Effect of inhomogeneous broadening on optical properties of excitons in quantum wells

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The effect of inhomogeneous broadening on exciton-light coupling in quantum wells (QW's) is studied by a semiclassical model, in which the exciton resonance frequency is assumed to have a Gaussian distribution. The presence of disorder may be observed in absorption, but not in reflectivity spectra. The integrated absorption of light in single and multiple QW's is nonzero only in the presence of a finite damping and increases with enhancement of either homogeneous or inhomogeneous broadening: only when broadening is larger than the exciton radiative width does the integrated absorption attain the saturation value expected from the exciton oscillator strength and the number of QW's. This behavior is similar to that of exciton-polariton absorption in bulk semiconductors, but unlike the bulk case it is not due to spatial dispersion. For multiple QW's, QW's in a microcavity and in a thick film of bulk, in general, disorder produces a decrease of the period of oscillations in the time-resolved transmission and yields a faster decay of the signal. However, inhomogeneous broadening is also found to lead to beatings in the time-resolved transmission or reflection of light from a *single* QW: the oscillations, which originate from interference between upper and lower wings of the exciton distribution, may be observed in high-quality samples. [S0163-1829(98)06507-2]

I. INTRODUCTION

The effect of disorder on optical properties of quantum well (QW) excitons is an important question that is currently of great interest.[1,2] The microscopic origin of disorder is most commonly interface roughness or alloying. Disorder manifests itself in both cw and time-resolved experiments. Typical phenomena related to disorder include the Stokes shift and inhomogeneous broadening of exciton lines, mixed Lorentzian-Gaussian line shapes,[3] modification of exciton radiative lifetime,[4,5] change of beat period in linear and nonlinear pulse propagation,[6–10] motional narrowing, and modification of Rabi splitting for QW excitons in microcavities.[11–13] Disorder is also responsible for momentum broadening, giving rise to resonant Rayleigh scattering,[14–16] finite risetime in the time-resolved behavior of secondary radiation (Rayleigh scattering and luminescence),[17–21] etc. While the first class of phenomena can be viewed as effects of *dephasing*, or partial breakdown of temporal coherence, the second kind of phenomena imply *scattering* and partial breakdown of spatial coherence.

In this work we study the effect of disorder on linear optical properties of excitons in QW's by means of a simple semiclassical model for inhomogeneous broadening. The model assumes conservation of the in-plane wave vector and a Gaussian distribution of exciton energies, which is taken into account in the dielectric susceptibility of the structure. This approach may be related to the microscopic model of in-plane potential fluctuations with a scale that is much smaller than the wavelength of light. Our purpose is threefold: (i) to give representative results for the effects of inhomogeneous broadening on both cw and time-resolved experiments in several situations like single and multiple QW's, thin layers, microcavities; (ii) to study in detail the effect of disorder in a paradigmatic system, namely, the single quantum well (SQW), particularly for time-resolved experiments. We obtain in fact a new and unexpected result, namely, that a *continuous* distribution of excitonic frequencies may give rise to beatings in the time-resolved transmission; (iii) to address the question of integrated absorption in single and multiple QW's as a function of both homogeneous and inhomogeneous broadening.

The problem of integrated absorption in multiple quantum wells (MQW's) is related to the issue of exciton-polariton absorption in bulk semiconductors. The mixed modes of the exciton and photon fields, i.e., the polaritons,[22,23] are stationary states in bulk crystals unless dissipative processes are introduced. It was found experimentally that the integrated excitonic absorption increases as a function of temperature up to a critical temperature T^*, reflecting the increase of damping, and only for $T > T^*$ does it attain the value expected from the exciton oscillator strength;[24–27] this behavior is explained theoretically when spatial dispersion is taken into account[28–30] (see Sec. IV for a fuller discussion). Similar experiments performed on MQW's seemed to give the same result, namely, that the integrated absorption increases with temperature;[27] however, the interpretation was not clear since spatial dispersion of the exciton in uncoupled MQW's can only be present for a finite angle of propagation with respect to the growth direction. In this work we reexamine the question of integrated absorption in MQW's, and show that different interpretations follow from different definitions of ''absorption'' (either dimensionless, or with the dimensions of an inverse length) which may be given in quasi-2D systems.

Since the present model assumes conservation of the in-

plane wave vector, it cannot account for phenomena related to the breakdown of spatial coherence, like resonant Rayleigh scattering. Also, applications are presented for the simplest physical situations: we neglect exciton excited states and the electron-hole continuum, as well as the effects related to the presence of a cladding layer.[31] The latter are of course important in real structures, particularly for a precise determination of reflectivity line shapes, but are unrelated to the problem of inhomogeneous broadening, which is the main issue here.

It must be emphasized that disorder is a static perturbation, which by itself induces no dephasing since all scattering processes are elastic. Disorder produces a (partial) exciton localization and results in an inhomogeneous distribution of exciton energies; an incident pulse excites all oscillators in phase, and each level has a coherent time evolution. However the different phases of excitons with different energies give rise to interference, which may be called disorder-induced dephasing.[17] The interference of different energy levels in the emitted radiation is responsible for most features in the time-resolved response presented in this paper. Scattering terms changing the in-plane wave vector (which are neglected here) remove excitons emitting in the phase-matching directions and produce emission in all other directions, thus yielding resonant Rayleigh and luminescence signals with a finite rise time. The emission in the transmission/reflection directions is thus the spatially coherent signal from excitons which do not undergo scattering events. Corrections to the present model therefore depend on the total amount of scattered light, and of its time evolution. This point will be discussed again in Secs. II and V.

Previous investigations of the effects of disorder will now be briefly discussed. Microscopic theories for momentum scattering of QW excitons due to interface roughness have been developed[17,32,33,12] in which the exciton center-of-mass motion problem in a disordered potential is solved by numerical integration. Semiclassical models for the effect of inhomogeneous broadening on excitons in MQW's are described in Refs. 34, 35 and 31: in these papers, however, only *vertical* disorder is considered, i.e., due to the different exciton energies in different wells. The present model, on the other hand, accounts for *lateral* disorder due to interface roughness and applies also to a single QW. The difference between lateral and vertical disorder in the optical response of the structure holds even in the case of wave vector conservation and can be formulated as follows: lateral disorder influences the dielectric susceptibility of each quantum well while vertical disorder just shifts the exciton resonance energy in different wells. A Green's-function approach for describing the effect of a disorder potential on exciton-light coupling is presented in Refs. 7 and 8. These works are closely related to the results presented here. Several papers address the question of microcavity-embedded QW excitons in the presence of disorder.[36–40,10,12,41] A more detailed comparison with existing approaches and results will be given in the course of the paper.

The rest of this paper is organized as follows. In Sec. II we describe the model for inhomogeneous broadening. In Sec. III results for cw optical properties are presented. Section IV is devoted to the problem of integrated absorption in single and multiple QW's. In Sec. V we discuss time-resolved transmission and reflection in single and multiple QW's, thin films, and microcavities. Section VI contains concluding remarks.

