How fast is excitonic electroabsorption?

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Many semiconductor light modulators rely on changes in excitonic absorption induced by electric fields. We study their temporal response in the framework of a one-dimensional model, for which we solve exactly the time-dependent Schrödinger equation. For a homogeneously broadened system, the electroabsorption response time is found to be simply the inverse of the (field-induced) exciton linewidth, which can be as short as 50 fsec.

Recently, nonlinear-optical and optoelectronic effects in semiconductor quantum wells (QW's) have attracted much attention because of their potential applications in optical information processing and, from a more fundamental point of view, because of specific features that distinguish them from similar effects in bulk materials.[1] Although discovered and understood only recently,[2] QW electroabsorption is already utilized in light-modulation devices operating at speeds as fast as 5 GHz,[3,4] and extensions to the terahertz domain are currently being explored. For example, subpicosecond excitonic electroabsorption has been observed in test QW structures,[5] mechanisms for generation of terahertz electrical transients by virtual excitons in biased QW's have been proposed,[6,7] and generation, propagation, and detection of terahertz electrical transients in coplanar strip lines on free-standing micrometer-thick QW films have been demonstrated.[8] In this context, it is important to understand the fundamental limits on the response time of semiconductor electroabsorption and whether these limits are already approached in recent experiments.

Several natural time scales involved in excitonic transitions in applied electric fields are worth noting. For excitons in a static electric field F, a characteristic time scale is the tunneling time, i.e., the time it takes the electron to tunnel out of the attractive Coulomb well owing to the hole. The width of the corresponding tunnel barrier is approximately given by $l \sim R/(eF)$, where R is the exciton binding energy. Together with the electron orbital velocity, $v \sim (R/m)^{1/2}$, this yields a tunneling time $\tau_{\text{tunnel}} \sim (mR)^{1/2}/(eF)$, where m is the reduced electron–hole (e–h) mass (in this Letter we use $\hbar = 1$). Only if the changes in the electric field $F(t)$ occur on a time scale slow compared with τ_{tunnel} can the system respond adiabatically. In the opposite limit of fast changes in $F(t)$, the problem is best thought of in terms of multiphoton transitions. These two limiting cases are well established in the photoionization of atoms.[9,10] Another characteristic time scale is, of course, the electron orbit time R^{-1} itself. Finally, as far as optical properties are concerned, we have to consider the inverse linewidth as a possible limitation on the response time. In the case of (biased) room-temperature GaAs QW's, all these scales are of the order of 50 to 500 fsec, within the time resolution of current experiments and significantly longer than the shortest optical pulses that have been generated (<10 fsec). Hence there is a need for theoretical study of the dynamics of excitonic electroabsorption in an idealized case.

A realistic analysis must be nonperturbative both in the e–h interaction and in $F(t)$ because optical transitions near the band edge of semiconductors are dominated by e–h correlation and because significant electroabsorption usually involves fields of the order of or larger than the ionization field. Furthermore, the full time dependence of the problem must be considered. It is unrealistic to carry out such an analysis for a Hamiltonian describing semiconductors in their full complexity. We can, however, consider a model Hamiltonian that contains all the important physics of excitonic electroabsorption and yet is simple enough to be solved exactly. In this Letter we investigate such a model in one dimension. We note that simple one-dimensional models have been applied successfully to related problems in photoionization of atoms.[10–12] Our model is the same as that of Refs. 10 and 11 but is considerably simpler than that of Ref. 12.

To address directly the intrinsic speed of response of excitonic electroabsorption, we consider an *idealized electroabsorption experiment* in which an electric field F applied to a sample is abruptly switched off at time $t = 0$, i.e., $F(t) = F\Theta(-t)$, where $\Theta(t)$ is the Heaviside step function, and the transmission of an ideal broadband probe pulse, $E(t) = E\delta(t - T)$, delayed by T from time $t = 0$, is measured by a slow, time-integrating detector placed behind an optically dispersive element. Experimentally this might correspond to a situation in which an electric field is shorted out by an e–h gas created optically by an ultrashort pump pulse.[5,8] The absorption and refraction of the material are described through the effective susceptibility $\chi(\omega) = P(\omega)/E(\omega)$, where $P(\omega)$ is the induced polarization at frequency ω. As usual, this susceptibility can be related by Fourier transformation to a linear response function, $\chi(t, t')$, which determines the polarization at time t in response to the optical electric

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field at time t'. Hence

$$\chi(\omega) = \int_0^\infty dt\, \exp(i\omega t)\chi(t + T, T). \quad (1)$$

Note that $\chi(t, t')$ is nonlocal (in time); it cannot be written as $\chi(t - t')$ because the absolute time matters.

