

Propagation of exciton polaritons in inhomogeneous semiconductor films

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A theoretical study of the propagation of short light pulses in semiconductor slabs containing excitons has been performed using the *scattering-state* technique and the *steepest-descent* method. These methods have allowed us to calculate numerically and to describe analytically the temporary behavior of the dielectric polarization inside the films as well as the time-resolved optical transmission spectra. The inhomogeneous broadening of exciton resonances has been taken into account within a local model. The appearance of a grating of the dielectric polarization in a semiconductor film illuminated by a short light pulse is predicted. This grating, which is due to interfering exciton polaritons, moves backward with respect to the light-propagation direction. We have shown that reabsorption and re-emission of photons by excitons in quantum wells and semiconductor films have a crucial effect on the coherent time-resolved spectra of these structures. The time-resolved transmission decay rate is found to depend on the thickness of semiconductor films with inhomogeneously broadened exciton resonances, which is not the case for a purely homogeneously broadened exciton. This fact, which follows from the energy dependence of the polariton damping in the former case, has also been explained in terms of multiple reabsorption–re-emission of photons by excitons.

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I. INTRODUCTION

Exciton polaritons are quasiparticles exhibiting properties of both photons and excitons, which result in semiconductor crystals from the coupling between an exciton and a photon.^{1–3} An exciton polariton may be considered as a virtual pair of an electron and a hole, which recombine, emitting a photon, and then form an exciton by absorbing the same photon. The importance of exciton polaritons for fundamental issues of light-matter interaction in solids, as well as their strong influence on the optical properties of semiconductor structures, have inspired a huge number of theoretical and experimental works in the last 40 years. Up to now, exciton polaritons in bulk semiconductors, quantum wells, and microcavities, and even in quantum wires and quantum dots, have been studied.⁴ Nevertheless, many important problems of polariton physics still remain unsolved. In particular, the effect of random potential fluctuations on the propagation of polaritons is still an open question, which deserves great consideration and analysis.

The present paper is devoted to a study of the optical response of semiconductors when the exciton resonance is inhomogeneously broadened. In recent decades a great number of experimental and theoretical works were devoted to this problem.^{5–10} The theory has adopted either quantum^{5–7} or semiclassical^{8–10} approaches, which are equivalent in linear optics. Here we will use a semiclassical approach, in which the inhomogeneous broadening is taken into account assuming that, instead of a single exciton resonance energy, the exciton states are distributed in energy with some spectral function $f(\omega)$.

Without going deeply into the details of the different the-

oretical approaches, we would like to point out that one of the principal difficulties for all models is connected with the different scales of roughness affecting excitons and photons. Actually, the excitons are sensitive to potential fluctuations having the scale of their Bohr radius, while the scale of roughness affecting the properties of the photons is much larger (about the wavelength of light in the media). Which scale is important for the mixed exciton-photon state (exciton polariton), and whether exciton polaritons behave as plane waves or as quasiparticles, are still unanswered questions.

The idea that potential fluctuations are averaged by extended exciton-polariton states has inspired a series of works on the *motional narrowing* effect.^{7,9–11} Speaking about motional narrowing one should distinguish between the *horizontal* effect, i.e., averaging in the plane of the light wave,^{7,9} and the *vertical* effect, i.e., averaging of fluctuations in the direction of light propagation.^{10,11} In both cases the theoretical model supposes the existence of extended exciton-polariton modes having macroscopic dimensions.

On the other hand, consideration of excitons localized in the fluctuation potential as individual scatters yields the effect of the Rayleigh scattering of light recently discussed and observed experimentally. Plane-wave and individual-scatter approaches to the exciton-polariton problem in disordered media are similar in some extent to the Bragg and von Laue models of x-ray diffraction by crystals.¹² The difference is that in the von Laue model all atoms are identical and the crystal has a regular lattice (which yields a formal equivalence between the von Laue and Bragg models) while in our case the excitons from the inhomogeneous distribution have different energies, and are randomly distributed in the fluc-

tuation potential. That is why the conclusions of the ‘‘Laue-like’’ model⁵ are somewhat different from the conclusions of the ‘‘Bragg-like’’ model^{7,10,13} (mostly, the difference is related to the effect of Rayleigh scattering of light by localized excitons).

In reflection or coherent transmission geometry, the difference between the two approaches does not seem to be crucial. A theoretical study of reflection of light by a regular grating of identical quantum dots¹⁴ yielded the same form of the reflection and transmission coefficients as in a homogeneous quantum-well (QW) model (which confirms the analogy with the Bragg–von Laue case). Estimations¹³ have shown that even for an irregular grating of nonequivalent quantum dots with real parameters, only a small fraction (less than 1%) of light is scattered, while most of the photons keep their in-plane wave-vector constant. The Bragg-like model has allowed one to explain both *horizontal*¹⁵ and *vertical*^{10,11} motional narrowing effects in microcavities and multiple QW structures, respectively.

An important advantage of the Bragg-like model consists in its simplicity. Actually, it allows one to find the linear optical response of complicated semiconductor structures with the use of Maxwell equations, taking fully into account the acts of absorption and emission of light by excitons (i.e., the polaritonic effect).

In the present work we address the problem of propagation of exciton polaritons excited by short pulses of light in disordered media, adopting a Bragg-like semiclassical model of extended exciton-polariton states whose in-plane size is much larger than all fluctuation scales in the problem. We assume that the photon (polariton) wave vector is conserved in the plane, and that there is no Rayleigh scattering. Our model semiconductor film is optically homogeneous both in the plane and in the growth direction. Moreover, we neglect spatial dispersion of exciton-polaritons.

Within these approximations we were able to find the temporary dynamics of the dielectric polarization due to propagation of exciton polaritons arising in semiconductor films after illumination by a short light pulse. Both cases of homogeneous and inhomogeneous broadening of excitons have been analyzed. Unexpected results have been found: in particular, the appearance of a grating of dielectric polarization which moves backward with respect to the direction of light propagation. The grating results from the interference between exciton polaritons belonging to the upper and lower branches, respectively, and is seen both in cases of homogeneous and inhomogeneous broadening. The time-resolved transmission spectra of light differ drastically for the two mechanisms of exciton broadening. That is, the decay rate of the time-resolved transmission is independent of the slab thickness in the case of homogeneous broadening, whereas it depends strongly on the thickness in the case of inhomogeneous broadening. The period of oscillations due to the interference between the two polariton branches is also quite sensitive to the broadening mechanism.

We have described these phenomena analytically using the *steepest-descent* method, and explained them in terms of classical optics. In addition, the quantum model of exciton-polariton propagation in disordered media has been developed, yielding a picture which is qualitatively similar to the mesoscopic model for the electronic transport.¹⁶ This leads

to the expectation that typical mesoscopic effects like weak localization¹⁷ should also appear for exciton polaritons.

