Ultrafast Adiabatic Following in Semiconductors

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The full semiconductor Bloch equations are solved to analyze the ultrafast exciton dynamics in semiconductors. Transient adiabatic following is obtained for large detunings and arbitrary intensities, leading to exciton Stark shift, bleaching, and recovery on the time scale of the pump pulse. The many-body Coulomb effects strongly influence the semiconductor response for resonant excitation conditions, almost doubling the effective Rabi frequency of the applied field.

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One of the most exciting aspects of the ultrashort laser pulses is the possibility to explore the similarities and differences of light-matter interaction in atomic systems and dielectric media. Investigating the transient response of semiconductor excitons for various excitation conditions and comparing the results with those of a discrete-level system allows identification of the influence of the many-body Coulomb effects on the dynamic response. Research along these lines led to the discovery of the optical Stark effect in semiconductors.[1] For large detuning and low intensities it has been shown recently that one obtains an almost ideal blueshift of the exciton without saturation.[2,3] For the same detuning but larger intensities the dominant transmission change is a saturation and recovery of the exciton resonance during the pulse excitation.

One often exploits the apparent similarities of the optical Stark effect in semiconductors and in atomic systems to analyze the basic features, using *two-level Bloch equation theory* to compute the ultrafast response. However, it is well known[4-10] that the proper *semiconductor Bloch equations* contain additional terms, the so-called exchange terms, which in general do not allow a reduction to an effective two-level description. To clarify the role of the many-body Coulomb effects we investigate the following questions: Can the ultrafast exciton recovery be explained by the semiconductor Bloch equations, or do we need additional assumptions concerning the semiconductor decay constants? What is the role of the Coulomb interaction? How does the Hartree-Fock energy renormalization and the reduction of the Coulomb attraction due to phase-space filling contribute to the ultrafast recovery? What is the relation between the excitonic response and the well-known discrete-level dynamics? Under which conditions can Rabi oscillations occur in semiconductors; i.e., when is the final carrier density determined only by the area under the pulse, so that full dynamic recovery occurs only for integer multiples of 2π pulses?

To answer all these questions we numerically solve the full semiconductor Bloch equations for the situations of resonant and nonresonant femtosecond excitation of the exciton and compare the results with those of the discrete-level quantum optics Bloch equations.[11] For moderate to large detunings and arbitrary excitation intensities we find that the complete Bloch vector, whose components are the complex interband polarization $P_{\mathbf{k}}$ and the carrier distribution function $f_{\mathbf{k}}$, is completely determined by the dynamics of the pump pulse. This is the phenomenon of ultrafast adiabatic following known from atomic physics,[11] which we thus identify for the case of semiconductors. For the case of resonant excitation, however, we find that Coulomb effects lead to dramatic renormalizations of the applied field, drastically modifying the exciton response in comparison with that of an atomic system.

The semiconductor Bloch equations are the Hartree-Fock equations for the many-body electron-hole Hamiltonian of a two-band semiconductor dipole coupled to an external field $E(t)$. As derived in Refs. 5–10, these equations can be written as

$$i\hbar\frac{\partial}{\partial t}P_{\mathbf{k}}=\left(\epsilon_{\mathbf{k}}-\frac{i\hbar}{T_2}-2\sum_{\mathbf{k}'\neq\mathbf{k}}V_{\mathbf{k}-\mathbf{k}'}f_{\mathbf{k}'}\right)P_{\mathbf{k}}-(1-2f_{\mathbf{k}})\left(\sum_{\mathbf{k}'\neq\mathbf{k}}V_{\mathbf{k}-\mathbf{k}'}P_{\mathbf{k}'}+\mu E\right),$$

$$\hbar\frac{\partial}{\partial t}f_{\mathbf{k}}=-\frac{\hbar(f_{\mathbf{k}}-f_{\mathbf{k}}^0)}{T_1}+2\,\mathrm{Im}\left(\mu^*E^*P_{\mathbf{k}}+\sum_{\mathbf{k}'\neq\mathbf{k}}V_{\mathbf{k}-\mathbf{k}'}P_{\mathbf{k}'}^*P_{\mathbf{k}}\right),$$