II. MODEL

In the framework of linear nonlocal response theory,[42] Maxwell equations for a light wave normally incident on a single quantum well in the vicinity of the exciton resonance frequency can be written in the form

$$\nabla\times\nabla\times\mathbf{E}=\frac{\omega^2}{c^2}\mathbf{D}, \tag{1}$$

where

$$\mathbf{D}(z)=\epsilon_\infty\mathbf{E}(z)+4\pi\mathbf{P}_{\rm exc}(z), \tag{2}$$

$$\mathbf{P}_{\rm exc}(z)=\int\chi(z,z')\mathbf{E}(z')dz', \tag{3}$$

and the nonlocal susceptibility is expressed as

$$\chi(\omega,z,z')=\chi(\omega)\Phi(z)\Phi(z'), \tag{4}$$

where $\Phi(z)$ is the exciton envelope function taken with equal electron and hole coordinates, and z is the normal to QW plane direction. Taking into consideration only the ground exciton state, one can write, in the absence of inhomogeneous broadening,

$$\chi(\omega-\omega_0)=\frac{\epsilon_\infty\omega_{\rm LT}\pi a_B^3\omega_0^2/c^2}{\omega_0-\omega-i\gamma}, \tag{5}$$

where ω_0 is the exciton resonance frequency, γ is its nonradiative homogeneous broadening, $\omega_{\rm LT}$ is the exciton longitudinal-transverse splitting in the bulk material, and a_B is the bulk exciton Bohr radius.

Solving Eq. (1) using the susceptibility (5) and supposing for simplicity the background dielectric constants in the well and barrier to be equal, one can obtain the amplitude reflection and transmission coefficients in the form[43,44]

$$r=\frac{i\Gamma_0}{\tilde{\omega}_0-\omega-i(\gamma+\Gamma_0)},\quad t=1+r, \tag{6}$$

where the renormalized frequency $\tilde{\omega}_0$ differs from ω_0 by a radiative shift

$$\tilde{\omega}_0-\omega_0=\frac{k\omega_{\rm LT}\pi a_B^3}{2}\int\int dz\,dz'\sin k|z-z'|\Phi(z)\Phi(z'), \tag{7}$$

the optical wave vector $k=\sqrt{\epsilon_\infty}\omega/c$, and the radiative decay rate Γ_0 of the exciton wave function is given by

$$\Gamma_0=\frac{k\omega_{\rm LT}\pi a_B^3}{2}\left[\int\Phi(z)\cos kz\,dz\right]^2=\frac{\pi}{\sqrt{\epsilon_\infty}}\frac{e^2}{mc}\frac{f_{xy}}{S} \tag{8}$$

in terms of the oscillator strength per unit area f_{xy}/S. The exciton radiative lifetime is $1/(2\Gamma_0)$. Γ_0 is typically a fraction of a meV (0.02–0.05 meV for GaAs/Al_xGa_{1-x}As or In_xGa_{1-x}As/GaAs QW's).

We suppose that due to inhomogeneous broadening the exciton resonance frequency is described by a Gaussian distribution function and neglect possible dependence of the exciton envelope function on energy. The validity of the latter assumption has been confirmed within a microscopic model,[17] provided the envelope function corresponding to the average QW width is used in the calculation. Thus, we substitute the resonant dielectric susceptibility (5) by a function

$$\tilde{\chi}(\omega)=\frac{1}{\sqrt{\pi}\Delta}\int d\nu\,\chi(\omega-\nu)\exp\left[-\left(\frac{\nu-\omega_0}{\Delta}\right)^2\right]$$

$$=\frac{i\epsilon_\infty\omega_{\mathrm{LT}}\pi^{3/2}\omega_0^2a_B^3}{c^2\Delta}w(z), \tag{9}$$

where

$$z=\frac{\omega-\omega_0+i\gamma}{\Delta} \tag{10}$$

and

$$w(z)=e^{-z^2}\mathrm{erfc}(-iz), \tag{11}$$

$\mathrm{erfc}(z)$ being the complementary error function.[45] Here we have supposed that $\Delta,\gamma>0$. Solving Eq. (1) with the susceptibility (9) one can obtain, instead of Eq. (6),

$$r=-\frac{\sqrt{\pi}\Gamma_0 w(z)}{\Delta+\sqrt{\pi}(\Gamma_0+i(\tilde{\omega}_0-\omega_0))w(z)},\quad t=1+r. \tag{12}$$

In the following we will neglect the exciton radiative shift, which is usually $\ll\Gamma_0$, and therefore assume $\tilde{\omega}_0-\omega_0\approx 0$. The form (12) of the reflection coefficient is convenient, since the w function can be evaluated by series expansions.[45]

In case of small inhomogeneous broadening one can use the asymptotic expression

$$w(z)\to\frac{i}{\sqrt{\pi}z},\quad |z|\to\infty, \tag{13}$$

and Eq. (12) reduces to Eq. (6).

We would like to stress at this point that for any form of the susceptibility $\chi(\omega-\omega_0)$ the reflection coefficient r can be represented as

$$r=\frac{i\alpha\chi}{1-i\alpha\chi}, \tag{14}$$

where $\alpha=\Gamma_0c^2/(\varepsilon_\infty\omega_{\mathrm{LT}}\pi\omega_0^2a_B^3)$. This follows from Maxwell equations (1)–(3) with nonlocal dielectric susceptibility (4), and corresponds to a full inclusion of reabsorption, or polaritonic effects beyond perturbation theory. The procedure is appropriate if the scale of in-plane potential fluctuations acting on the exciton center of mass is much smaller than the wavelength of light in the medium λ, so that all excitons give a coherent contribution to the susceptibility. Indeed in microscopic models of interface roughness due to growth islands of small linear extension the potential correlation length is of the order of the exciton radius,[17] i.e., much smaller than λ. The distribution of the exciton resonance frequency is taken into account here by averaging the susceptibility χ in Eq. (14); the present model is similar to that of Refs. 7 and 8, where a random potential with short-range correlation is assumed and the linear susceptibility is obtained by a configuration average. Both here and in Refs. 7 and 8 scattering processes that break wave-vector conservation are neglected: the effect of scattering is now discussed.

In general, an extinction coefficient can be defined, which is the sum of absorption and scattering coefficients:[46] absorption represents dissipation of energy in the system, while the scattering term accounts for light that is reemitted in directions other than those of transmission and reflection. The scattering efficiency for resonant excitation of free excitons in QW's is usually considered as small, however, systematic investigations of its value are not available in the literature. Measurements of the intensity of resonant Rayleigh scattering in QW's indicate that the ratio of total elastically scattered light to reflected light is of the order of 10^{-3} or smaller.[47] A similar value is obtained by estimating the scattering coefficient using Rayleigh's classical formula.[48,49] Thus the scattering term is indeed small, and the approximation of identifying extinction with absorption is well justified, at least for cw experiments. A discussion of the effect of scattering for time-resolved experiments is deferred to Sec. V.