The paper is organized as follows: In Sec. II we present our formalism. Both the *scattering-state* approach and the analysis based on the steepest-descent method have been developed, in order to describe the propagation of exciton polaritons in disordered media. Section III is devoted to discussion of the time-resolved transmission and of the time- and frequency-dependent polarization for a homogeneously broadened slab. These results are to be compared with those for an inhomogeneously broadened media, which are presented in Sec. IV. Section V contains concluding remarks, and addresses the mesoscopic effects in the propagation of exciton-polaritons in a disordered media. The derivation of the transmitted signal with the steepest descent method in the presence of inhomogeneous broadening is given in the Appendix.

II. MODEL AND FORMALISM

A. Semiclassical approach to the exciton-polariton problem

We describe the interaction of light with excitons in semiconductor structures by means of the linear semiclassical approach. In the framework of linear-response theory, Maxwell equations for light incident on the structure may be written in the form:

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

where

$$\mathbf{D}(z) = \varepsilon_B \mathbf{E}(z) + 4\pi \mathbf{P}_{\text{exc}}(z). \quad (2)$$

ε_B is the background dielectric constant, and $\mathbf{P}_{\text{exc}}(z)$ is the excitonic contribution to the dielectric polarization. In the local model the relation between $\mathbf{P}_{\text{exc}}(z)$ and $\mathbf{E}_{\text{exc}}(z)$ is given by

$$\mathbf{P}_{\text{exc}}(z) = \chi_b \mathbf{E}(z), \quad (3)$$

with

$$\chi_b(\omega, \omega_0) = \frac{1}{4\pi} \frac{\varepsilon_b \omega_{\text{LT}}}{\omega_0 - \omega - i\gamma}, \quad (4)$$

where ω_0 and ω_{LT} represent, respectively, the exciton resonance frequency and the exciton longitudinal-transverse splitting in the bulk material, and γ is the exciton nonradiative homogeneous broadening, which we suppose to be frequency independent. Throughout the paper we will assume the background refractive indices $n_b = \sqrt{\varepsilon_b}$ of the slab and of the surrounding medium to be the same, an assumption which allows us to avoid complications caused by all the optical interference effects.

The local description is appropriate for thick semiconductor films; it may also be used for QW's when the conditions $\omega_{\text{LT}} \ll \gamma$ and $\sqrt{\varepsilon_b}(\omega_0/c)L_z \ll 1$ are fulfilled, where L_z is the QW width.¹⁸

In order to describe the inhomogeneous broadening we assume that the excitons have a frequency distribution centered at the frequency ω_0 given by a function $f(\omega, \omega_0)$. As discussed in Ref. 13 in detail, the coherent contribution of all

excitons from the distribution is correctly taken into account within the constant in-plane wave-vector approximation by replacing the dielectric susceptibility χ_b [Eq. (4)] by that averaged with the exciton distribution function:

$$\chi(\omega) = \int_{-\infty}^{\infty} \chi_b(\omega, \mathbf{v}) f(\mathbf{v}, \omega_0) d\mathbf{v}. \quad (5)$$

We emphasize here that solving Maxwell equations with the weighted dielectric susceptibility is not equivalent to solving Maxwell equations for a single-exciton resonance with a further averaging of the electric field. Such a difference is quite essential because the two procedures describe different schemes of interaction between excitons and light. This point will be analyzed in detail in Sec. IV.

In this work we have assumed the exciton distribution function to be a Gaussian:

$$f(\omega, \omega_0) = \frac{1}{\sqrt{\pi}\Delta} \exp\left[-\left(\frac{\omega_0 - \omega}{\Delta}\right)^2\right]. \quad (6)$$

Δ will be referred to as the parameter of inhomogeneous broadening. The advantage of the Gaussian distribution is the possibility of calculating analytically the susceptibility (5), which becomes

$$\chi(\omega) = i \frac{\varepsilon_b}{4\sqrt{\pi}} \frac{\omega_{LT}}{\Delta} w(\zeta), \quad (7)$$

where $\zeta = (\omega - \omega_0 + i\gamma)/\Delta$, and $w(\zeta) = e^{-\zeta^2} \text{erfc}(-i\zeta)$, $\text{erfc}(\zeta)$ being the complementary error function.¹⁹

B. Scattering-state approach

The scattering-state approach allows us to calculate numerically the time-dependent dielectric polarization $P_{\text{exc}}(z, t)$ induced in a semiconductor film (or in a QW) by a short light pulse with a spectral function $g(\omega)$.^{20,21} In this framework Maxwell equations are solved in the frequency domain with appropriate boundary conditions. The frequency- and coordinate-dependent solutions of Eq. (1) called scattering states of the system $E(z, \omega)$ are Fourier-integrated in order to obtain the time-dependent dielectric polarization:

$$P_{\text{exc}}(z, t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega g(\omega) \chi_b E(z, \omega) \exp(-i\omega t). \quad (8)$$

The time-resolved reflection and transmission of the structure are given, respectively, by

$$R(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega g(\omega) [E(0, \omega) - 1] \exp(-i\omega t). \quad (9)$$

$$T(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega g(\omega) E(d, \omega) \exp(-i\omega t), \quad (10)$$

where $z=0$ and $z=d$ correspond to the beginning and end of the structure.

The scattering states $E(z, \omega)$ are easily found using the *transfer-matrix* technique.²² Within this method, each layer of the structure is characterized by a 2×2 matrix connecting

the in-plane components of electric and magnetic fields at the beginning and end of the layer. The transfer matrix across the entire structure is a product of the transfer matrices across all the layers,

$$\tilde{M} = \prod_{j=N}^0 \tilde{M}_j, \quad (11)$$

where the layers are numbered in order of the propagation of light through the structure. The amplitude reflection coefficient of the entire structure depends on the elements m_{ij} of the matrix \hat{M} as

$$r(\omega) = \frac{n_b(m_{11} + m_{12}n_b) - (m_{12} + m_{22}n_b)}{n_b(m_{11} + m_{12}n_b) + (m_{12} + m_{22}n_b)}. \quad (12)$$

Now the scattering states of the system can be found as

$$E(z, \omega) = m_{11}^z [1 + r(\omega)] + m_{12}^z [1 - n_b r(\omega)], \quad (13)$$

where m_{ij}^z are the elements of the matrix \hat{M}_z describing the transfer from the beginning of the structure till the point z which can be found as a product of the transfer matrices of individual layers from z till 0 in the same way as the matrix \hat{M} [Eq. (11)].

