Light Propagation and Disorder Effects in Semiconductor Multiple Quantum Wells

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Propagation of femtosecond light pulses in semiconductor multiple quantum wells with interface roughness is investigated theoretically and experimentally. It is shown that the nonlocal susceptibility causes exciton broadening, decreases the relative magnitude of the 1*s* and 2*s* resonances, and leads to multiple reflections. The time resolved signal exhibits modulations that are sensitive to the barrier thickness. The absorption spectrum and time dependence of the beat frequency are influenced by static disorder.

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Propagation of light pulses in semiconductor heterostructures, in particular, multiple quantum well structures (MQWS), is a problem concerning a wide range of applications in modern optics. Examples are light transmission in waveguides and semiconductor lasers, or any femtosecond optical experiment investigating the optical response of quantum confined systems. In this Letter, we theoretically and experimentally investigate propagation in the growth direction of a semiconductor multilayered structure. The results show that a complete description of propagation in a heterostructure must simultaneously treat the effects of the nonlocal susceptibility, Coulomb interaction, and disorder. The experimental observation cannot be theoretically reproduced if the different effects are treated separately.

In optically thick bulk semiconductors, polariton effects lead to slow propagation [1], beating [2], and pulse breakup [3–6]. In heterostructures, the transmission of ultrashort light pulses is sensitive to not only the interactions between generated electronic excitations but also to sample geometry, including structural disorder (e.g., well width fluctuations).

Propagation in resonant dispersive media is typically described using the slowly varying envelope approximation (SVEA). While this approach is justified in bulk semiconductors where the dielectric function is spatially homogeneous, in semiconductor heterostructures, which have a nonlocal susceptibility, the SVEA does not hold because the spatial dependence of the polarization is determined by the geometry of the heterostructure and not by the spatial dependence of the exciting light field. The lack of translational invariance in the growth direction means that momentum is not conserved in this direction. This leads to a finite density of states for excitonic radiative decay [7,8], thus increasing the exciton linewidth. Furthermore, partial reflections play an important role [9,10]. We show that, within linear response theory, the problem of pulse propagation in an idealized semiconductor multiple quantum well structure can be solved analytically and thus provides a good example for investigating the effects of nonlocality.

Realistic semiconductor heterostructures are generally characterized by a certain amount of intrinsic disorder, caused by imperfections of the interface separating the epitaxial layers. We therefore include weak static disorder in our analysis. The coupling of the relative and center of mass (c.m.) coordinates by the disorder potential is treated using Green's function theory, allowing the inclusion of the Coulomb interaction and static disorder on an equal footing. Our calculations reveal characteristic features in the transmitted pulse shape, which are also observed in our femtosecond pulse propagation experiments in a InGaAs/GaAs MQWS.

For our calculations we consider a multilayer structure consisting of N wells of average thickness L and $N + 1$ barriers of thickness $D - L$. If z is the direction of growth, the incident laser pulse is described classically by a linearly polarized plane wave traveling in the positive z direction with a frequency dependent amplitude $E_0(\omega)$. For technical simplicity we assume perfect quantum confinement conditions (infinitely high potential barriers) so that the optical excitation, i.e., the interband polarization P is completely localized within the wells. We can then characterize each quantum well by a transmission coefficient T, a reflection coefficient R, and an absorption coefficient $\alpha = (1 - |T|^2 - |R|^2)/2$.

To compute T and R first for a single quantum well, we write the solutions of Maxwell's equation for the barriers as

$$E(z,\omega) = \begin{cases} E_0(\omega)[e^{ik_0 z} + R(\omega)e^{-ik_0 z}] & z < -L/2\,, \quad (1a) \\ E_0(\omega)T(\omega)e^{ik_0 z} & z > L/2\,, \quad (1b) \end{cases}$$

where k_0 is the wave vector of the incident light field in the barrier. Then, within the quantum well, we expand the polarization in terms of the confinement wave functions. For sufficiently thin quantum wells this expansion can be restricted to the ground state confinement wave functions [11,12] $P(z,\omega) = P^{2D}(\omega)\cos^2(\pi z/L)$, where $P^{2D}(\omega)$ has to be obtained as the solution of the two-dimensional electron-hole problem. As a consequence of the simple analytical z dependence of $P(z,\omega)$, Maxwell's wave equation can be integrated analytically. The two frequency

