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A scattering-state approach is proposed to study the propagation of extremely short optical pulses through semiconductor heterostructures. The formalism is applied to the propagation of exciton polaritons: Our simulated experiments predict the formation of an exciton-induced polarization grating when the light pulse is resonant with the excitonic transition, and suggest proper physical conditions for its experimental detection. Moreover, our analysis of the polariton transport in thick semiconductor layers reveals a decrease of the average polariton group velocity as a function of time, which we ascribe to a re-emission—reabsorption of light by excitons. [S0163-1829(99)01043-7]

Femtosecond spectroscopy of low-dimensional semiconductor structures has been made possible by rapid development of up-conversion techniques[1] as well as phase-dependent reflectivity experiments.[2,3] These advanced techniques allow us to study the coherent dynamics of electronic excitations in semiconductors and give much insight into the physics of ultrafast optical processes in multilayer structures. In particular, the phase-dependent reflectivity technique allows to study the *linear* optical response of quantum systems to ultrashort pulses of light, which allows to investigate basic properties of single unscreened excitons, including their radiative coupling and exciton-polariton kinetics.[3]

Optical-interference effects appear to be of key importance in a wide class of semiconductor structures including multiple quantum wells and vertical cavity surface-emitting lasers (VCSEL's). A proper account of multiple reflection of laser pulses in light-absorbing or -emitting systems like VCSEL's requires an adequate theoretical approach. In general, time-dependent Maxwell equations must be solved taking into account absorption or emission of light in the system due to electronic transitions. A recently developed quantum-mechanical approach[4] provides a time-dependent solution of a set of coupled Maxwell and semiconductor Bloch equations. This method, while being quite powerful for the analysis of nonlinear problems, has some disadvantages with respect to electrodynamic macroscopic models in the linear-response regime.[5] In particular, the classical theory provides simple analytical expressions for the reflectivity and transmissivity coefficients, which within a fully microscopic approach can only be obtained by tedious numerical calculations.

In this paper we present a semiclassical formalism allowing to model propagation of ultrashort light pulses in multilayer structures accurately taking into account multiple-reflection as well as interference effects. The proposed theoretical approach allows to reduce the problem of ultrafast light-pulse propagation to the evaluation of *scattering states*, i.e., states corresponding to a monochromatic light beam incident on our multilayer structure. In spite of its linear-response character, the method allows to describe in a very simple manner excitonic absorption and re-emission of light in the structure, the so-called exciton-polariton effect. This theoretical scheme is applied to the propagation of exciton polaritons in semiconductor films. We find unexpected aspects of this old problem. In particular, for resonance conditions *we predict the formation of a excitonic polarization grating*, which moves backward with respect to the light propagation. The velocity of this motion is found to decrease with time, which is ascribed to multiple reabsorption/re-emission of light by excitons.

As starting point, let us consider Maxwell's equations describing light incident on a planar multilayer structure. For the simplest case of a TE-polarized wave in a nonmagnetic material ($\mu=1$) we have in the frequency domain

$$\tilde{\mathcal{L}}\tilde{E}(z,\omega)\equiv\left[\Delta+\frac{\omega^2\varepsilon(z,\omega)}{c^2}\right]\tilde{E}(z,\omega)=0, \tag{1}$$

where $\tilde{E}(z,\omega)$ is the Fourier transform of the time-dependent electric field $E(z,t)$, i.e.,

$$E(z,t)=\int d\omega\tilde{E}(z,\omega)e^{-i\omega(t-t_{\bigcirc})}, \tag{2}$$

and the polarization field is expressed in terms of a frequency-dependent dielectric function, i.e., $\tilde{P}(z,\omega)=[\varepsilon(z,\omega)-1]\tilde{E}(z,\omega)$.

The stationary solutions $\tilde{E}$ describe the spatial propagation through our multilayer structure of a monochromatic light beam with frequency ω. A part from their amplitude,

such stationary solutions can be regarded as *scattering states of light*. More specifically, we can write

$$\tilde{E}(z,\omega)=c(\omega)s(z,\omega), \tag{3}$$

where $s(z,\omega)$ are the scattering states of the linear operator $\tilde{\mathcal{L}}$, i.e., solutions of Eq. (1) with $s(z=0,\omega)=1+r(\omega)$, where $r(\omega)$ is the amplitude reflection coefficient of the structure, and $c(\omega)$ are arbitrary Fourier coefficients, whose value will be dictated by the initial condition $E(z,t_0)=E_0(z)$.