C. Analytical calculation of the time-resolved transmission

The transmitted signal $T(t) = \int_{-\infty}^{\infty} dt' E_I(t') G(t-t')$ is calculated as the convolution between the incident pulse $E_I(t)$ and the response function $G(t)$. $G(t)$ coincides with $T(t)$ for an incident δ pulse, and is expressed in terms of the transmission coefficient $t(\omega)$ as $G(t) = \int_{-\infty}^{\infty} (d\omega/2\pi) e^{-i\omega t} t(\omega)$, with

$$t(\omega) = \frac{4n_b n(\omega)}{[n_b + n(\omega)]^2 e^{-ikd} - [n_b - n(\omega)]^2 e^{ikd}}.$$

Here $n(\omega)$ is the complex refractive index $n(\omega) = n_b \sqrt{1 + 4\pi\chi(\omega)/\varepsilon_b}$, $k = k(\omega) = n(\omega)\omega/c$, and d is the thickness of the semiconductor slab. When $d \gg \lambda$, where λ is the wavelength of the light in the medium, multiple reflections at each interface of the slab—which are described by the second term in the denominator of $t(\omega)$ —may be disregarded, so that $G(t)$ becomes

$$G(t) = \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{4n_b n(\omega)}{[n_b + n(\omega)]^2} e^{i\Phi(\omega)}, \quad (14)$$

with $\Phi(\omega) = \omega[n(\omega)d/c - t]$ being a rapidly oscillating phase. $G(t)$ may now be approximately evaluated with the steepest descent method^{23,24} [which coincides with the stationary phase method for $\text{Im}\Phi(\omega) = 0$]. This approach is based on the observation that the integral in Eq. (14) is dominated by frequencies corresponding to large values for $e^{i\Phi}$, i.e., by the saddle points for $\text{Im}\Phi(\omega)$ (in fact the imaginary part of an analytical function does not have maxima or minima). Thus, in order to calculate the integral, it is convenient to choose integration paths passing through the saddle points and concentrating large values for $e^{-\text{Im}\Phi}$ on the shortest interval. These paths are simply determined using the

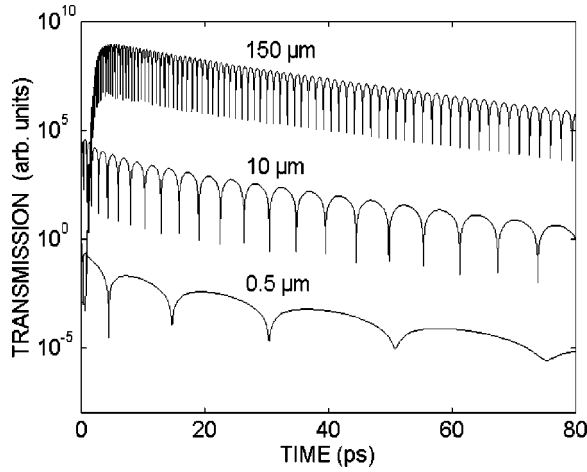


FIG. 1. Time-resolved transmission through slabs of GaAs of different thicknesses: 150, 10, and 0.5 μm . The excitonic parameters used in this calculation are $\hbar\omega_{LT}=0.08$ meV, $\hbar\omega_0=1515$ meV, and $\hbar\gamma=0.05$ meV. The background refractive index is 3.54. The 300-fs-long incident pulse is centered at the exciton resonance frequency.

Riemann condition for analytical functions, which states that the lines of maximum variation for $\text{Im}\Phi(\omega)$ are equipotential lines for $\text{Re}\Phi(\omega)$.

III. PROPAGATION OF EXCITON POLARITONS IN HOMOGENEOUS FILMS

A. Time-resolved transmission

Figure 1 shows the time-resolved transmission of GaAs films of different thicknesses induced by a 300-fs-long pulse of light; the thicknesses have been varied in the range between 500 and 150 μm . The excitonic parameters used in this calculation are: $\hbar\omega_{LT}=0.08$ meV, $\hbar\omega_0=1515$ meV, $\hbar\gamma=0.05$ meV, and $n_B=3.54$. The central energy of the pulse incident on the structures has been chosen equal to the exciton resonance energy. In Ref. 23, $G(t)$ was evaluated with the steepest-descent method for a homogeneous medium described by the local susceptibility χ_b [Eq. (4)], in the time window $(d/v)\ll t\ll(d/2v)(\omega_0/\omega_{LT})$ ($v=c/n_b$). It was shown that the response function is given by

$$G(t) = \frac{1}{\sqrt{2\pi}} \left(4 \frac{d\omega_c^2}{v\hat{t}^3} \right)^{1/4} e^{i\Phi_0^{(\text{hom})}} e^{-\gamma\hat{t}} \cos \left(\Delta\Phi_S^{(\text{hom})} + \frac{\pi}{4} \right), \quad (15)$$

where

$$\Delta\Phi_S^{(\text{hom})} = 2\pi c \left(\frac{d}{v} \right)^{1/2} \hat{t}^{1/2} \quad (16)$$

and

$$\Phi_0^{(\text{hom})} = -\omega_0\hat{t} - \frac{d\omega_{LT}}{v}. \quad (17)$$

ω_c is the polariton splitting: $\omega_c = (\omega_0\omega_{LT}/2)^{1/2}$, and $\hat{t} = t - d/v$.

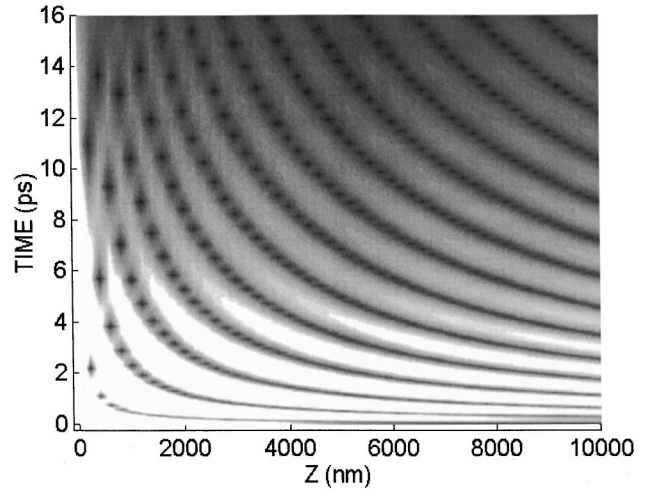


FIG. 2. Coordinate- and time-dependent dielectric polarization induced by excitons excited by a 300-fs light pulse in a 10- μm -thick slab of GaAs with the same parameters as in Fig. 1. The brightness of the color is proportional to the absolute value of the dielectric polarization.

As one can see from Fig. 1 and Eq. (15), in the case of pure homogeneous broadening the decay rate of the signal is independent of the slab thickness. Moreover with the parameters of Fig. 1 the exponential decay $\exp(-\gamma\hat{t})$ largely overwhelms the $\hat{t}^{-3/4}$ decay entailed by the pre-exponential factor. The spectra exhibit pronounced oscillations; the time spacing $\tau^{(\text{hom})}$ between two successive minima depends on the time and on the thickness of the slab, and is given by

$$\tau^{(\text{hom})} = \frac{\pi}{\omega_c} \left(\frac{vt}{d} \right)^{1/2}, \quad (18)$$

as may be deduced from Eq. (15). The oscillations represent a typical feature of interference between exciton-polariton branches, which was already experimentally observed²⁵ a few years ago. The nature of the beats can be simply understood if one considers the exciton polaritons as wave packets composed of plane waves having different frequencies and different group velocities. The intensity of the transmitted signal at time t is governed by the polariton waves with group velocity $v = \partial\omega/\partial k = d/t$.