where $\epsilon_{\mathbf{k}}=\hbar^2k^2/2m+E_g-\hbar\omega_0$, E_g is the band-gap energy at zero density, ω_0 is the central frequency of the applied field, T_1 and T_2 are the phenomenological population and phase relaxation times, $f_{\mathbf{k}}^0$ is the equilibrium carrier distribution, and μ is the dipole interband matrix element. Since the lowest subband heavy-hole exciton in a thin quantum well

is well separated from all higher-lying excitons, we employ the two-band model where the Coulomb potential V is evaluated with the $n=1$ eigenfunctions of an infinitely high well. The exchange terms are $2\sum_{k'}V_{k-k'}f_{k'}P_k$, $2f_k\sum_{k'}V_{k-k'}P_{k'}$, and $\sum_{k'}V_{k-k'}P^*_{k'}P_k$. The E field contains the excitation and the probe field propagating in the directions $\mathbf{K}_x$ and $\mathbf{K}_p$, respectively, which leads to polarization components in $\mathbf{K}_p$, $\mathbf{K}_x$, and $2\mathbf{K}_x-\mathbf{K}_p$ directions. After linearizing with respect to the probe field amplitude and Fourier transformation with respect to t, the polarization contribution in the $\mathbf{K}_p$ direction yields the probe spectrum. Screening of the Coulomb potential V and T_1-relaxation processes are of minor importance in the fs Stark effect. We therefore assume $f_e=f_h=f$.[12]

First, we discuss the case of nonresonant excitation well below the exciton resonance so that no spectral overlap between the pump pulse and the resonance exists, thus eliminating the possibility for real carrier generation. Taking the temporal width of the pump-pulse intensity $\Delta t=120$ fs (FWHM) and the exciton dephasing time as 0.8 ps, we obtain the probe absorption spectra shown in Fig. 1 for positive pump-probe delays t_p, i.e., when the pump precedes the probe. We clearly see the blueshift of the exciton resonance which is accompanied by pronounced bleaching and some negative absorption (transient gain) below the resonance. All these features recover gradually for positive t_p, in very good qualitative agreement with the experimental observations.[1,3] For the present case of relatively large detuning below the exciton resonance the pump pulse generates carriers, which are often called "virtual" although they are real carriers which are present in the medium only during the time in which the pulse is present. The carriers reduce the exciton oscillator strength via phase-space filling, causing the exciton bleaching and recovery. Figure 1 shows that, in agreement with the experimental findings,[3,13,14] exciton bleaching and recovery occurs during the presence of the pump pulse. As already noted in Ref. 7, the time dependence of the corresponding carrier density follows the pump-pulse amplitude, even though the characteristic medium response times T_1 and T_2 are both much longer than the pulse duration. For negative t_p (not shown) we observe coherent oscillations.[4]

To investigate the influence of the many-body effects on the dynamic semiconductor response, we solved the semiconductor Bloch equations also for the case of resonant excitation. Before we discuss these results, however, let us briefly recall some properties of simple discrete-level systems, which are well known to show dramatically different dynamic responses for different values of the relevant parameters.[11] (i) For the case of strong damping, polarization, and population in a discrete-level system follow the amplitude of the applied field adiabatically. (ii) For not too strong damping and resonant excitation, the dipole-coupled level populations undergo Rabi oscillations. (iii) For large detuning, one again observes adiabatic following which in this case, however, is independent of the system damping. The absence of Rabi oscillations can be explained completely by the spectral detuning of the pulse with respect to the atomic transition energy. In this Letter we study the semiconductor response for situations corresponding to (ii) and (iii).