The scattering states s can be readily found by applying a transfer-matrix method[6] to our electromagnetic problem. 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 at the end of the layer. The total transfer matrix—relative to the entire structure—is obtained as the product of the transfer matrices corresponding to the individual layers: $T=\Pi_{j=N}^{j=0}T_j$, where the layers are numbered starting from $z=0$ till the end of the structure.

The reflection coefficient r of the structure can be expressed as a function of the elements T_{mn} of the matrix T according to

$$r(\omega)=\frac{n_i(T_{11}+T_{12}n_f)-(T_{21}+T_{22}n_f)}{n_i(T_{11}+T_{12}n_f)+(T_{21}+T_{22}n_f)}. \tag{4}$$

Here, n_f is the refraction index in the last (semi-infinite) layer of the structure. The scattering states s are then given by

$$s(z,\omega)=T_{11}^z[1+r(\omega)]+T_{12}^z[1-n_i r(\omega)], \tag{5}$$

where T^z is the transfer matrix from 0 to z.

Given the above scattering states s, by inserting Eq. (3) into Eq. (2) we finally obtain the desired space and time evolution of our light pulse

$$E(z,t)=\int d\omega c(\omega)s(z,\omega)e^{-i\omega(t-t_\bigcirc)}, \tag{6}$$

where the Fourier coefficient is given by the scalar product between the electromagnetic-field distribution at the initial time $t_\bigcirc$ and the corresponding scattering state

$$c(\omega)=\int dz s^*(z,\omega)E_\bigcirc(z). \tag{7}$$

The above expression follows directly from the orthonormality and completeness of our scattering-state set.[7] We stress that for the case of an initial light pulse located far from the structure—the one considered in our simulated experiments—the scattering state s in Eq. (7) can be replaced by its incident part: $s(z,\omega)\propto\exp(in_i\omega/cz)$ and the coefficient $c(\omega)$ reduces to the spatial Fourier transform of the initial condition $E_\bigcirc$: $c(\omega)\propto\tilde{E}_\bigcirc(k)=\tilde{E}_\bigcirc(n_i\omega/c)$. The difference of our approach from that of Jahnke *et al.*[4] formulated for multiple quantum well structures and microcavities is that we do not solve directly the time-dependent Maxwell equations (which is quite hard in many cases) but we operate with the scattering states of the system.

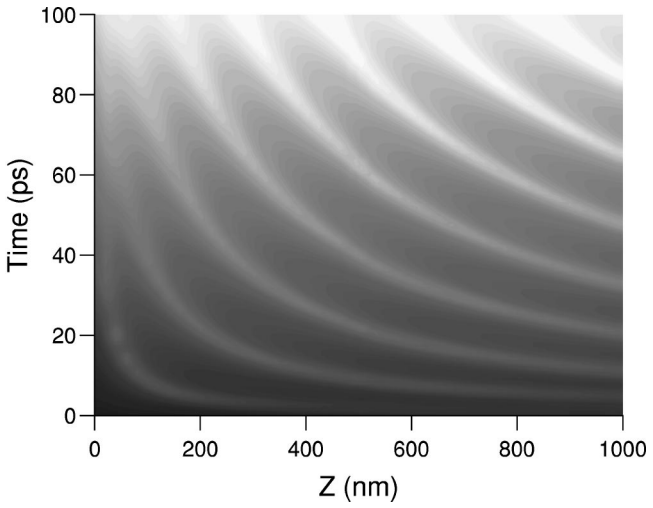

FIG. 1. Evolution of the excitonic polarization induced by a 150 fs-long Gaussian pulse in a 1000-nm-thick layer of GaAs with the following excitonic parameters: $\hbar\omega_0=1.515$ eV, $\hbar\omega_{LT}=0.08$ meV, and $\hbar\Gamma=0.1$ meV. The pulse is centered at the exciton resonance frequency. Darkness is proportional to the logarithm of the excitonic polarization.

The above theoretical scheme has been applied to the propagation of exciton-polaritons in semiconductors. This has been a subject of hundreds of works during recent 40 years starting from the pioneering papers by Pekar, Hopfield, and Agranovich.[8] The semiclassical approach to this problem consists in the introduction of the frequency-dependent complex dielectric constant

$$\varepsilon(\omega)=\varepsilon_B\left[1+\frac{\omega_{LT}}{\omega_0-\omega-i\Gamma}\right], \tag{8}$$

where ε_B is the background dielectric constant, ω_0 is the exciton resonance frequency, ω_{LT} is the longitudinal-transverse splitting, and Γ is the damping. Here, the spatial dispersion has been neglected for simplicity. The exciton-polariton wave vector K is connected with ε by the relation $\varepsilon(\omega)=\omega^2/K^2c^2$, while the corresponding group velocity is given by $v(\omega)=\partial\omega/\partial K$. Two polariton branches are usually supposed to produce two waves of polarization with different wave vectors, which may interfere giving rise to quantum beats in the time-resolved transmission spectra.[9,10] Actually, for a short light pulse containing all frequencies around the exciton resonance, the polariton group velocity cannot be defined in a simple way. In this respect two basic questions need to be answered: *What is the initial spatial distribution of this polarization*? *How it changes with time due to coherent emission and reabsorption of light by excitons*?