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dependent integration constants are determined assuming continuity of the field amplitude and its first derivative at the interfaces. We obtain

$$E(z,\omega) = E_0(\omega)e^{ik_0z} + \frac{4\pi}{L}P^{2D}(\omega)f(z,\omega), \qquad (2a)$$

$$f(z,\omega) = [e^{ik_0L/2}\cos(k_0z) - 1] - \left[1 - \left(\frac{2\pi}{k_0L}\right)^2\right]^{-1}[\cos(k_0z) + \cos(2\pi z/L)] \qquad (2b)$$

for the electric field within the quantum well and

$$E_0(\omega)R(\omega) = \frac{4i}{L}\,dP^{2D}(\omega)\sin(k_0L/2), \quad T(\omega) = 1 + R(\omega) \qquad (3)$$

for the reflectivity and transmitivity of the well [13], with $d = \pi\{1 - [1 - (2\pi/k_0L)^2]^{-1}\}$. Hence, the absorption coefficient $\alpha = -\mathrm{Re}(RT)$, showing that finite absorption in a nonlocal situation requires nonvanishing reflection and transmission.

So far our results are valid for arbitrary 2D polarization. However, for the purposes of this Letter, we restrict the analysis to the linear regime, where we can write

$$P^{2D}(\omega) = \int_{-L/2}^{L/2} dz\,dz'\chi(\omega,z,z')E(\omega,z)\delta(z-z'), \qquad (4)$$

with

$$\chi(\omega,z,z') = -\frac{2}{L}|\mu|^2\cos(\pi z/L)\cos(\pi z'/L)\frac{1}{V} \times \int d^2R\,d^2R'\langle\mathbf{R},\mathbf{r}=0|(\omega + i\delta - \mathcal{H}_{ex})^{-1}|\mathbf{R}',\mathbf{r}'=0\rangle, \qquad (5)$$

and $E(z,\omega)$ is the solution of Eq. (2b). In Eq. (5), μ is the dipole moment, $\mathbf{R}$ and $\mathbf{r}$ are the in-plane c.m. and relative coordinates of the electron hole pair.

In the presence of interface roughness the exciton Hamiltonian $\mathcal{H}_{ex}$ contains the center of mass kinetic energy, the excitonic part, and the disorder potential, i.e., the fluctuation of the confinement energy due to interface roughness. This random potential can be characterized by its correlation function only, the average value of the disorder potential is taken as zero. To account for the Coulomb interaction, which we have taken purely two dimensionally, we expand the susceptibility in terms of the excitonic wave functions. Because the spot size is usually large compared to the correlation length of the disorder potential, all observed quantities are a configuration average over many independent regions. It is therefore sufficient to calculate the configuration average of the linear susceptibility. For this purpose we apply the self-consistent Born approximation, assuming a short-ranged correlation function for the well width fluctuation $\langle\Delta L(\mathbf{R})\Delta L(\mathbf{R}')\rangle_{\mathrm{conf}} = 4A\sigma^2L^6/\pi^4\delta(\mathbf{R}-\mathbf{R}')$. Here, $\sigma = \pi^2\Delta L/2L^3$ is the standard deviation of disorder potential and A is the mean cross section of the independent regions. In our numerical evaluation we include four bound excitonic states. Since the relevant disorder parameter for the c.m. particle scales with the spatial extension of the excitonic states, the effects of disorder on the continuum states can be neglected.

Without disorder the exciton line shape is a Lorentzian with width $\Gamma = \delta + \Gamma_r$, where δ is due to "pure dephasing" processes such as scattering with other quasiparticles and Γ_r is the radiative linewidth. It is interesting to note that the radiative linewidth ("natural linewidth") in a semiconductor heterostructure depends on the magnitude of the in-plane momentum of the exciton. For bulk or surface polaritons, only scattering at the crystal edges leads to a radiative decay. However, perpendicular to the layers of a heterostructure, momentum conservation does not apply and there is a finite density of states for excitons with $\mathbf{K}_\parallel = 0$ to emit photons and decay into the crystal ground state. This leads to an intrinsic broadening of the excitonic resonances [7,8].

For $k_0\chi \ll 1$ one can expand the solution $T \approx 1 + 2\pi ik_0\chi + O((k_0\chi)^2) \approx \exp(2\pi ik_0\chi)$ and $\alpha = 2\pi k_0\mathrm{Im}(\chi) + O((k_0\chi)^2)$. Hence, in first order the absorption reduces to $-\ln(|T|)$.