For each group velocity one can find two different polariton waves coming from the two different branches. Interference between these two waves gives rise to oscillations in time-resolved transmission spectra. Note the picosecond time scale in the spectra, which is a typical time scale for the exciton-polariton transport processes (to be distinguished from the time-of-flight of the free photon across the structure, which is about 100 fs for a 10- μm -thick slab). Longer times correspond to the interference between polaritons with larger group velocity, i.e., between polaritons which are closer to resonance; the decrease of the splitting between two interfering modes—which eventually reaches the longitudinal-transverse splitting ω_{LT} —corresponds to an increase of the period of oscillations with time given by the inverse of the frequency splitting.

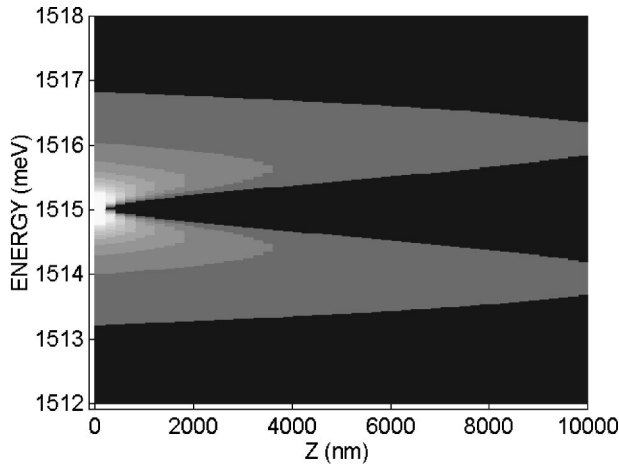


FIG. 3. Coordinate- and frequency-dependent dielectric polarization induced by excitons excited by a 300-fs light pulse in a 10- μm -thick slab of GaAs with the same parameters as in Fig. 1. The brightness of the color is proportional to the absolute value of the dielectric polarization.

B. Time-dependent dielectric polarization

Figure 2 shows the temporary evolution of the excitonic polarization in a 10- μm -thick GaAs slab excited by a short light pulse, as a function of coordinate and time calculated within the scattering state approach [Sec. II, Eqs. (9) and (13)]. All the other parameters are the same as Fig. 1; the brightness of color is proportional to the absolute value of the dielectric polarization.

One can see that the dielectric polarization in Fig. 2 is a strongly nonmonotonic function of the coordinate and the time. That is, a pronounced grating of the dielectric polarization appears. The period of oscillation increases as a function of time, and decreases as a function of the coordinate, which is consistent with Eq. (18) (where one should replace the layer thickness d with the coordinate z). Interestingly, the grating moves backward with respect to the light propagation direction, which seems, at first glance, to be in contradiction with the wave-vector conservation law. Note, however, that the excitons in our model are infinitely heavy, so that the motion of the dielectric grating cannot be associated with the motion of excitons, but only with absorption and coherent re-emission of light by different excitons in the structure. Thus the wave-vector conservation law is not violated in this calculation.

Let us interpret the peculiar behavior of the excitonic polarization in terms of reabsorption-re-emission of light by excitons. At very short times the polarization is homogeneous, which is a natural consequence of the homogeneity of the film. Then coherent re-emission of light starts: photons re-emitted at different points z_1 and z_2 interfere at z_3 , and a z -dependent intensity pattern develops, which in turn influences the reabsorption processes responsible for further changes of the dielectric polarization, thus producing an intensity pattern which is also time dependent. The time independence of the polarization at $z=0$ follows from the fact that—due to wave-vector conservation—no interference pattern produced by photons re-emitted at different points arises, and the polarization is only determined by the photons emitted at $z=0$.

Appearance of the grating may also be analyzed from the point of view of the scattering-state model. Figure 3 shows the excitonic polarization of a 10- μm GaAs slab defined as

$$P_{\text{exc}}(z, \omega) = g(\omega) \chi_b E(z, \omega) \quad (19)$$

as a function of frequency and coordinate. Once again, the brightness of the color is proportional to the absolute value of the dielectric polarization. One can see that the initial single-resonance distribution of the dielectric polarization at the exciton resonance frequency drastically changes as one goes deeper into the semiconductor medium. A pronounced camelback structure appears, with two maxima split by about 2 meV. These maxima are associated with eigenenergies of exciton polaritons propagating in the media [these are the eigenstates in the (ω, z) space, to be distinguished from those in the (ω, k) space]. Clearly, the beats in temporary dynamics of the polarization are due to the double-resonance structure in its frequency-resolved spectra. Such a structure is characteristic of the regime of strong coupling between excitons and photons. The splitting of the maxima of the polarization is roughly proportional to the number of acts of absorption and re-emission of a photon by an exciton during the propagation of a polariton mode, and thus increases with the coordinate. The period of the beats in time is inversely proportional to the splitting between the two eigenstates of the system, so that it decreases with the coordinate. Thus the behavior of the excitonic polarization in the frequency domain fits well into the quantum absorption–re-emission model.

The classical model explains appearance of the camelback structure by the interplay between transmission and absorption of light in a semiconductor slab. If we neglect the reflection by the surface, the intensity of the transmitted light at the point z can be represented as

$$T(z, \omega) = I - \xi(z) \alpha(\omega), \quad (20)$$

where $\alpha(\omega)$ is the absorption coefficient, and $\xi(z)$ is a monotonically increasing function of z . Since

$$P_{\text{exc}}(z, \omega) \propto \alpha(\omega) T(z, \omega), \quad (21)$$

the peak of the excitonic polarization experiences a bifurcation at a certain value of z , which is seen in Fig. 3.

IV. EXCITON POLARITONS IN THE CASE OF INHOMOGENEOUS BROADENING

A. Weighted susceptibility

It was pointed out in Sec. II that the correct way to describe the inhomogeneous broadening within the constant in-plane wave-vector approximation is to consider the weighted dielectric susceptibility [Eq. (5)] and not the weighted optical response (reflection or transmission coefficient) of the structure. Due to its relevance for all the following discussion of the inhomogeneous broadening effects, we will address this question here in detail.

The physical difference between the solution of Maxwell equations obtained considering first a single-exciton resonance and then averaging the electric field related to different resonances, and that with the weighted susceptibility, may be easily understood with the aid of Fig. 4. In the former case

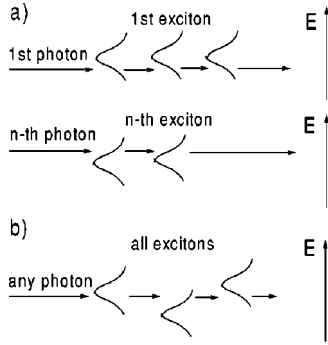


FIG. 4. Illustration of the two schemes of interaction between a photon and an inhomogeneously broadened exciton resonance (see the text). (a) First the optical response of each single exciton state belonging to the inhomogeneous distribution is found, then the total response of the structure is calculated as the sum of the individual responses. (b) First the dielectric susceptibility containing contributions of all excitons is calculated, then the light interaction with a layer characterized by this susceptibility is considered.

