [10], we can neglect $|c_e|^2/M_e$ and take $M_p \approx M_l/|c_l|^2 \sim 10^{-5} m_0$. It is this small value of M_p which makes motional narrowing important in microcavity structures.

The effect of disorder on the polariton is more complicated than for the exciton, since scattering can occur both between the two polariton branches and to regions of k space where the k dependence of the coefficients c_e, c_l and the nonparabolicity become important. However, if the energy scale of the disorder potential is sufficiently small, each branch can be treated independently, and only small k behavior needs to be considered. This requires that the linewidths must be small compared with the splitting between the modes, a condition which is well fulfilled in our structure, where the Rabi splitting is 7.3 meV and the linewidths are $\sim$1 meV. In this approximation, the polariton center of mass acts like a simple particle with mass M_p moving in a disorder potential $V_p(R) = \langle p(R)|V_{\rm dis}|p(R)\rangle = |c_e|^2 V_e(R)$. This has the same spatial dependence as $V_e(R)$, but the amplitude is reduced by a factor $|c_e|^2$, since only the exciton component of the polariton feels the disorder. Taking $M_p = M_l/|c_l|^2$, as described above, the center of mass Hamiltonian for the polariton is

$$H_{\rm eff} = -|c_l|^2 \frac{\hbar^2 \nabla^2}{2M_l} + |c_e|^2 V_e(R) .$$

In the polariton case, the comparison of length scales leads to a different conclusion than for the exciton: λ_c is unchanged at $\sim$100 Å, but, on resonance, $\lambda_{\rm qm} = \hbar|c_l|/\sqrt{2M_l\Gamma} \sim 10^4$ Å, using our linewidth of $\Gamma \approx 1$ meV. This comparison shows that the classical approximation, which is good for the exciton, is totally inappropriate for the polariton. Since the disorder correlation length is much smaller than $\lambda_{\rm qm}$, it is more realistic to approximate the potential by white Gaussian noise [1]. This has zero correlation length scale and is characterized solely by a correlation function of the form $\langle V_e(R_1) V_e(R_2)\rangle = \alpha\delta(R_1 - R_2)$. Though it is not possible to obtain analytical results for the broadening, the lack of a length scale in the white noise potential means that we can obtain a simple expression for the variation of the linewidth as c_e, c_l change on passing through the resonance. If we choose a length scale $R_0 = |c_l|^2/|c_e|^2$, an energy scale $E_0 = |c_e|^4/|c_l|^2$, and define variables $R' = R/R_0$, $E' = E/E_0$, the Hamiltonian transforms to

$$H'_{\rm eff} = -\frac{\hbar^2\nabla^2}{2M_l} + \frac{|c_l|^2}{|c_e|^2} V_e\left(\frac{|c_l|^2}{|c_e|^2} R'\right) = -\frac{\hbar^2\nabla^2}{2M_l} + V'_e(R') .$$

The important property of this transformation is that the correlation function for $V'_e(R')$ is just the same as for the original $V_e(R)$. Hence the transformation produces a Hamiltonian with identical properties, regardless of the values of c_e, c_l. The scaled spectra will therefore always have the same width, Γ_e, giving a real linewidth of $E_0\Gamma_e = |c_e|^4/|c_l|^2\,\Gamma_e$. The singular behavior of this function as $|c_l|^2 \to 0$ is clearly unphysical—it arises where the white Gaussian noise approximation is no longer applicable because the polariton mass becomes large. The linewidth must, in reality, tend to the bare exciton width, but, in the limit $|c_l|^2 \to 0$, the exciton-like mode becomes unobservable, so this width is never measured. We have, however, obtained the linewidth of the bare exciton by etching off the top mirror of one of our samples (inset to Fig. 1). The value of 3.1 meV is, as the theory predicts, considerably larger than any of the measurements with the mirror in place.

In order to compare the theoretical expression with experiment, it is necessary to account for the contribution to the linewidths of the broadening caused by the finite finesse of the microcavity. The cavity photon can escape through the mirrors, so it has a finite lifetime, with an associated homogeneous broadening Γ_l. The polariton spends only a fraction $|c_l|^2$ of its time as a photon, so it is less likely to escape, and the lifetime contribution to its width is reduced to $|c_l|^2\Gamma_l$. To obtain the total linewidth, the lifetime and disorder contributions are combined by convolving the line shapes for the two processes [11].

Figure 3 shows the experimental linewidths (FWHM) of the spectra in Fig. 1 plotted as a function of $|c_e|^2$, the exciton fraction in the polariton, along with our theoretical fit. The values of $|c_e|^2$ as a function of temperature were obtained from the experimental data by fitting the separation of the polariton modes to the two-state model discussed above. Plotted in this way, our theory predicts that the experimental points for both polariton branches should fall on a single curve. The figure also shows, as triangles, the widths measured using electric field tuning at zero magnetic field and a constant temperature of 6 K. The close correspondence between the electric field and temperature tuning results allows us to rule out the possibility of the linewidth variations being due to any thermal broadening mechanism [12] or magnetic field effects.