Within the present approximation of neglecting scattering, absorption is defined as

$$A(\omega)=1-|r|^2-|t|^2; \tag{15}$$

absorption of a quantum well for vanishing inhomogeneous broadening is

$$A(\omega)=\frac{2\gamma\Gamma_0}{(\omega_0-\omega)^2+(\gamma+\Gamma_0)^2}, \tag{16}$$

so that the integrated absorption is

$$\int d\omega\,A(\omega)=\frac{2\pi\gamma\Gamma_0}{\gamma+\Gamma_0}. \tag{17}$$

In case of inhomogeneous broadening the integrated absorption depends on both broadenings γ and Δ.

Time-resolved reflection and transmission spectra from the single quantum well have the same shape because the corresponding amplitude coefficients are related by $t=1+r$. For example, the time-resolved transmission in case of incident δ pulse is described by a function

$$G(\tau)=\int\frac{d\omega}{2\pi}\exp(-i\omega\tau)t(\omega). \tag{18}$$

The experimentally detectable quantity is the intensity of light that is proportional to $|G(\tau)|^2$. In case of no inhomogeneous broadening $G(\tau)$ can be calculated analytically:

$$G(\tau)=-\Gamma_0\exp[-(\gamma+\Gamma_0+i\omega_0)\tau]\quad(\tau>0). \tag{19}$$

In order to generalize this formalism for the case of MQW's one should derive the amplitude reflection and transmission coefficients of MQW in terms of the single QW reflection and transmission coefficients (12). This can be done by using a transfer matrix method. At normal incidence, in the basis

of amplitudes of incoming and outgoing light waves the transfer matrix across the period of the MQW structure has a form

$$\hat{M}=\frac{1}{t}\begin{bmatrix}(t^2-r^2)e^{ikd} & re^{-ikd}\\ -re^{ikd} & e^{-ikd}\end{bmatrix}, \tag{20}$$

where d is the period of the structure. The eigenvalues of this matrix can be written in form

$$e^{\pm iQd}=\frac{m_{11}+m_{22}}{2}\pm\sqrt{\frac{(m_{11}+m_{22})^2}{4}-1}, \tag{21}$$

where m_{11} and m_{22} are the diagonal elements of the matrix $\hat{M}$. The eigenvectors of this matrix can be represented as

$$I_{\pm}=\begin{bmatrix}1\\ a_{\pm}\end{bmatrix}, \tag{22}$$

where

$$a_{\pm}=\frac{re^{ikd}}{e^{-ikd}-te^{\pm iQd}}. \tag{23}$$

Representing the light wave inside the structure as a linear combination of two eigenvectors and using the Bloch theorem one can obtain the amplitude reflection and transmission coefficients of the MQW in the form

$$r_{\mathrm{MQW}}=\frac{a_+a_-(1-e^{2iNQd})}{a_--a_+e^{2iNQd}}; \tag{24}$$

$$t_{\mathrm{MQW}}=\frac{(a_--a_+)e^{iNQd}}{a_--a_+e^{2iNQd}}e^{-ikd}. \tag{25}$$

The eigenfrequencies of exciton polaritons in the structure[50,51] must satisfy the equation

$$a_+e^{iNQd}=a_-e^{-iNQd}. \tag{26}$$

Substituting the amplitude coefficients (24),(25) into Eqs. (15),(18) one can obtain, respectively, the absorption and time-resolved transmission spectra of the MQW.

For excitons in thick films (with a thickness much larger than the exciton Bohr radius) one can use the local response theory. The inhomogeneous broadening can be taken into account in the frequency-dependent dielectric function of the layer, which can be written in the vicinity of the exciton resonance as

$$\epsilon(\omega)=\epsilon_\infty+\frac{\tilde{\chi}(\omega)c^2}{\pi a_B^3\omega^2}, \tag{27}$$

where $\tilde{\chi}(\omega)$ is the same as in Eq. (9). In order to take into account the spatial dispersion one should replace in $\tilde{\chi}(\omega)$: $\omega_0\rightarrow\omega_0+(\hbar k^2/2M)$, where M is the exciton mass. In this case, to calculate the optical properties of the system one should introduce additional boundary conditions on the excitonic contribution to the dielectric polarization. In the following, however, we will ignore the exciton spatial dispersion and use therefore only the usual Maxwell boundary conditions.

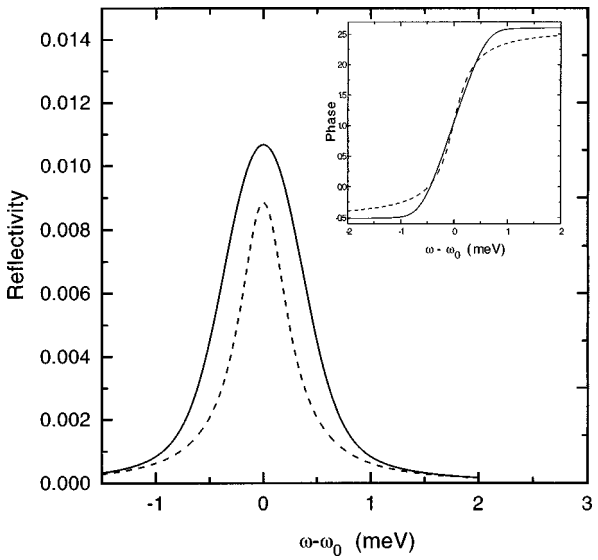

FIG. 1. Reflectivity of a single quantum well, for $\gamma=0$, $\Delta=0.4$ meV (solid line) and $\gamma=0.25$ meV, $\Delta=0$ (dashed line). Inset: phase of the reflection coefficient. In this and the following figures the index of refraction is $n=3.26$ and the radiative width is taken to be $\Gamma_0=0.026$ meV, corresponding to a $GaAs/Al_{0.4}Ga_{0.6}As$ QW of about 10 nm thickness.

III. CW SPECTRA

In Fig. 1 we show the reflectivity of a single QW for the cases of homogeneous broadening (dashed line) and inhomogeneous broadening (solid line). Note that γ represents the half-width at the half-maximum (HWHM) of the homogeneous line, whereas the HWHM of the inhomogeneous line is $\Delta\sqrt{\ln 2}$. The tails of the reflectivity curves are Lorentzians in both cases: in fact from the asymptotic behavior of the w function [Eq. (13)] one sees that the line shape (12) tends to the Lorentzian form (6) far from the line center. Thus it is difficult to distinguish between homogeneous and inhomogeneous broadening in reflectivity experiments. The phase of the reflection coefficient is shown in the inset of Fig. 1 and is seen to have a similar behavior for both sources of broadening.