[Fig. 4(a)] each given photon can interact only with a single-exciton resonance from the frequency distribution; thus the photon is always emitted and reabsorbed by the same exciton. The averaging of all single-photon–single-exciton interaction schemes yields the optical response of the system within this model.

On the other hand, if Maxwell equations are solved with the weighted dielectric susceptibility [Eq. (5)], a photon emitted by a given exciton is allowed by the model to be absorbed by another exciton from the distribution [Fig. 4(b)]. Thus averaging of the dielectric susceptibility allows one to take into account all possible chains of coherent absorption-emission acts. The difference between the two approaches is almost negligible for cw optical spectra, but it appears to be huge in time-resolved spectra where fine interference effects play an important role.

This is illustrated in Fig. 5 which shows the cw (inset) and time-resolved reflection spectra of a single QW. We have used the following set of excitonic parameters $\hbar\omega_{LT} = 0.095$ meV, $\hbar\gamma = 0.05$ meV and $\hbar\Delta = 1.9$ meV. These parameters have yielded the best fit to the experimental reflection and absorption spectra of a 172-Å-thick $\text{In}_{0.06}\text{Ga}_{0.94}\text{As}/\text{GaAsOW}$.²⁶ The time-resolved reflection spectrum [Fig. 5(a)] and the cw reflection spectrum [$R(\omega) = |r_{\text{QW}}(\omega)|^2$; inset to Fig. 5, solid line] have been calculated within the local model according to Eqs. (9) and (13), with the averaged dielectric susceptibility (7). Note that in Eq. (9) we substituted

$$E(0, \omega) - 1 = r_{\text{QW}}. \quad (22)$$

These spectra are to be compared with time-resolved and cw reflection spectra [Fig. 5(b) and the dashed line in the inset to Fig. 5] calculated by averaging the reflection coefficient of the quantum well,

$$\bar{r}_{\text{QW}}(\omega, \omega_0) = \int_{-\infty}^{+\infty} r_{\text{QW}}(\omega, \omega'_0) f(\omega'_0, \omega_0) d\omega'_0, \quad (23)$$

where $r_{\text{QW}}(\omega, \omega_0)$ is given by Eq. (12) with the dielectric susceptibility (4). The time-resolved reflection in this case

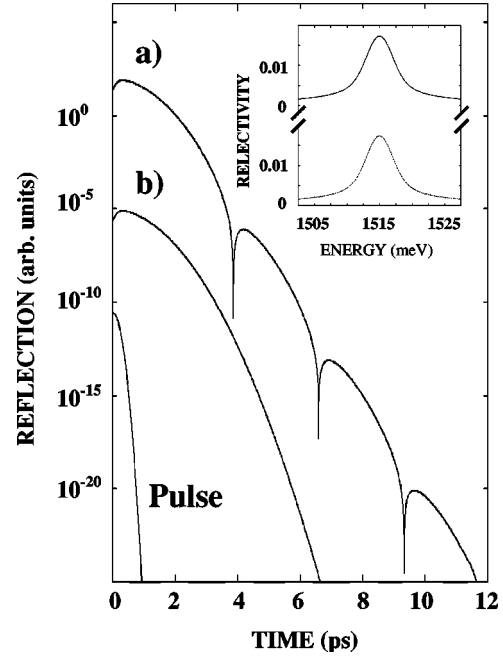


FIG. 5. Calculated time-resolved reflectivities for a single QW containing an inhomogeneously broadened exciton resonance. (a) The inhomogeneous broadening is taken into account in the dielectric susceptibility of the QW. (b) Frequency-resolved reflection spectra calculated for individual exciton resonances from the inhomogeneous distribution are averaged with the exciton frequency distribution function. Time-resolved spectra were obtained by an inverse Fourier transform of the frequency-resolved spectra [shown in the inset by solid (a) and dashed (b) lines] convoluted with the spectral function of the incident pulse (shown in the figure).

has also been found from Eq. (9), with a 300-fs-long pulse shown in Fig. 5, and with the substitution $E(0, \omega) - 1 = \bar{r}_{\text{QW}}$.

One can see that the time-resolved reflection spectrum calculated with the averaged dielectric susceptibility exhibits pronounced oscillations that are absent in the spectrum calculated with the averaged reflection coefficient. Quantum beats in time-resolved spectra of a QW with a single inhomogeneously broadened exciton resonance were predicted in Ref. 13. A direct experimental evidence of the beats has not yet been reported to the best of our knowledge, while an indirect proof was recently obtained.²⁷ Appearance of the beats in the time-resolved response of a *single* optical resonance is a striking and unexpected effect. (Note that usually the beats take place between *two* discrete, closely lying quantum states).

In Ref. 13 the beats were interpreted in the framework of the semiclassical approach as a result of interference of photons emitted at frequencies slightly lower and slightly higher than ω_0 . The beats may also be interpreted in the framework of the quantum absorption–re-emission model. Considering the propagation of exciton polaritons in a semiconductor as a chain of emission-absorption acts (as illustrated in Fig. 4), we attribute the beats to the interference between those photons which interact with only one exciton resonance from the energy distribution [they induce a monotonic decay of $R(t)$ shown in Fig. 5(b)] and those which interact with different excitons during their lifetime in a QW. Passing through dif-

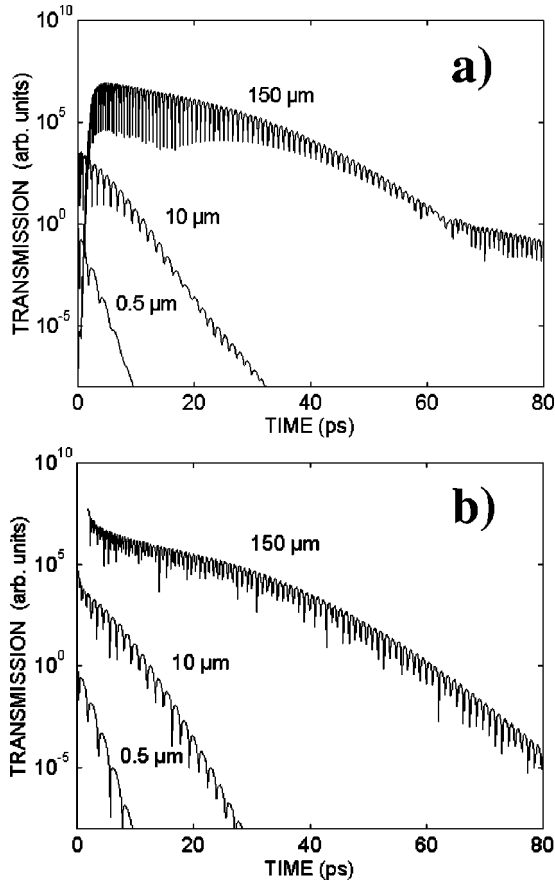


FIG. 6. Time-resolved transmission through slabs of GaAs of different thickness: 150, 10, and 0.5 μm . (a) Numerical calculation by the scattering state method. (b) Results of the analytical model. The excitonic parameters used in this calculation are $\hbar\omega_{LT} = 0.08$ meV, $\hbar\omega_0 = 1515$ meV, $\hbar\gamma = 0.05$ meV, and $\hbar\Delta = 1$ meV. The 300-fs-long incident pulse is centered at the exciton resonance frequency.

ferent chains of emission-absorption acts, the photons gain different phases which gives rise to interference.