We have addressed these questions by performing a simulation of the exciton-polariton dynamics in a semiconductor film using our scattering-state formalism for the frequency-dependent dielectric constant (8).

Figure 1 shows the excitonic polarization in a 1000-nm-thick film of GaAs excited by a short light pulse, as a function of space and time. Here, the central energy of the pulse is equal to the exciton resonance energy (1.515 eV) while its spectral width is 150 fs. Moreover, we have taken $\hbar\omega_{LT}=0.08$ meV and $\hbar\Gamma=0.1$ meV. The excitonic polarization

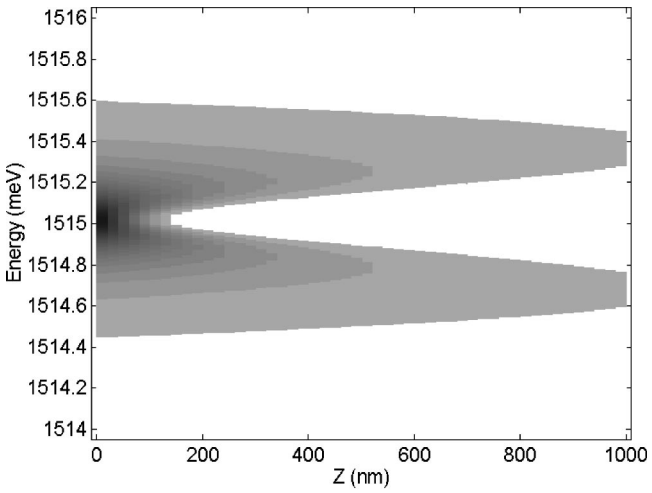

FIG. 2. Space- and frequency-dependent excitonic polarization induced in a 1000-nm-thick layer of GaAs at the same conditions as in Fig. 1.

has been computed as the product of the electric field times the resonant term in the dielectric function. In order to exclude any optical interference effect due to reflection from the surfaces we have chosen in the media before and after the GaAs film the same background dielectric constants as in GaAs ($\varepsilon_B = 13$). Thus our results must be also valid for a bulk. Note the picosecond time scale, in contrast to the femtosecond time-scale corresponding to the light propagation time through the film. This means that on this time scale the contribution of the fast photonic components of the pulse to the dielectric polarization is negligibly small. The light pulse has quickly passed through the crystal, and the light was partially absorbed in the crystal by excitons, which we have supposed to be infinitely heavy and unable to move. For these conditions, the observed motion of the excitonic polarizations in the crystal is only due to emission and reabsorption of light.

We clearly see a periodic dielectric grating in the crystal, which moves backward with respect to the light propagation direction, first quickly then slower and slower. Appearance of the grating in a homogeneous media after a single-pulse excitation seems to be a striking effect. In order to understand the nature of this grating we have plotted in Fig. 2 the excitonic polarization in the same structure as a function of frequency and coordinate. One can see that an initial single-resonance distribution of the dielectric polarization at the exciton frequency drastically changes as one goes deeper into the semiconductor media. A pronounced camel-back structure appears, with two maxima split by almost 1 meV in the end of the structure. Clearly, the beats in the time dynamics of the polarization are due to a double-resonance structure in its frequency-resolved spectra. This structure is typical for a regime of strong coupling between excitons and photons. The new propagation eigenstates, i.e. exciton-polaritons, are split proportionally to the coupling between an exciton and a photon. This splitting of the maxima of the polarization is roughly proportional to the number of absorption and re-emission processes of a photon by an exciton during the propagation of the polariton mode, and thus it increases with the coordinate. The period of the beats in time is inversely proportional to the splitting between the two eigenstates of the system as follows from the Heisenberg relations. This explains the decrease of the oscillation period with the coordinate seen in Fig. 1.

Further interpretation of the temporary behavior of the excitonic polarization grating requires a more detailed analysis of the evolution of polariton packets. Let us consider a single maximum in the excitonic polarization. As stressed before, the polarization changes in time are only due to coherent emission and reabsorption of light by excitons. The width of the excitonic spectral line in absorption is proportional to Γ, which has been chosen to be much smaller than the spectral width of the pulse. Due to multiple absorption, the spectral function of the pulse gets narrower and tends to the form of the exciton line in absorption (see Fig. 2). This is consistent with the observed broadening of the maxima of the excitonic-polarization in time and space during their motion (Fig. 1). The average group velocity tends in this case to the exciton velocity, which is always very small and is equal to zero in our model.