Figure 2 shows the calculated absorption line shape for both homogeneous broadening (dashed lines) and inhomogeneous broadening (solid line) for the cases of one and five quantum wells. Here the inhomogeneously broadened spectrum decays like a Gaussian in the tails; thus it is possible to distinguish whether the line is mainly homogeneously or inhomogeneously broadened by performing a careful lineshape analysis of absorption measurements.[3] Figure 2 also shows that absorption obviously increases with the number of QW's, while the line shape does not change appreciably. Peculiar, asymmetric line shapes are obtained for MQW's when broadening is comparable to or smaller than the radiative width. To give an example, the inset of Fig. 2 shows the absorption line shape for five QW's with very small values of both broadenings. The asymmetry and the peak structure originate from radiative coupling between the QW's, which gives rise to a splitting of the eigenmodes.[50–53,31] However, in practice the inhomogeneous broadening grows rapidly with the number of QW's due to unavoidable differences between the well widths, so that the peculiar line shape is

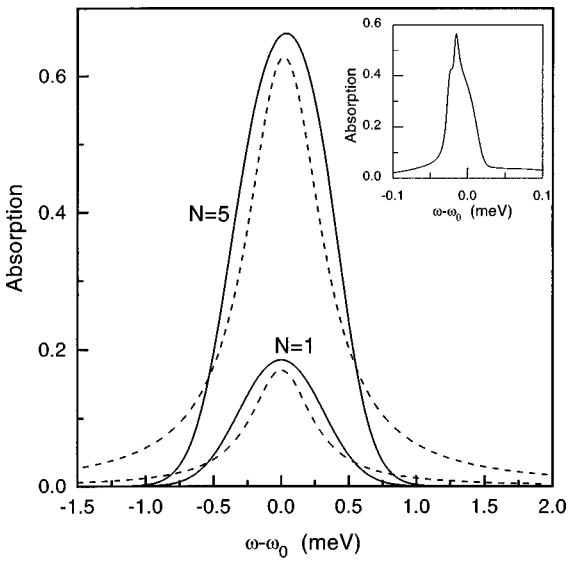

FIG. 2. Absorption of a single quantum well and a MQW with five wells, for $\gamma=0$, $\Delta=0.4$ meV (solid line) and $\gamma=0.25$ meV, $\Delta=0$ (dashed line). Inset: absorption for five QWs, with $\gamma=0$, $\Delta=0.01$ meV.

washed out and only the Lorentzian-Gaussian line shape shown in Fig. 2 remains.

Figure 3 shows the calculated reflection spectra for five Bragg arranged QW's, i.e., a MQW structure with a period $d=\lambda/2$, where λ is the wavelength of light in the media at the exciton resonance frequency. In the absence of inhomogeneous broadening the spectrum reminds typical spectra of Bragg mirrors, i.e., shows a plateau and symmetric oscillating tails. The plateau corresponds to the optical stop band and the oscillations are associated with eigenfrequencies of exciton-polariton modes in the system. In the presence of a very weak disorder ($\Delta=0.01$ meV) the spectral line shape far from the exciton resonance frequency remains almost unchanged while the plateau transforms to two well-distinct peaks. This indicates that the positive interference of all reflected light waves in the structure that is responsible for the

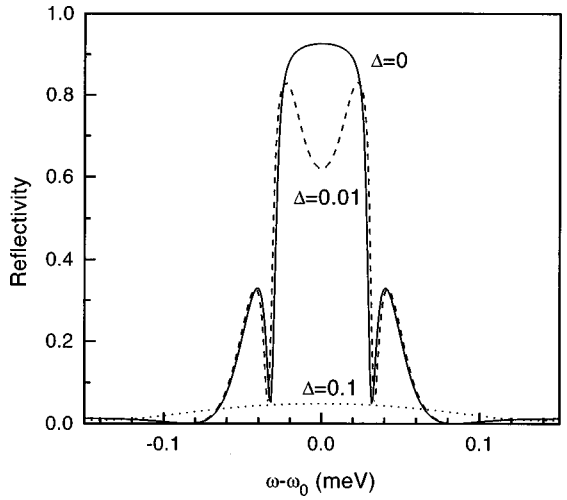

FIG. 3. Reflectivity of Bragg arranged MQW with five wells (period $d=\lambda/2$), for different values of the inhomogeneous broadening.

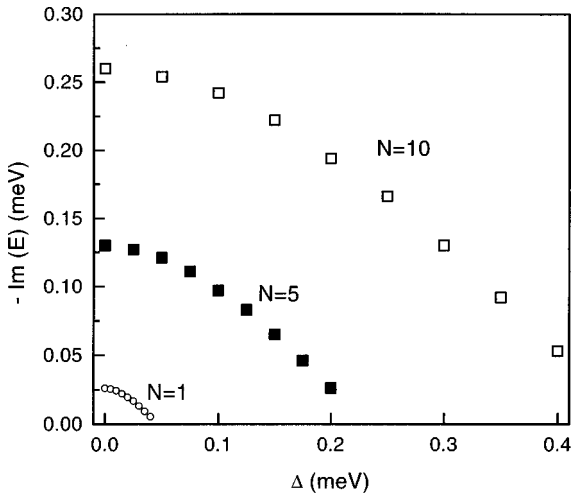

FIG. 4. Radiative width for the optically active mode of a Bragg arranged MQW with different number of wells (period $d=\lambda/2$), for different values of the inhomogeneous broadening.

stop-band appearance is lifted in case of even small disorder. At $\Delta=0.1$ meV there is no more oscillations in the spectrum and its form tends to a usual Lorentzian. This is a limit of strong disorder (in comparison with the radiative broadening) that suppresses all interference effects in the structure. This is realized in the majority of real samples.

Electromagnetic coupling between excitons in MQW's gives rise to eigenmodes with different energies, or in other words, to a radiative splitting of the exciton states. The state that matches the wave vector of light in the sample has the largest radiative width:[50,52] this can lead to a radiative lifetime that is much shorter than for a single QW. Since this ''superradiant'' radiative recombination requires phase coherence between excitons in different wells, it is obviously of interest to examine in which way disorder competes with radiative coupling. For a single QW it was already shown that disorder decreases the radiative width;[4] a model based on vertical disorder suggested that for MQW's inhomogeneous broadening has to be compared with the *superradiant* decay rate.[35] In Fig. 4 we show the decay rate (i.e., minus the imaginary part of the complex energy) for the optically active mode in Bragg-arranged MQW's. It has been found from Eq. (26) solved numerically in the complex plane. It can be seen from Fig. 4 that the superradiant width is indeed suppressed by disorder, and that the energy scale for this suppression grows with the number of QW's. Collective effects in the radiative properties of excitons in Bragg and anti-Bragg arranged MQW's have been recently observed.[54]

IV. INTEGRATED ABSORPTION

It is often taken for granted that the frequency integral of the exciton absorption line is a temperature-independent constant, which is proportional to the exciton oscillator strength. Such a statement follows from a treatment of exciton-radiation coupling within second-order perturbation theory[55,42] and therefore neglects the oscillatory exchange of energy between exciton and photon, which is characteristic of polaritonic effects. Within the exciton-polariton picture[22,23,56,42] optical absorption cannot be due to coupling of light to the exciton alone, but must follow from the pres-

ence of dissipative processes like exciton-phonon coupling. In physical terms, the incoming photon produces an excitonic transition, and is then reemitted with the same wave vector, and so forth, unless scattering events break the translational symmetry and transfer energy to the crystal lattice.