To some extent this makes propagation of exciton-polaritons in quantum structures containing inhomogeneously broadened exciton resonances analogous to the transport of electrons in the mesoscopically disordered media. This analogy will be discussed in detail in Sec. V of this paper. It should be noted that the emission-absorption acts we are speaking about are virtual. Moreover, in the case of a QW there is no real propagation of exciton polaritons, since the excitons are confined in the z direction. The analogy relies on the fact that in a QW in the presence of inhomogeneous broadening, the photon is coupled with *all* excitons from the inhomogeneous contour, which means that during the polariton lifetime it can be virtually emitted or absorbed by different excitons from the distribution, in the same way as an electron may be scattered by any scattering center in the medium.

B. Time-resolved transmission through inhomogeneous slabs

Figures 6(a) and 6(b) show the time-resolved transmission of GaAs films of different thicknesses (from 500 nm to 150 μm) induced by a 300-fs pulse of light. The pulse is centered

at the exciton resonance frequency, while its spectral width much exceeds the exciton broadening. The excitonic parameters used for this calculation are the same as in Fig. 1; the only difference is that in Fig. 6 the exciton has been assumed to be also inhomogeneously broadened with $\hbar\Delta = 1$ meV. A striking agreement between the numerical calculation, performed with the scattering-state method [Fig. 6(a)], and the analytical one, which is discussed later in this section and in the Appendix [Fig. 6(b)], is obtained.

A remarkable feature of these spectra is that—in contrast to the homogeneous case—the decay rate of the transmission depends strongly on the thickness of the slab. That is, it decreases with an increase of the thickness, finally approaching (for the 150- μm -thick slab) a value close to the decay rate in the case of only homogeneous broadening, as one can judge comparing Figs. 6 and 1. Note also the different dependence on time and thickness of the slab with (Fig. 6) and without (Fig. 1) inhomogeneous broadening. Clearly, time-resolved transmission is very sensitive to the mechanism of the exciton broadening.

The observed features are easily interpreted with the aid of our analytical predictions. The main steps of the derivation of the time-resolved transmission of a δ pulse in the presence of inhomogeneous broadening through a slab characterized by the local susceptibility (7) with the steepest-descent method, and the obtained expressions are given in the Appendix; here we discuss the results.

Homogeneous and inhomogeneous broadenings contribute both to the decay of the signal [Eq. (A3)] through the term $\exp(-\text{Im}\Phi_s)$. As far as γ is supposed to be energy independent, the former produces a thickness-independent exponential decay. On the other hand, the latter is responsible for a nonexponential, time-varying decay rate, since exciton polaritons have an energy-dependent damping in the presence of inhomogeneous broadening. Polaritons which are closer to resonance exhibit a larger damping, whereas—for reasonable values of Δ —those with a larger splitting have a damping which is close to the homogeneous one. For this reason the decay is also expected to be thickness dependent: polaritons which determine the transmitted signal at a fixed time t are closer to resonance on decreasing the slab thickness, which in turn implies a faster decay for thinner slabs.

Let us first examine the case $(\Delta/\Omega) \ll 1$: we will refer to this situation as the “small disorder case.” Using the asymptotic expansion of $w(\zeta)$ for $\zeta \rightarrow \infty$, and retaining only corrections up to the lowest order in (Δ/Ω) to Eq. (A1), the stationary points ω_s are found to be given by

$$\omega_s = \omega_0 \pm \Omega \left[1 + \frac{3}{4} \left(\frac{\Delta}{\Omega} \right)^2 \right] - i\gamma. \quad (24)$$

(with plus and minus signs referring to the upper and lower polariton branches, respectively). The resulting transmitted signal is given by

$$G(t) = \frac{1}{\sqrt{2\pi}} \left(4 \frac{d\omega_c^2}{\nu \hat{\tau}^3} \right)^{1/4} \left[1 - \frac{3}{8} \left(\frac{\Delta}{\Omega} \right)^2 \right] e^{-\gamma \hat{\tau}} e^{i\Phi_0^{(\text{hom})}} \times \cos \left(\Delta \Phi_s + \frac{\pi}{4} \right), \quad (25)$$

with

$$\Delta\Phi_S = \Delta\Phi_S^{(\text{hom})} \left[1 + \frac{1}{4} \left(\frac{\Delta}{\Omega} \right)^2 \right]; \quad (26)$$

all the other quantities have been explained in the Appendix.

Equation (24) shows that in this case Δ produces only a small correction in the real part of the eigenenergies of the polariton modes. This, in turn, implies that the decay of the transmitted signal is still quasiexponential, as in the homogeneous case; however, the period of the beats is modified and shortened due to the presence of the inhomogeneous broadening, and is given by

$$\tau^{(\text{inh})} = \frac{\tau^{(\text{hom})}}{1 + \frac{3}{4} \left(\frac{\Delta}{\Omega} \right)^2}. \quad (27)$$

The dependence of the time-resolved transmission on the thickness of the slab may also be qualitatively explained with the quantum model, schematizing the exciton-polariton propagation as a chain of virtual emission-absorption acts.

In case of homogeneous broadening only, any photon is always absorbed and re-emitted by the same exciton characterized by the homogeneous broadening γ , which governs the decay time of the time-resolved transmission. On the other hand, a photon can be absorbed and re-emitted by any exciton from the frequency distribution in the case of inhomogeneous broadening, as already discussed in Sec. IV A. Suppose that during its motion across the slab the photon experiences N absorption–re-emission acts (clearly, on average, N increases with increase of the thickness of the slab). If $N = 1$ (which might be called “the QW limit”) the probability ρ for a photon having a frequency ω to be absorbed by an exciton with a frequency ω' depends on the imaginary part of the variation of the dielectric susceptibility due to this exciton, and on the probability of finding this exciton. Thus, it can be estimated as²⁷

$$\rho(\omega, \omega') \propto \frac{\varepsilon_B \omega_{LT} \gamma}{(\omega' - \omega)^2 + \gamma^2} f(\omega', \omega_0). \quad (28)$$

Equation (28) shows that a photon can coherently interact only with excitons situated in the γ vicinity of its frequency ω . Moreover, photons with frequencies close to ω_0 have better chances to be absorbed by an exciton than those having frequencies far from the center of the excitonic distribution. For the same reason, excitons in the center of the distribution have better chances to be excited by photons than those from the wings of the distribution. In this QW limit the transmission decays with a Gaussian envelope and decay parameter given by $1/\Delta$, as discussed in detail in Ref. 13.