As is well known from Elliott's theory,[13] the strength of optical transitions is determined by the probability of finding an electron and a hole in the same unit cell. In one dimension, $\chi(t, t') = \chi(x = 0; t, t')$, where x is the e–h relative coordinate. $\chi(x; t, t')$ satisfies the inhomogeneous Wannier–Schrödinger equation

$$\left[i\frac{\partial}{\partial t} + i\gamma + \frac{1}{2m}\frac{\partial^2}{\partial x^2} + V(x) - eF(t)x\right]\chi(x; t, t') = -\mu^2\delta(x)\delta(t - t'), \quad (2)$$

where μ is the interband dipole matrix element, $V(x)$ is the e–h Coulomb interaction potential, and we have included a phenomenological exciton linewidth γ. In what follows, we solve for $\chi(x; t, t')$ and, where appropriate, take the value at $x = 0$. Note immediately from Eq. (2) that, if the probe pulse $E(t)$ arrives after the time $t = 0$ when $F(t)$ is set to zero (i.e., if $t' = T > 0$), for all times of interest $\chi(x; t + T, T)$ is independent of T and, consequently, the resulting absorption spectrum is always the same. Hence, although the field abruptly drops to zero at delay time $T = 0$, the measured absorption spectrum will show changes only for small *negative* time delays for which the probe pulse has already passed through the sample. In this case, the rapid electric-field transient continues to interact with the decaying polarization that is left by the probe pulse for (negative) time delays of the order of the inverse of the linewidth. This result is consistent with other models of transient absorption spectroscopy involving two-level systems that explain well the oscillatory differential signals obtained for negative time delays in many experiments.[14]

To treat the e–h correlation exactly, we choose a simplified form for the e–h interaction, namely, $V(x) = V\delta(x)$.[10,11] This retains the most important excitonic feature, namely, a single bound state with binding energy $R = (mV^2)/2$ and Bohr radius $a = 1/(mV)$. In what follows, we measure energies (times) in units of R (R^{-1}), χ is measured in units of μ^2/V, and $F(t)$ is measured in units of the ionization field $F(t) = [f(t)R]/(ea)$. In these units, Eq. (2) reads

$$\left[i\frac{\partial}{\partial t} + i\gamma + \frac{\partial^2}{\partial x^2} + 2\delta(x) - f(t)x\right]\chi(x; t, t') = -2\delta(x)\delta(t - t'). \quad (3)$$

To solve Eq. (3), we introduce the free e–h pair susceptibility (i.e., the susceptibility for $V = 0$), $\chi_{e\text{-}h}(x; t, t')$, in the presence of the electric field $f(t)$. This enables us to perform a standard Green's function manipulation; we formally introduce the e–h interaction as a perturbation and then sum the perturbation series to infinite order. The resulting expression, which is exact (i.e., it is not a perturbation approximation), reduces for $x = 0$ to

$$\chi(t, t') = \chi_{e\text{-}h}(t, t') + \int_{t'}^{t} dt''\chi_{e\text{-}h}(t, t'')\chi(t'', t'). \quad (4)$$

Here the free e–h pair susceptibility, $\chi_{e\text{-}h}(t, t') = \chi_{e\text{-}h}(x = 0; t, t')$, has the exact analytical form

$$\chi_{e\text{-}h}(t, t') = \frac{\Theta(t - t')(1 + i)}{[2\pi(t - t')]^{1/2}}\exp[-\gamma(t - t')] \times \exp\left\{-i\int_{t'}^{t} dt''q^2(t'') + i\frac{\left[\int_{t'}^{t} dt''\, q(t'')\right]^2}{(t - t')}\right\}, \quad (5)$$

where $q(t)$ is the vector potential, i.e., $dq(t)/dt = f(t)$. We solve Eq. (4) numerically for $t' = T$, treating exactly the square-root singularity of the kernel. Typically, we use of the order of 10^4 points, which guarantees both convergence and a large enough mesh for subsequent Fourier transformation.

Figure 1 shows excitonic electroabsorption spectra for various static electric fields f and $\gamma = 0.2$. These changes are similar to what has been observed in parallel field QW or bulk electroabsorption[1,2]; however, we note that the case of perpendicular field quantum-confined Stark effect modulators is not described by the present model. For $f = 0.6$ the absorption spectrum displays a small Stark shift, broadening, and the loss of oscillator strength of the exciton as well as Franz–Keldysh oscillations in the e–h continuum of scattering states, while for $f = 2$ mainly broadening and loss of oscillator strength of the exciton are apparent.