Absorption experiments performed on bulk CdS,[24] GaSe,[25] ZnSe,[26] and GaAs (Ref. 27) crystals indicated that, on decreasing the temperature T, the integrated absorption $K(T)=\int_{\text{line}}\alpha(\omega)\,d\omega$ remains constant only above a characteristic temperature T^*, then for $T<T^*$ it decreases towards smaller values. The temperature T^* marks the crossover from the excitonic ($T>T^*$) to the polaritonic ($T<T^*$) regime; only in the former case the exciton-light coupling can be treated by perturbation theory. From a theoretical point of view, it was shown[57] that if the exciton has no spatial dispersion the integrated absorption has to be constant and temperature independent: this can also be viewed as a consequence of the sum rules embodied in Kramers-Kronig relations, which, however, do not necessarily hold if the dielectric constant is wave-vector dependent. In fact, when excitonic spatial dispersion is taken into account the integrated absorption is found to depend on damping γ compared with a critical value $\gamma_c=\omega_0\sqrt{2\epsilon_\infty\hbar\omega_{\text{LT}}/(Mc^2)}$:[29,30] for $\gamma<\gamma_c$ the integrated absorption grows with γ, while for $\gamma>\gamma_c$ it is a constant. Thus the crossover from polaritonic to excitonic regimes is governed by the temperature dependence of the exciton damping γ.

Recent experiments performed on GaAs/Al_xGa_{1-x}As MQW's (Ref. 27) indicated a temperature dependence of integrated absorption that is similar to that observed in bulk GaAs, namely, an increase at low temperature followed by saturation. However, since the QW's in Ref. 27 are electronically uncoupled, exciton motion along the growth direction cannot occur, and spatial dispersion is present only for in-plane motion of the exciton. The temperature dependence of integrated absorption is attributed in Ref. 27 to the finite angle of incidence of the incoming beam; however, no definite conclusions are drawn. In another theoretical work[58] the integrated absorption of a MQW structure has been calculated taking into account the optical coupling of homogeneously broadened exciton resonances in QW's; this work predicted the increase of integrated absorption with temperature increase also for a normal incidence. An important question for this kind of analysis is which definition of absorption has to be used, i.e., whether the quantity $K(T)$ refers to the integral of the absorption coefficient (with the dimensions of an inverse length) or of the optical density $A=1-R-T$ (which is obviously dimensionless). A reexamination of the issue of integrated absorption in single and multiple QW's is clearly needed, and is the purpose of this section.

Here we concentrate on the definition $A=1-R-T$, which is directly measured in an optical experiment; this quantity will be referred to as ''absorption'' or ''optical density.'' In the case of homogeneous exciton broadening, the frequency integral $\int_{\text{line}}A(\omega)\,d\omega$ is seen to depend on exciton damping [Eq. (17)]: for $\gamma\ll\Gamma_0$ the integrated absorption is proportional to γ, while for $\gamma\gg\Gamma_0$ it is a constant and is proportional to the oscillator strength per unit area. This result admits a simple physical interpretation in terms of wave-vector conservation. An incoming photon has a finite probability of being absorbed by the exciton; however, since the in-plane wave vector is conserved the photon is reemitted in either the transmission or the reflection directions, and in both cases no energy is absorbed (i.e., dissipated) in the crystal. Only if the exciton suffers scattering events (embodied in the damping γ) before decaying radiatively is the incoming photon removed from the transmitted/reflected beams. Thus the total energy dissipated in the crystal increases with the rate of damping processes compared to radiative recombination.

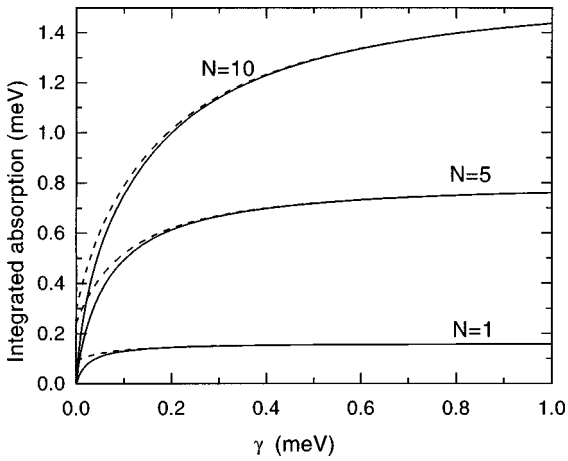

FIG. 5. Integrated absorption (see text) for MQW's with a period $d=20$ nm, for different number of wells, as a function of homogeneous broadening. Solid lines: $\Delta=0$. Dashed lines: $\Delta=0.1$ meV. Here again $\Gamma_0=0.026$ meV and $n=3.26$.

In Fig. 5 we show the integrated absorption for MQW's as a function of damping γ, for homogeneous broadening only (solid lines) and for a finite value of the inhomogeneous broadening Δ (dashed lines). In all cases the integrated absorption increases with the damping; however, when $\Delta=0$ (homogeneously broadened case) the integrated absorption vanishes for $\gamma\to0$, while if $\Delta\neq0$ it tends to a constant value for $\gamma\to0$ (thus when $\Delta\neq0$ the integrated absorption has a discontinuity at $\gamma=0$, since it must vanish there). For large damping the integrated absorption saturates to a constant value, which reflects the total oscillator strength of the exciton in the MQW, and equals $2\pi N\Gamma_0$. The critical value of γ, for which the integrated absorption starts to saturate, also increases with the number of wells reflecting the increase of the radiative width;[50,51] this is quite important for an experimental observation, since the critical value of γ for one QW is of the order of Γ_0 and is too small for the increase of integrated absorption to be observed. The behavior shown in Fig. 5 indicates that an increase of integrated absorption as a function of temperature is expected.

Figure 6 shows again the integrated absorption for MQW's, but now as a function of inhomogeneous broadening Δ. The solid lines refer to the case of pure inhomogeneous broadening. Of course when $\gamma=0$ exactly, optical absorption is zero for all frequencies: the solid curves in Fig. 6 are calculated for a very small, but finite value of γ. The dashed curves instead refer to the case of mixed homogeneous-inhomogeneous broadening. The overall behavior of the curves is similar to that of Fig. 5. This is interesting, in view of the different physical meaning of the two sources of broadening: the damping γ describes a decay of the total exciton population, while the parameter Δ describes a redistribution of states within the inhomogeneously broad-

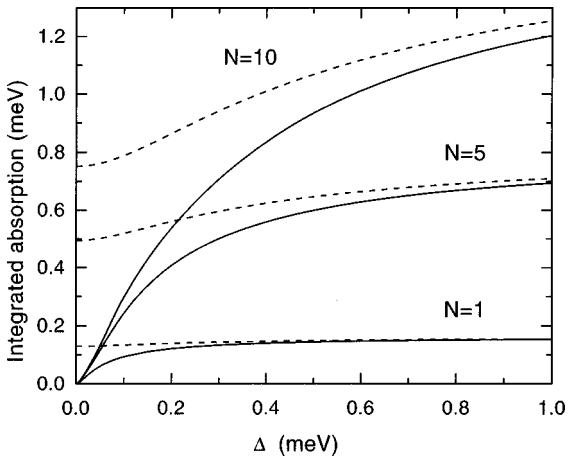

FIG. 6. Integrated absorption (see text) for MQWs with a period $d=20$ nm, for different number of wells, as a function of inhomogeneous broadening. Solid lines, $\gamma=0$; Dashed lines, $\gamma=0.1$ meV.