Because of multiple reflections, the transmission coefficient T_N of a structure with N quantum wells deviates from a simple power law $T_N \neq T^N$, since the radiative coupling of the quantum wells is sensitive to the sample period. Hence, we have to use a transfer matrix to compute R and T for a MQW from the single-well results. The problem becomes translationally invariant on the length scale of the wavelength only if $k_0\chi \ll 1$ *and* $\exp(ik_0D) \approx 1$. Then the transmitted pulse can be described by an effective k vector corresponding exactly to the solution obtained within the SVEA, where the medium is replaced by an effective homogeneous medium with an average susceptibility $\chi_{\mathrm{eff}}(\omega) = \chi(\omega)/D$. In this limit, the effects of propagation effects are essentially the same as in 3D [2]. Generally, however, one has to take the strong frequency dependent reflection into account for the proper analysis of MQW transmission experiments.

As an illustration of our results, we show in Fig. 1 the transmitted intensity $|T|^2$ and the absorbed intensity $|A|^2 = 2\alpha = 1 - |T|^2 - |R|^2$ as a function of frequency (normalized to the bulk exciton binding energy) calculated for a single 10 nm quantum well without disorder, using $\delta = 0.2$ meV [14]. On resonance, a large percentage of the intensity is reflected, as can be seen from Fig. 1, where the reflected intensity is the difference between the two curves. From the width of the absorption spectrum, we see that the effective radiative linewidth of the exciton is approximately 0.7 meV. Because the oscillator strength in a real quantum well is less than in our 2D model, this is a upper limit to the excitonic radiative linewidth.

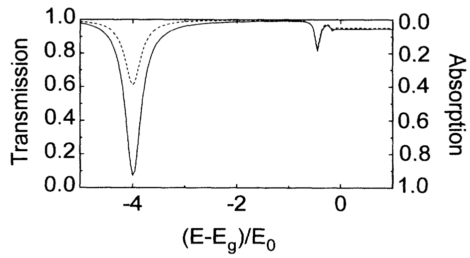

FIG. 1. Transmitted intensity (solid line) and absorbed intensity (dashed line) for a single GaAs quantum well without disorder.

Although measuring the transmission and reflection is experimentally simple, it is very difficult to unravel the relative linewidth contributions due to propagation from those due to disorder. We therefore studied pulse propagation in a MQW, providing a clear separation of the two effects. The sample is a 20 period $In_{0.08}Ga_{0.92}As/GaAs$ quantum well [15]. The sample has 40 nm barriers and the 10 nm InGaAs wells. As the light-hole transitions are blueshifted by 30 meV due to built-in strain from the lattice constant mismatch, the heavy-hole states are isolated. At 5 K the linear absorption spectrum (inset Fig. 2) displays (i) a strong heavy-hole (hh) exciton resonance at 1.455 eV with a 1.7 meV linewidth and (ii) absorption due to heavy-hole–free-electron transitions above 1.462 eV. The homogeneous linewidth, obtained from transient four wave mixing (TFWM) experiments, is 0.2 meV. Additionally, time resolved TFWM verifies that the sample is inhomogeneously broadened [16].

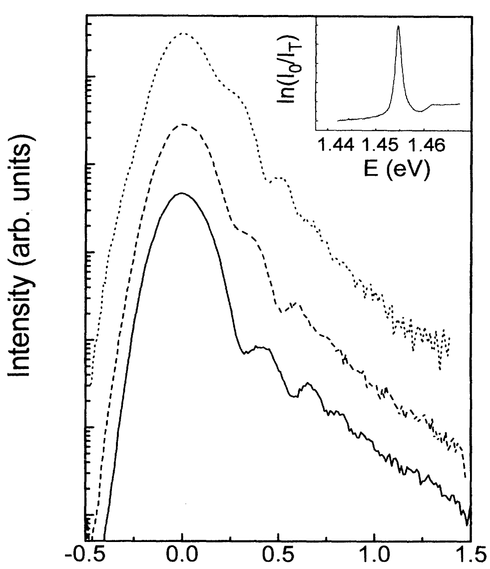

FIG. 2. Transmitted pulse measured in a 20 layer $In_{0.08}Ga_{0.92}As/GaAs$ MQWS at a detuning of (i) 11.4 meV (solid line) and (ii) 4.5 meV (dashed line) below the 1*s*-hh resonance and (iii) +1.5 meV below the band gap (dotted line). Inset: Measured transmission spectrum.

The binding energy of 7 meV is less than the 2D value (16.8 meV) because of the finite extent of the wave function. Hence, comparison between theory and experiment must be made carefully. Agreement can only be expected if the overlap of the exciting light pulse with the absorption spectrum is qualitatively comparable.