Now suppose that $\Delta \gg \gamma$. The probability that the same photon is virtually absorbed and re-emitted by an exciton from its γ vicinity N times is proportional to $f^N(\omega', \omega_0)$. With increasing N this function becomes sharper and sharper, finally approaching the Dirac delta function $\delta(\omega' - \omega_0)$. In turn, N must increase with increasing slab thickness, since in our model the exciton polaritons are supposed to maintain spatial coherence over the whole slab thickness. (Note that this is still true when dephasing processes—neglected in our model—are taken into account, provided the

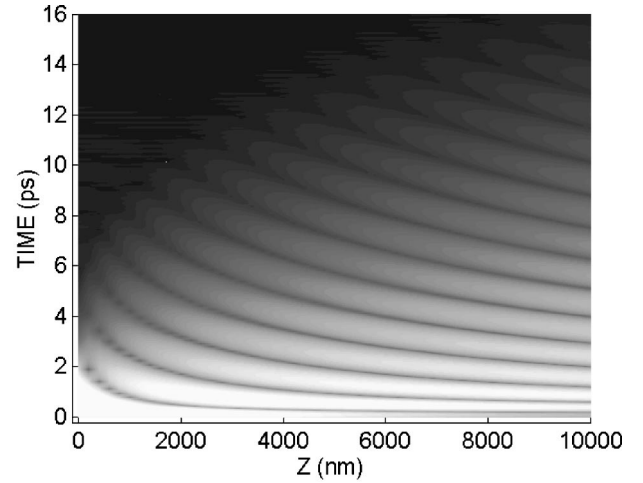


FIG. 7. Coordinate- and time-dependent dielectric polarization induced by excitons excited by a 300-fs light pulse in a 10- μm -thick slab of GaAs with the same parameters as in Fig. 6. The brightness of the color is proportional to the absolute value of the dielectric polarization.

thickness of the slab does not exceed the coherence length of polaritons.) That is why in the case of *very thick* films only an exciton-polariton mode with a frequency $\omega = \omega_0$ survives. This mode has a homogeneous broadening γ which governs its decay rate. So, in this limit, the influence of inhomogeneous broadening on the decay time of the transmitted signal is completely suppressed. This is what we see in Fig. 6 (the case of the 150- μm -thick slab).

Variation of the transmission decay time with the thickness of the sample is a manifestation of the *vertical motional narrowing* effect. Actually, the origin of the effective narrowing of the frequency distribution of exciton polaritons with an increase of the thickness of the slab is the averaging of disorder in the slab by extended polariton modes which filter out the exciton states from the wings of the Gaussian distribution. Recently, the vertical motional narrowing effect has been observed in multiple-QW structures, where an increase of the decay time of the time-resolved reflection with an increase of the number of QWs has been observed.¹¹ Here we call for a similar experimental study of the motional narrowing effect in a thick semiconductor film, which seems to have a substantial fundamental importance and possible application in ultrafast opto-electronics.

Finally, note the second scale of modulation of the transmitted signal in Fig. 6(a) (the weak minima at 20 ps for the 10- μm -thick slab and at 62 ps for the 150- μm -thick slab). This modulation has the same nature as the oscillations in the time-resolved reflection of the single QW shown in Fig. 5; that is, the interference within a single-polariton branch caused by an inhomogeneous broadening. This effect is absent in the simplified analytical calculation [Fig. 6(b)].

C. Dynamics of the dielectric polarization

The time-dependent excitonic polarization in a 10- μm -thick film of GaAs induced by a 300-fs-long light pulse in the case of inhomogeneous broadening is shown in Fig. 7. We used the same set of excitonic parameters as in Fig. 6. Comparing Fig. 7 with Fig. 2, one can see that both in cases

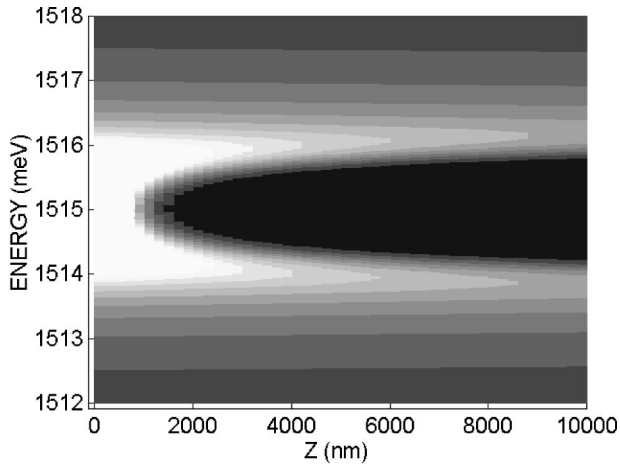


FIG. 8. Coordinate- and frequency-dependent dielectric polarization induced by excitons excited by a 300-fs light pulse in a 10- μm -thick slab of GaAs with the same parameters as in Fig. 6. The brightness of the color is proportional to the absolute value of the dielectric polarization.

of homogeneous and inhomogeneous broadening, a grating of the dielectric polarization forms in the crystal, which then moves toward the beginning of the structure ($z=0$). In the inhomogeneous case, the period of this grating does not change strongly with time and coordinate.

The frequency-dependent excitonic polarization in the same slab is shown in Fig. 8. This figure is to be compared with Fig. 3, which shows the same quantity in the presence of homogeneous broadening only. One can see that in both cases the maximum of the frequency-dependent polarization experiences a bifurcation at some value of z . The splitting between the two resulting peaks is greater in the presence of inhomogeneous broadening, and the line shape of the excitonic polarization is different in the homogeneous and inhomogeneous cases.

V. CONCLUSIONS

Here we have presented an analysis of exciton-polariton propagation in semiconductor slabs in the framework of a simplified semiclassical model, which assumes the slabs to be optically homogeneous even though the exciton resonance is inhomogeneously broadened. The model neglects all scattering processes in the semiconductor, but takes into account the coherent multiple acts of reabsorption–re-emission of light by excitons with different resonance energies, an effect which has a huge influence on the time-resolved optical response of the slabs.

It seems quite instructive to consider the propagation of exciton polaritons as a chain of virtual emission-absorption acts. A photon entering into the structure can excite an exciton state, which can coherently re-emit the photon, which is then absorbed again, etc. Due to the random potential fluctuations, exciton states in semiconductors are characterized by slightly different energies (this is what we call the *inhomogeneous broadening case*). In this case, during its passage through the film a photon may interact with a few different excitons from this distribution. Since different photons interact with different exciton resonances, transmission of light through an optically homogeneous medium with an inho-

geneously broadened exciton resonance presents some analogies with the problem of the electronic transport in mesoscopic structures containing randomly placed elastic scatterers.

Consideration of the exciton-polariton propagation as a problem of mesoscopics assumes a quantum approach emphasizing the quasiparticle nature of polaritons. We speculate on this using results of a semiclassical model, because, in the linear regime, quantum and classical approaches for the light-matter interaction problems are formally equivalent.

In this paper we calculate numerically and describe analytically the appearance of peculiar interference phenomena related to the propagation of exciton-polaritons in semiconductor slabs. Oscillations in the time-resolved spectra of QW's arise due to interference within a *single*-polariton branch induced by the inhomogeneous broadening, which have been recently predicted. Photons absorbed and emitted by different excitons interfere in this case. The phase difference of photons passing through different chains of emission-absorption acts has the same origin as that of electrons having different paths in mesoscopic problems. This effect is only seen in calculations assuming the weighted dielectric susceptibility of the media, and highlights the important role of the interference between photons passing through different chains of emission-absorption acts, a phenomenon completely ignored in the model in which the optical response of the structure is obtained by weighting those related to different exciton resonances.