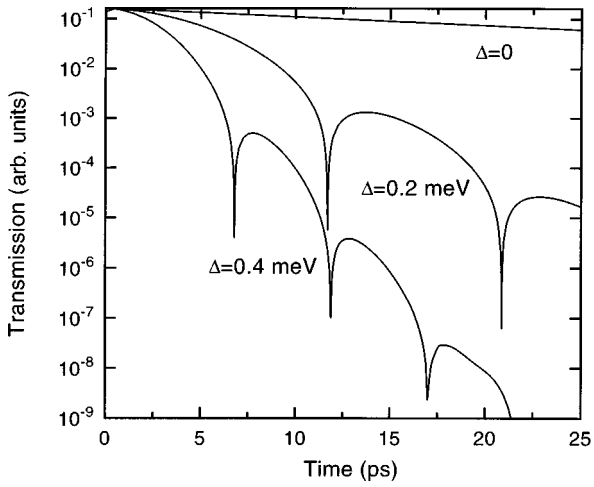

FIG. 7. Transmission response function $|G(\tau)|$ from a single quantum well, with $\Gamma_0=0.026$ meV and $\gamma=0$, for different values of the inhomogeneous broadening.

ened exciton line, without decay of the total population. Nevertheless, the two broadening mechanisms act in a similar way for what concerns integrated absorption.

An obvious question that can now be posed is how the bulk limit is recovered for a large number of wells. The qualitative behavior shown in Figs. 5 and 6 is found to hold also for large N; however, the definition of absorption as $A=1-R-T$ is no longer appropriate in the bulk limit. For a large number of wells, i.e., for a thick enough MQW of thickness x it becomes more convenient to define an absorption coefficient per unit length α in terms of the logarithm of the transmission, or rather

$$T=\frac{(1-R)^2e^{-\alpha x}}{1-R^2e^{-2\alpha x}} \tag{28}$$

when multiple reflections are taken into account.[59] When this definition is adopted, the integrated absorption coefficient $K(T)=\int_{\text{line}}\alpha(\omega)\,d\omega$ is found to have the same behavior as in a homogeneous film with an excitonic resonance, namely, K is a constant when spatial dispersion is absent. Thus the same results may be analyzed in two different ways, according to the definition that is used. The definition (15) is preferable for one or a few QW's, where attenuation of the incoming beam is small and the dimensionless absorption probability $A(\omega)$ has a simpler physical interpretation; the usual definition (28) corresponds to treating the MQW as a thin film of a bulklike material, and becomes useful when the incoming beam is considerably attenuated in propagating through the structure.

V. TIME-RESOLVED SPECTRA

A. Single quantum well

In this section we study the effect of exciton inhomogeneous broadening on time-resolved optical spectra. We calculate the absolute value $|G(\tau)|$ of the response function at finite times $\tau>0$ (i.e., we neglect the δ-function term for transmission). The transmitted intensity is given by $|G(\tau)|^2$.

In Fig. 7 we show the response function $|G(\tau)|$ for time-resolved reflection or transmission for a single QW, assuming $\gamma=0$ and different values of the inhomogeneous broadening Δ. For $\Delta=0$ the response function decays exponentially with the radiative decay rate Γ_0 [Eq. (19)]. For finite values of Δ the transmitted signal decays with a Gaussian envelope and shows oscillations, whose period decreases with increasing Δ. Considering that $\Delta=0.4$ meV corresponds to a FWHM of $2\sqrt{\ln 2}\Delta=0.66$ meV, the curves shown in Fig. 7 should be accessible to experimental verification in high-quality samples.

We emphasize that beatings in the time-resolved signal from a single QW can only be obtained when reabsorption/polariton effects are taken into account, as done in the present treatment by the solution of Maxwell equations leading to Eq. (14). The observation of beatings in coherent time-resolved spectroscopies from QW excitons has been reported several times;[9] however, beatings usually originate from coherent superposition of two quantum transitions, like excitons localized in different interface islands,[60] heavy- and light-hole excitons,[61] upper and lower polaritons,[62] exciton-polariton eigenmodes in MQW's,[54] etc. The prediction of beatings in the time-resolved transmission from a continuous, inhomogeneously broadened exciton in a *single* QW is, to our knowledge, a new and peculiar phenomenon. Thus it is useful to understand the origin of oscillations in physical terms. Two simple arguments are given in the following.

An analytical approximation of the shape of the time-resolved signal can be given in the limit of strong disorder, i.e., $\Delta>\Gamma_0$. In this case the reflection coefficient (12) can be represented as a sum

$$r=\sum_{n=1}^{\infty} f^n, \quad f=-\frac{\sqrt{\pi}\Gamma_0}{\Delta}w(z). \tag{29}$$

Keeping only the first two terms in Eq. (29) one can obtain

$$G(\tau)=-\Gamma_0\exp\left[-\gamma\tau-\frac{\Delta^2\tau^2}{4}-i\omega_0\tau\right] \times\left[1-\sqrt{2\pi}\frac{\Gamma_0}{\Delta}\text{erf}\left(\frac{\tau\Delta}{2\sqrt{2}}\right)e^{\Delta^2\tau^2/8}\right]. \tag{30}$$

One can see from Eq. (30) that this function is nonmonotonic in the presence of inhomogeneous broadening and that $|G(\tau)|$ has a minimum (zero) when the bracket in the right-hand part of Eq. (30) vanishes. Clearly, if we keep only the term linear in susceptibility in the reflection coefficient, the oscillations disappear. In case $\tau \ll 2\sqrt{2}/\Delta$ the condition for the minimum has a simple form

$$\Gamma_0 \tau e^{\Delta^2 \tau^2/8} = 1. \tag{31}$$

In the range of parameters of Fig. 7 the above equation has the approximate solution $\tau \sim n/\Delta$, with a numerical factor n of the order of three to five: this shows that the period of oscillations is inversely proportional to the inhomogeneous broadening parameter.