Figures 2(a) and 2(b) show time-resolved differential absorption spectra near the exciton resonance as measured by $\Delta(\mathrm{Im}\,\chi)$, the difference between the absorption without field, $\mathrm{Im}\,\chi_0$, and the absorption with field, $\mathrm{Im}\,\chi$, for $f(t) = f\Theta(-t)$, with $f = 0.6$ [Fig. 2(a)] and $f = 2$ [Fig. 2(b)]. As explained above, because of causality the ideal broadband probe beam measures the absorption spectrum corresponding to the field value $f = 0$ for all positive time delays $T > 0$. For negative delays, corresponding to the probe pulse's passing

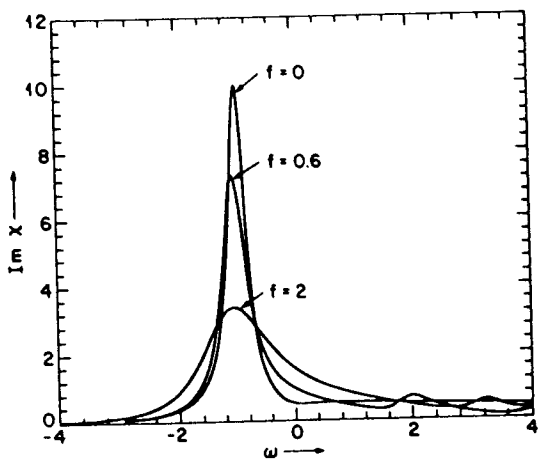

Fig. 1. Excitonic electroabsorption spectra for various static electric-field strengths f (in units of the ionization field R/ea) and a phenomenological exciton width at zero field $\gamma = 0.2R$.

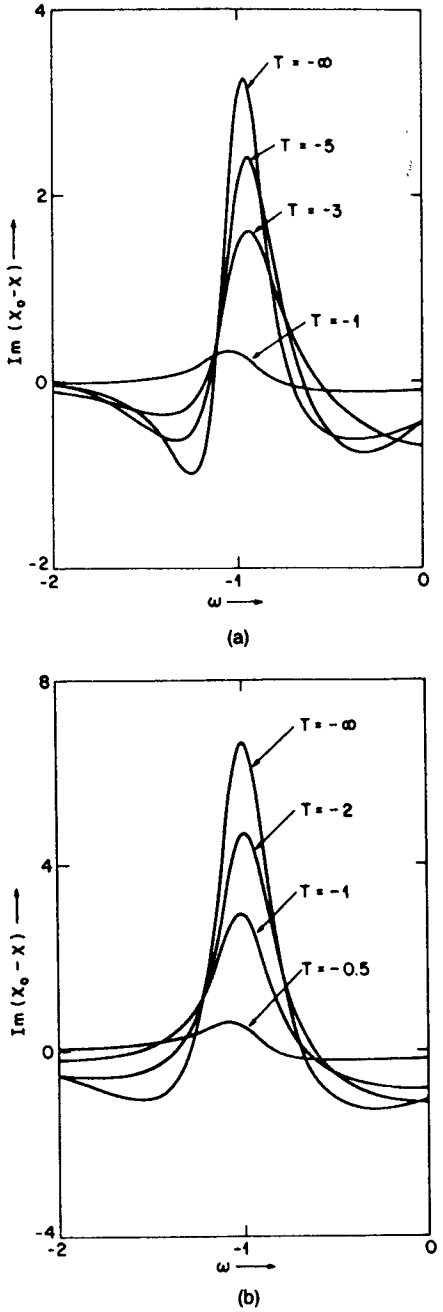

Fig. 2. Differential absorption spectra, as measured by the difference between the absorption without field and the absorption with field, for various time delays T (in units of R^{-1}) for (a) $f = 0.6$ and (b) $f = 2$.

through the sample just before the transient, Figs. 2(a) and 2(b) show a smooth evolution of the differential absorption spectra from $\Delta(\text{Im}\ \chi) = 0$ at $T = 0$ to the $\Delta(\text{Im}\ \chi)$ spectra corresponding to Fig. 1 at $T = -\infty$.

Although the electric field changes abruptly, the temporal response of the exciton absorption is perfectly well behaved. Comparison of Figs. 2(a) and 2(b) shows that the differential signal vanishes with a time constant that decreases with increasing field. This rules out the inverse exciton binding energy as a fundamental limit and leaves the inverse exciton linewidth in the presence of the field $\gamma(f)$ (i.e., including contributions from field ionization) as the mechanism governing the response time. The interpretation is further supported by a quantitative comparison with the profiles of Fig. 1. For $f = 0.6$ ($f = 2$) the exciton broadening has increased to ~0.3 (~0.8). The inverse of these numbers agrees with the negative delays for which the differential absorption has decreased to approximately half its value. Further calculations for different values of the zero field width γ confirmed this picture.

In conclusion, we have studied the temporal response of excitonic electroabsorption for a simple one-dimensional model under ideal conditions. Our results show that the fundamental limit on the response time is simply the inverse of the exciton linewidth (in the presence of an applied field), at least in the case of a homogeneous linewidth. In relevant experiments[5,8] the inverse linewidths correspond to approximately 50 fsec, but time resolutions of 330 fsec (Ref. 5) and 180 fsec (Ref. 8) have been shown. Therefore the present experiments have closely approached the limits that are suggested by the present model. We have noted that the interaction of the field transient with the decaying polarization owing to the probe pulse is the origin of this effect and that this result is related to recent models of femtosecond nonlinear-optical response of two-level systems.[14]

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