Both for homogeneously and inhomogeneously broadened excitons, we predict the formation of a grating of dielectric polarization induced by a short light pulse propagating through a semiconductor slab. Interestingly, the grating moves backward with respect to the light-propagation direction.

Striking differences are predicted in the time-resolved transmission spectra of semiconductor films for inhomogeneously and homogeneously broadened excitons. In particular, in the former case the decay rate of the transmission depends strongly on the thickness of the slab. The signal decays with a nonexponential envelope governed mostly by the inhomogeneous broadening parameter Δ in the limit of very thin films. On increasing the slab thickness the decay becomes slower and slower, eventually approaching a value characteristic of the decay in homogeneously broadened media. This behavior—which is a manifestation of the vertical motional narrowing effect—has a simple, intuitive explanation if the propagation of the exciton-polaritons is interpreted in terms of virtual reabsorption and re-emission of light by excitons, and may be summarized as follows: statistically, photons have a larger probability to interact with excitons in the center of the energy distribution. In the limit of an infinite number of emission-absorption acts all exciton resonances but one are filtered out, so that the total decay rate equals the (nonradiative) decay rate of this single exciton resonance.

By use of the steepest-descent method we have derived an analytic formula for the time-resolved transmission, and interpreted the preceding results in terms of classical optics. In this framework inhomogeneous broadening is predicted to influence the dispersion and group velocity of exciton polaritons, as well as their damping rates. Interference of polariton modes from upper and lower branches having the same

group velocities governs the polarization at a given point and at a given time. The dependence of the damping rate on the frequency is responsible for the thickness-dependent decay of the transmitted signal.

We hope that the quasiparticle nature of exciton polaritons and the mesoscopic phenomena (for instance weak localization) in their transport will be revealed experimentally soon. Single- or double-pulse coherent optical spectroscopies with a picosecond resolution are required for these studies. For instance, a dielectric grating excited by a first pulse could be probed by the second pulse, etc. We believe that intriguing fundamental problems of the physics of exciton polaritons will attract enhanced attention from the scientific community.

APPENDIX: CALCULATION OF THE TIME-RESOLVED TRANSMISSION THROUGH A SEMICONDUCTOR SLAB WITH THE STEEPEST-DESCENT METHOD

In the case of distributed resonance frequency the refractive index has a nonvanishing, imaginary part even for $\gamma \rightarrow 0$, which in turn implies that the phase Φ in Eq. (14) has both real and imaginary parts: $\Phi(\omega) = \text{Re } \Phi(\omega) + i \text{Im } \Phi(\omega)$ [with $\text{Im } \Phi(\omega) > 0$]. In order to calculate $G(t)$ [Eq. (14)] with the steepest-descent method, the stationary points should be evaluated at each time t . These points are the solutions of the equation

$$\zeta w(\zeta) = \frac{i}{\sqrt{\pi}} \left[1 + \frac{1}{2} \left(\frac{\Delta}{\Omega} \right)^2 \right], \quad (\text{A1})$$

with $\Omega = \Omega(t) = \omega_c [(d/1)/v/t]^{1/2}$. The stationary points

$$\omega_S = \omega_S(t) = \omega_0 \pm \Omega(t) - i\gamma, \quad (\text{A2})$$

(with the plus and minus signs referring to the upper and to the lower polariton branches, respectively) are correctly obtained in the limiting case $\Delta \rightarrow 0$ from Eq. (A1), as may be easily verified using the asymptotic expansion for $\zeta \rightarrow \infty$ up to third order in $(1/\zeta)$.²⁴

Once the points $\zeta_S = \zeta_R + i\zeta_I$ have been calculated [where ζ_S are the complex solutions of Eq. (A1), and ζ_R and ζ_I are real and imaginary parts of ζ_S , respectively] the response function at a time t may be evaluated. It is given by

$$G(t) = \left(\sqrt{\frac{2}{\pi} \frac{\sin(2\beta)}{\text{Re } \Phi''}} \right)^{1/2} e^{i\Phi_0} e^{-\text{Im } \Phi_S} \cos(\Delta \Phi_S + \beta). \quad (\text{A3})$$

In Eq. (A3) all quantities are meant to be calculated at the stationary points.

Here

$$\Phi_0 = \Phi_0^{(\text{hom})} - \frac{\omega_{LT}}{4} \frac{d}{\nu} \left(\frac{\Delta}{\Omega} \right)^2, \quad (\text{A4})$$

$$\Delta \Phi_S = -\zeta_R \Delta \left(\hat{t} + \left(\frac{\omega_c}{\Delta} \right)^2 \frac{d}{\nu} \frac{1 + \frac{1}{2} \left(\frac{\Delta}{\Omega} \right)^2}{\zeta^2} \right), \quad (\text{A5})$$

$$\begin{aligned} \text{Im } \Phi_S &= \text{Im } \Phi_S^{(\text{hom})} + \text{Im } \Phi_S^{(\text{inh})} \\ &= \gamma \hat{t} - \zeta_I \Delta \left(\hat{t} - \left(\frac{\omega_c}{\Delta} \right)^2 \frac{d}{\nu} \frac{1 + \frac{1}{2} \left(\frac{\Delta}{\Omega} \right)^2}{\zeta^2} \right), \end{aligned} \quad (\text{A6})$$

$$\begin{aligned} \Phi_S'' &= \frac{\partial^2 \Phi}{\partial \omega^2} \\ &= \text{Re } \Phi_S'' + i \text{Im } \Phi_S'' \\ &= 2 \frac{d}{\nu} \frac{\omega_c^2}{\Delta^3} \left\{ \frac{1}{\zeta} \left[1 + \frac{1}{2} \left(\frac{\Delta}{\Omega} \right)^2 \right] - \left(\frac{\Delta}{\Omega} \right)^2 \zeta \right\}, \end{aligned} \quad (\text{A7})$$

$$\beta = \frac{1}{2} \arctan \left[-\frac{\zeta_R \left(\frac{1 + \frac{1}{2} \left(\frac{\Delta}{\Omega} \right)^2}{\zeta^2} - \left(\frac{\Delta}{\Omega} \right)^2 \right)}{\zeta_I \left(\frac{1 + \frac{1}{2} \left(\frac{\Delta}{\Omega} \right)^2}{\zeta^2} + \left(\frac{\Delta}{\Omega} \right)^2 \right)} \right]. \quad (\text{A8})$$

In the *small disorder* situation ($\Delta/\Omega \ll 1$), Eq. (A1) may be solved analytically, as discussed in Sec. IV. The general case with an arbitrary value of the ratio Δ/Ω requires the numerical solution of Eq. (A1); this step, which is very rapidly and easily performed, is the only numerical one necessary in order to calculate the response function (A3).

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