Interestingly, the oscillations in time-resolved transmission of the single quantum well exist for a wide range of distribution functions for the exciton resonance frequency (not only Gaussian). As far as we judge from numerical simulations, any distribution function decreasing faster than Lorentzian provides nonmonotonic (oscillating in time) signal in transmission. The integral (18) can be done analytically for the distribution function

$$f(x) = \frac{\Delta^3}{\pi} \frac{1}{(x-\omega_0)^4 + \Delta^4}. \tag{32}$$

One can show that in this case oscillations appear if $\Delta > 2\Gamma_0$ and have a period

$$T = \frac{4\pi}{\sqrt{\frac{\Delta^2}{4} - \Gamma_0^2}}. \tag{33}$$

To obtain an intuitive physical understanding of the nonmonotonic shape of the time-resolved transmission (reflection) spectra from a single quantum well at the inhomogeneously broadened exciton resonance, let us consider a wave packet composed by a set of plane waves with amplitudes proportional to $\exp\{-[(\omega-\omega_0)/\Delta]^2\}$, which is a simplified analogy of the wave packet emitted by an inhomogeneously broadened exciton. As a zeroth order approximation let us suppose all these waves to have the same phase. Since

$$e^{i(\omega_0+\nu)\tau} + e^{i(\omega_0-\nu)\tau} = 2e^{i\omega_0\tau}\cos\nu\tau, \tag{34}$$

the time-dependent integrated signal is given by a function

$$G(\tau) \propto \int_0^\infty d\nu\, e^{-(\nu^2/\Delta^2)} \cos\nu\tau = \frac{\sqrt{\pi}}{2}\Delta \exp\left(-\frac{\Delta^2\tau^2}{4}\right), \tag{35}$$

which describes an initial Gaussian-like decay of the transmitted signal in agreement with Eq. (30). However, the function (35) is monotonic. The origin of the minima and oscillations is in the frequency dependence of the phase of the emitting oscillators. As one can see from Eq. (12) or from the inset of Fig. 1, the phase of the reflection coefficient of light changes by π as one tunes the frequency across the exciton resonance. The interference of the light waves emitted with the different phases results in the oscillations of the time-resolved transmission and reflection. In the first order

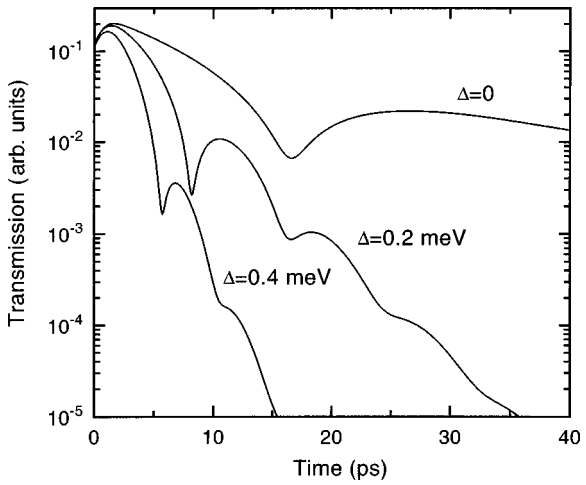

FIG. 8. Transmission response function $|G(\tau)|$ from a multiple quantum well with five wells, of period $d = 20$ nm, with $\Gamma_0 = 0.026$ meV and $\gamma = 0$, for different values of the inhomogeneous broadening.

and in the case of a strong disorder, one can approximate the phase as a function of frequency by

$$\varphi \approx \arctan\frac{\nu}{\Delta}, \tag{36}$$

where $\nu = \omega - \omega_0$. In this case

$$G(\tau) \propto \Delta^{-1} \int_0^\infty d\nu\, e^{-(\nu^2/\Delta^2)} (\Delta\cos\nu\tau - \nu\sin\nu\tau)$$

$$= \frac{\sqrt{\pi}}{2}\Delta \exp\left(-\frac{\Delta^2\tau^2}{4}\right)\left[1 - \frac{\tau\Delta}{2}\right]. \tag{37}$$

Thus we see that even in this oversimplified model the interference of modes having a different phase in the case of inhomogeneous broadening causes the pronounced nonmonotonity of the time-resolved signal that has a minimum $= 2/\Delta$. Physically, the upper and lower wings of the exciton distribution behave as two oscillators with different energies, which are excited in phase by the incoming pulse and decay coherently while producing beatings.

B. MQW's, thick films, microcavities

In Fig. 8 we report the transmission response function from a MQW with five wells, again for different values of Δ. Numerical convergence in the frequency integral was carefully checked: we found that spurious features could arise when parameters of numerical integration were not properly chosen. Already for $\Delta = 0$ oscillations are present, as previously reported;[8,35,53,31] they originate from interference between the various modes of the electromagnetically coupled QW's, and for increasing thickness they go over to the bulk polariton beatings.[62,63] Note, in particular, that this interference causes an initial increase of the time-resolved signal. The effect of inhomogeneous broadening is to decrease the period of oscillations, and to yield a faster (Gaussian) decay of the time-resolved signal. Note that the time-integrated transmitted signal decreases with increase of both homogeneous and inhomogeneous broadening, which is accompa-

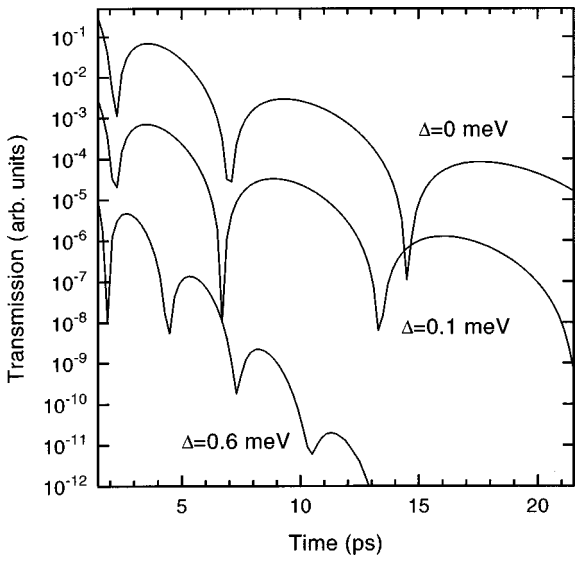

FIG. 9. Transmission response function $|G(\tau)|$ from a 1-μm thick GaAs film, for different values of the inhomogeneous broadening. Other parameters: $\hbar\omega_0=1.52$, $\hbar\omega_{LT}=0.08$ meV, $\gamma=0.1$ meV.

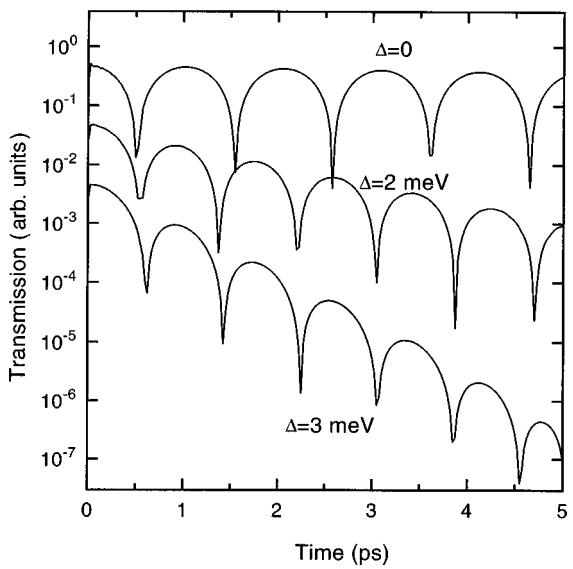

FIG. 10. Transmission response function $|G(\tau)|$ from a λ cavity with one embedded quantum well, for different values of the inhomogeneous broadening. Other parameters: mirror reflectivity $R=0.9994$, Rabi splitting=4 meV.

nied by a simultaneous increase of the integrated absorption. Also, the oscillations are somewhat smeared out for longer times. Similar conclusions were reported by Stroucken *et al.*[8] The results of our Fig. 8 differ in some details from those of Ref. 8, Fig. 3; our curve displays regular oscillations and a fast decay with no plateaus.