The most significant feature of the experimental data in Fig. 3 is the decrease in the linewidth near resonance ($|c_e|^2 = 0.5$), which occurs for the lower branch (filled symbols). This behavior cannot be explained by simply averaging the exciton and photon linewidths, as would be valid if both were homogeneously broadened [13]. It also cannot be explained by using a model which does not include motional narrowing but which takes into account the convolution effects discussed above (dashed line). By contrast, the motional narrowing model (solid line) gives a very good fit [14] to the data, particularly for the lower branch. The comparison for the upper polariton branch (open symbols) is less satisfactory, with the experimental points lying above the predicted values. The difference between the widths of the two branches is not presently

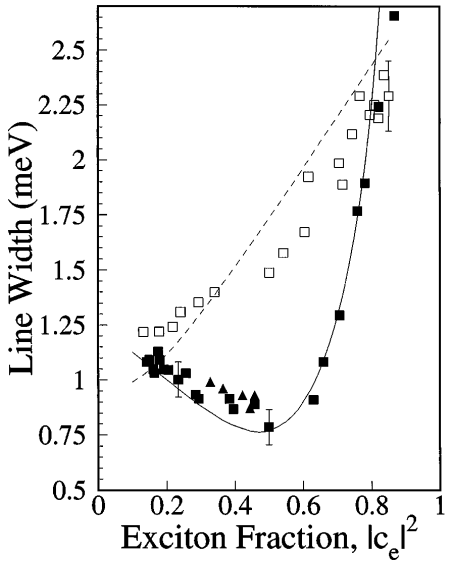

FIG. 3. The linewidths (squares) of the spectra of Fig. 1 plotted as a function of the exciton fraction in the polariton, $|c_e|^2$. The filled symbols are the experimental points for the lower branch and the open symbols are those for the upper branch. Also shown (triangles) are the corresponding results for electric field tuning. The solid line is the theoretical fit with our motional narrowing model, while the dashed line shows the "fit" obtained without motional narrowing.

understood—additional broadening for the upper branch has previously been ascribed to effects of absorption by exciton continuum states [9], but this cannot apply at high magnetic fields, where the nearest optically allowed transitions are at much higher energies. We speculate that the difference may be due to coupling with higher energy optically forbidden states, mixed into the wave function by the disorder potential.

Recently, Savona *et al.* [15] have reported numerical simulations of inhomogeneous broadening in a one (in-plane) dimensional microcavity model which treats disorder scattering and polariton coupling on the same footing. Their results show that, for disorder strengths comparable to ours, the scaling theory works very well. Furthermore, they find that the upper branch is broader than the lower branch, suggesting that disorder scattering can indeed explain the difference between the widths of the two branches. Although a realistic two dimensional simulation has yet to be carried out, these results provide strong support for our simpler scaling treatment.

In conclusion, we have shown theoretically that motional narrowing is to be expected in high quality semiconductor microcavities, and we have verified experimentally that it does indeed occur. This is the first experimental demonstration of motional narrowing of interband optical transitions in semiconductors. It is made possible by the control provided by a microcavity of the mixing between photon and exciton states, and hence the dispersion of the polariton.

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[1] B. I. Halperin, Phys. Rev. **139**, A104 (1965).
[2] See, for example, A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961), p. 446.
[3] R. Ouillon, Chem. Phys. Lett. **35**, 63 (1975); P. W. Anderson, Phys. Rev. **76**, 647 (1949).
[4] R. F. Schnabel, R. Zimmermann, D. Bimberg, H. Nickel, R. Losch, and W. Schlapp, Phys. Rev. B **46**, 9873 (1992); R. Zimmermann and E. Runge, J. Lumin. **60-1**, 320 (1994).
[5] J. Singh and K. K. Bajaj, Appl. Phys. Lett. **48**, 1077 (1986). But see also S. D. Baranovskii, U. Doerr, P. Thomas, A. Naumov, and W. Gebhardt [Phys. Rev. B **48**, 17 149 (1993)] for an argument that the relevant length scale may be shorter.
[6] T. A. Fisher, A. M. Afshar, M. S. Skolnick, D. M. Whittaker, and J. S. Roberts, Phys. Rev. B **53**, 10 469 (1996).
[7] C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. **69**, 3314 (1992).
[8] R. Houdré, C. Weisbuch, R. P. Stanley, U. Oesterle, P. Pellandini, and M. Ilegems, Phys. Rev. Lett. **73**, 2043 (1994).
[9] J. Tignon, P. Voisin, C. Delalande, M. Voos, R. Houdré, U. Oesterle, and R. P. Stanley, Phys. Rev. Lett. **74**, 3967 (1995).
[10] In a magnetic field, M_e is infinite, but even without a field $M_e \sim 0.25m_0$, which is much larger than M_l.
[11] P. Kinsler and D. M. Whittaker (to be published); see also S. Pau, G. Björk, J. Jacobson, H. Cao, and Y. Yamamoto, Phys. Rev. B **51**, 14 437 (1995).
[12] Indeed, the thermal broadening expected at 120 K is only 0.15 meV; see D. S. Chemla, D. A. B. Miller, P. W. Smith, A. C. Gossard, and W. Wiegmann, IEEE J. Quantum Electron. **20**, 265 (1984).
[13] Y. Zhu, D. J. Gauthier, S. E. Morin, Q. Wu, M. J. Carmichael, and T. W. Mossberg, Phys. Rev. Lett. **64**, 2499 (1990).
[14] The theoretical widths were obtained by convolving a homogeneous Lorentzian contribution of width $\Gamma_l|c_l|^2$ with an inhomogeneous Gaussian contribution of width $\Gamma_e|c_e|^4/|c_l|^2$, the constants Γ_l and Γ_e being determined by fitting the experimental widths at low and high temperatures.
[15] V. Savona, F. Tassone, C. Piermarocchi, A. Quattropani, and P. Schwendimann, Proceedings of the 23rd International Conference on Physics of Semiconductors, Berlin, 1996 (to be published).