The 110–120 fs [full width at half maximum (FWHM)] incident pulses are nearly transform limited, with a spectral width of 20–22 meV. (We have verified that the slight chirp does not qualitatively change the results.) The measurements are performed in the low-density limit with a 100 μm spot (much larger than the disorder correlation length) and the transmitted pulse is time resolved via cross correlation with a reference pulse in a second harmonic crystal. These measurements are similar to earlier ones where the density dependence was examined [17]. In Fig. 2 the experimental results for several detunings between the center of the pulse spectrum and the 1*s* exciton resonance are shown. At all detunings the pulse exhibits a long modulated tail and the modulation period is quite small.

For comparison with the experimental results we show in Fig. 3 the calculated cross correlation of the transmitted pulse for various different detunings between the central frequency and the 1*s*-hh-exciton resonance in a GaAs MQWS with 20 layers without disorder (left) and with weak disorder (right). Here, the amplitude of the transmitted pulse has been obtained as

$$E(z,t) = \int \frac{d\omega}{2\pi} e^{-i(\omega t - k_0 z)} T_N(\omega) E_0(\omega)$$

for $z \geq ND$, where $E_0(\omega)$ is the initial pulse, for which we take a 100-fs (FWHM) Gaussian line shape. The insets show the calculated transmission spectra per quantum well, $\ln[I_0(\omega)/I_T(\omega)]/ND$. The excitonic resonances

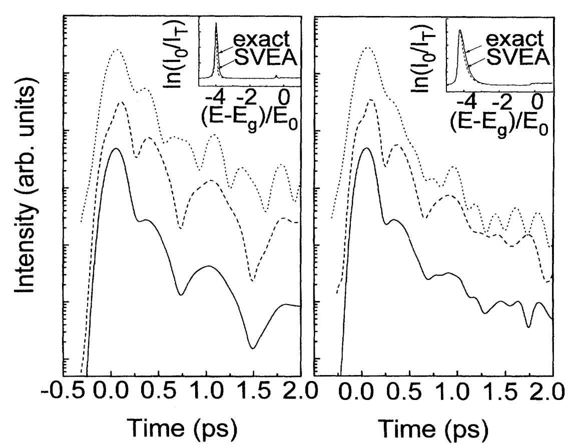

FIG. 3. Time resolved transmitted pulse in a 20 layer GaAs MQWS at a detuning of (i) $3E_0^B$ below the 1*s*-hh-exciton resonance (solid line), (ii) $1E_0^B$ below resonance (dashed line), and (iii) $0.5E_0^B$ below the band gap (dotted line) Left: Without disorder; right: With a weak disorder. Insets: Transmission spectra.

exhibit an additional broadening compared to the single quantum well spectrum. This additional broadening is a Fabry-Pérot effect due to the distributed feedback in the structure, whereas, as mentioned earlier, the broadening of the single quantum well spectrum is an intrinsic feature due to the breaking of translational invariance. Note that without disorder the relative magnitudes between the $1s$ and $2s$ resonances are much smaller than 27, as would be predicted within the SVEA. The $1s$ excitonic resonance has an asymmetric line shape, due to propagation effects. In the presence of disorder, the $1s$ resonance is slightly redshifted and has an enhanced asymmetry; the $2s$ resonance cannot be resolved. The asymmetric line shape is also observed experimentally.

The time resolved transmitted signal exhibits a long modulated tail even if excitation occurs far below resonance. If the pulse center frequency is above the $1s$ resonance, the interference of the $1s$ exciton and the exciton continuum leads to a fast modulation of the transmission tail. If excited below resonance, the period is much longer. Since at this central frequency the pulse has a negligible overlap with the continuum, the beat frequency depends strongly on detuning as observed experimentally. In the absence of disorder, the beat period increases with increasing time as $\sqrt{t}$, as is the case in a bulk crystal with a single excitonic resonance [2]. At all detunings, the modulation frequency is significantly increased by static disorder and increases even with increasing time, as is observed experimentally. In both cases, the modulation period depends on the barrier thickness due to multiple reflections. Thus, static disorder and propagation effects lead to qualitative modifications of the time resolved signal that are apparent in the experimental result as a rapid beating. These modifications cannot be derived if Coulomb, disorder, or propagation effects are treated seperately.

In summary, our experimental and theoretical investigations clearly show that both the spatially inhomogeneous dielectric function and static disorder modify the absorption spectrum of a semiconductor heterostructure and lead to characteristic features of the transmitted femtosecond pulses. The lack of translational invariance and static disorder cause a broadening of excitonic resonances, and reflections at the interfaces lead to interference effects that strongly modify the transmitted pulse shape. Since all effects act simultaneously, a correct interpretation of the experimental results requires an analysis, taking into account the full geometry of the sample, including interface roughness. Although we have only investigated pulse propagation within linear response theory, it is clear that in the nonlinear regime, the influence of a nonlocal susceptibility and disorder is also important.

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