On comparing Figs. 7 and 8 it can be noticed that for a given value of Δ the overall decay of the time-resolved transmission signal is much slower for MQW's than for a single QW. This can be interpreted by saying that the disorder-induced decay of the QW exciton is reduced by radiative coupling between QW's: since the excitation is delocalized over the MQW, the effect of disorder is averaged out in the MQW polariton state. This is a kind of quantum ''motional'' narrowing, and is analogous to the phenomenon occurring for polaritons in microcavities.[11,13]

Propagation quantum beats of the quadrupole polariton in Cu_2O were reported by Fröhlich *et al.*[62] They are due to interference of upper and lower polaritons that propagate at the group velocity in a film thicker than the wavelength of light. An analytic expression that describes the polariton beatings was recently given.[63] The quantum beats also require that coherence is maintained over the transit time in the crystal. In Fig. 9 we show the transmission response function through a 1-μm-thick layer of GaAs sustaining an excitonic resonance, for different values of the inhomogeneous broadening. Here again the effect of disorder is to reduce the period of oscillations and to make the whole signal decay with a faster (Gaussian) envelope. It is interesting to note that disorder does not result in a broadening of the structures, but only in a change of their temporal positions.

Semiconductor microcavities with embedded QW's represent a peculiar model system for studying the polaritonic beats. Here, two interfering polariton branches are produced by a confined photon mode in the cavity and an exciton resonance in a quantum well. The splitting between two confined polariton branches, referred to as a Rabi splitting, achieves 7–8 meV in real structures (Ref. 2). This splitting results in quantum beats seen in time-resolved transmission or reflection spectra. The period of these beats is $T=2\pi/\Omega$ (where Ω is the Rabi-splitting frequency), which is much smaller than a period of oscillations in typical MQW systems. Figure 10 shows the calculated response function for time-resolved transmission of a microcavity with parameters close to those of an experimentally studied structure[64] in the absence and in the presence of inhomogeneous broadening. Clearly, the inhomogeneous broadening effect on these beats is much weaker than in the case of single or multiple QW's, since the value of the polariton splitting, which has to be compared with the inhomogeneous broadening, is giant in the case of a microcavity. Thus, even when Δ is of the order of a few meV the oscillations are still seen. The period of oscillations decreases with increase of disorder, and the signal decays faster for stronger disorder. Similar results have been recently obtained within a quantum-mechanical approach to the polaritonic problem by Savona *et al.* (Refs. 10 and 33). The tendency of decrease of the period of oscillations and increase of the decay rate of the signal has been found for all systems considered in this work, including thick films of bulk, MQW's, microcavities, and, surprisingly, single QW's.

We now take up again the question of the validity of the present model, which neglects wave-vector scattering. The total scattered intensity is a small fraction of the incident light, as discussed in Sec. II; however, since the secondary radiation has a finite rise and decay time,[17–20] it could possibly modify the shape of the time-resolved signal in the phase matching directions. For a spectrally narrow excitation the resonant Rayleigh signal decays with the coherence time, but when the whole inhomogeneous line is coherently excited, the decay rate is expected to be faster and dominated by disorder-induced dephasing;[15] in this latter case not only the total (time-integrated), but also the time-dependent elastically scattered light should be a small fraction of the observed signal. Thus our results for the response functions,

which represent the response to a δ pulse, should not be appreciably modified by Rayleigh scattering. In any case a complete answer could only come from a theory of time-resolved response of disordered QW's that includes Rayleigh scattering and hot luminescence together with polariton effects; developing such a theory is a considerable challenge for future research.

VI. CONCLUSIONS

We have studied the effect of disorder on optical properties of excitons in QW's by means of a semiclassical model with a Gaussian distribution of exciton energies. Disorder affects both cw and time-resolved measurements. The presence of inhomogeneous broadening is apparent in the tails of absorption (but not of reflectivity) spectra. The integrated absorption of light in single and multiple QW's (defined as the integral of the optical density) is nonzero only in the presence of a finite damping and increases as a function of either homogeneous or inhomogeneous broadening. For a broadening larger than the radiative width in the coupled QW's the integrated absorption attains a saturation value corresponding to the total oscillator strength of the MQW excitons. This behavior resembles that of exciton-polariton absorption in the bulk, but in the MQW case it is not due to a spatial dispersion effect. These considerations follow from the definition of absorption appropriate to a single QW or a thin MQW, where the incoming light beam has a small probability of being absorbed by the sample; for a thick MQW, when the light beam undergoes exponential attenuation, it is more appropriate to define an absorption coefficient per unit length in terms of the logarithm of the transmission, and in this case the properties characteristic of a thin film (in particular, the fact that the integrated absorption coefficient is a constant in the absence of spatial dispersion) are recovered.

The most peculiar result of the present model is that the time-resolved transmission or reflection from a single QW displays oscillations in the presence of inhomogeneous broadening. These oscillations can be viewed as quantum beats between the upper and lower wings of the exciton line, and can be obtained only if polariton effects in the optical response are included. Approximate treatments substantiate the conclusion that the period of oscillations is inversely proportional to the parameter Δ. The time-resolved transmission in the presence of inhomogeneous broadening has a Gaussian-like decay: thus for an experimental observation it is important that inhomogeneous broadening be not too large, in order that one or a few oscillations can be detected with an appreciable intensity. The present results indicate that the beatings may be observed if the broadening Δ is of the order of 0.4 meV (corresponding to a FWHM of about 0.7 meV). Beatings are also predicted for different kinds of inhomogeneous distributions, i.e., they are not a special feature of a Gaussian distribution.

Quantum beats in the time-resolved transmission are present in MQW's, thin films of bulk, and QW's in microcavities due to coherence between quantum transitions with a common ground state. The effect of disorder is to induce a faster (Gaussian-like) decay of the signal and to decrease the period of beatings. Disorder-induced decay of the time-resolved signal is slower in MQW's than in a single QW: this is a consequence of radiative coupling between QW's, and is a kind of polariton motional narrowing. For a weak disorder the oscillations are not smeared out and the sharpness of structures is preserved. These conclusions are common to the various systems examined and can therefore be taken as general features for the effect of disorder on time-resolved spectroscopies.

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