This effect can also be understood in terms of the exciton-photon coupling strength. Actually, the wings of the pulse are weakly coupled to the exciton than the central part of the pulse tuned at the resonance. The polaritons created by these weakly coupled photons are fast and arrive first at the given point of the slab. The interference between these ''fast'' polaritons creates the short oscillations at short times. The long-period oscillations are created by the central part of the pulse strongly coupled with the exciton, which propagates slowly compared to the photonlike polaritons. The interference processes within this type of polaritons play a major role at longer times thus inducing an increase of the period of the beats with time.

Quantitatively, these effects can be described either in the framework of classical optics or in terms of the quantum interference. From the classical point of view, the appearance of a camel-back structure in the frequency-dependent dielectric polarization results from the interplay between optical absorption and transmission. Actually, the excitonic polarization at a given point Z is proportional to the absorption coefficient $A(\omega)$ (having the same Lorentzian shape in any point of the structure) times the amplitude of light transmitted to the point Z. This latter has a minimum at the exciton resonance frequency due to absorption. The product of these two terms behaves as

$$P(\omega,Z) \propto A(\omega)[1 - f(Z)A(\omega)], \tag{9}$$

where $f(Z)$ is a monotonic function which increases with Z. Clearly, the function $P(\omega,Z)$ experiences bifurcation at certain values of Z. Then, two maxima appear, whose splitting increases as a function of Z.

In terms of quantum interference, one should consider exciton polaritons in the structure as quasiparticles described by certain wave packets. The dielectric polarization at point Z and time t within the crystal, is governed by wave packet components that have a group velocity $V = Z/t$. Evidently, there are always two frequencies that correspond to the same group velocity, one from the upper polariton branch and another from the lower one. Thus, the polarization at a given point and time is always the result of interference between two waves, while the phase difference between such two

waves is a function of Z and t. This consideration allows to apply to our problem the stationary phase method developed by Andreani and Panzarini[10] for the explanation of the beats in time-resolved excitonic transmission through a semiconductor slab. Using the same arguments, we obtain an approximate expression for the period of the beats

$$T=\pi\left[\frac{2ct}{Z\sqrt{\varepsilon_B}\omega_0\omega_{LT}}\right]^{1/2}. \tag{10}$$

This appears to be in excellent agreement with the numerical results reported in Fig. 1 at the values of Z exceeding the critical value where the bifurcation of $P(\omega,Z)$ takes place. Actually, the beating period decreases as the square root of Z and increases as the square root of t. To summarize, *the grating arises and evolves due to the interference between two exciton-polariton branches*.

Interestingly, the dielectric grating created in the crystal by a short light pulse moves backward with respect to the propagation direction. This does not violate momentum conservation because such motion is not a motion of *mechanical excitons* (which are infinitely heavy in our model). The visible motion is completely a result of reabsorption of the light re-emitted by excitons. Due to the interference between two polariton branches, this reabsorption is a periodical function of the coordinate, drifting with time towards the beginning of the structure. Formally, this follows from the analysis by the stationary phase method and Eq. (10). Taking into account the finite mass of excitons this drift velocity is only slightly reduced.

We finally stress that two-pulse coherent experiments may be able to directly probe the presence of the dielectric (i.e., polarization) grating predicted by our analysis, by looking, e.g., to self-diffraction phenomena.

In conclusion, we have proposed a semiclassical theoretical approach for the study of the ultrafast dynamics of short light pulses in multilayer semiconductor structures. The method allows to reduce the complexity of a full space and time propagation to a scattering-state problem. Within our approach the coupling of light to the electronic system is taken into account within a standard local-dielectric-response model.

Quite peculiar and unexpected results have been obtained from the simulation of the ultrafast dynamics of exciton-polariton modes generated by short light pulses in thick semiconductor layers. When the optical pulse is energetically resonant with the excitonic transition, the induced polarization becomes an oscillating function of the coordinate. Peaks of the dielectric polarization formed in this way experience a drift backwards with respect to the light propagation direction. The velocity of this drift decreases as the time increases. Such features have been explained in terms of optical interference between two polariton branches and multiple reabsorption and re-emission of photons by excitons. We believe that the ultrafast coherent spectroscopy of semiconductor films will confirm our prediction of a dielectric grating induced by exciton-polariton effects.

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