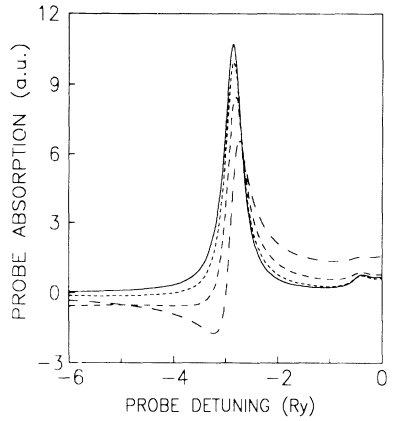
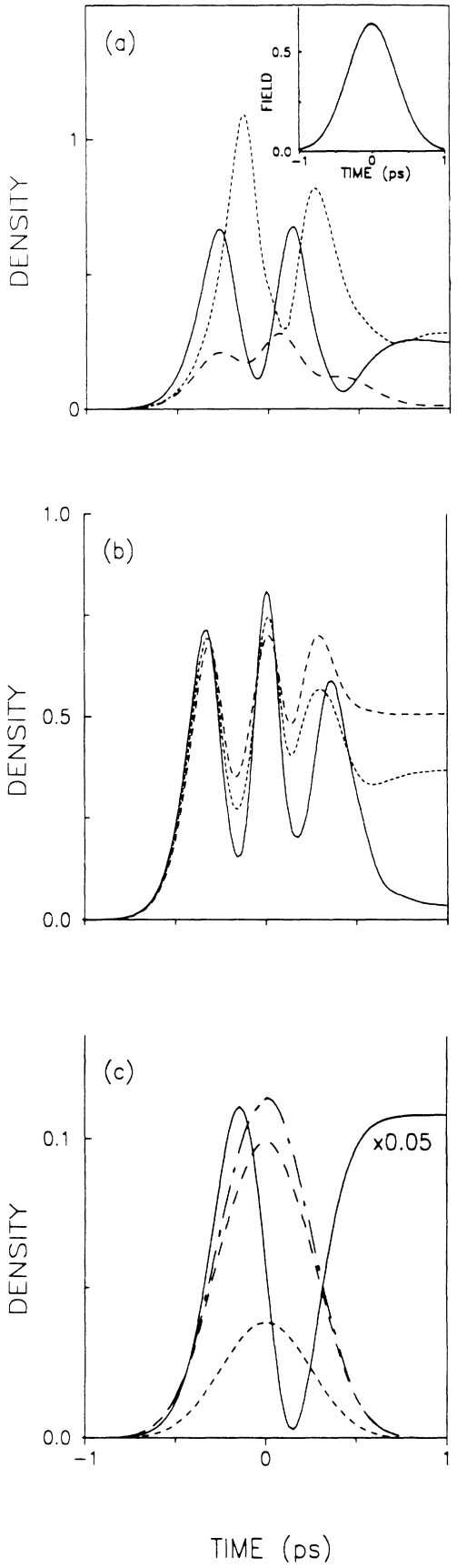


FIG. 1. Computed probe absorption spectra for a 50-Å GaAs quantum well vs probe detuning without pump (solid line) and for various pump-probe time delays: $t_p=0$ (long-dashed curve), $t_p=60$ fs (medium-dashed curve), $t_p=120$ fs (short-dashed curve). The probe detuning is plotted in units of the bulk exciton rydberg energy R^* ($=4.2$ meV in GaAs) with respect to the unrenormalized (zero-excitation) band gap. The pump detuning is $9R^*$ below the exciton and the peak pump intensity was $\mu E_{\max}=2.6R^*$.

Since we want to clearly distinguish between coherent and incoherent effects, we first investigate the limit $T_2\gg\Delta t$. As an illustrative example we show in Fig. 2(a) the time dependence of the semiconductor density for a 2.2π pulse, i.e., $(2/\hbar)\int dt\,\mu E(t)=2.2\pi$, exciting the system at the exciton resonance or at slightly detuned frequencies. It is well known that the time-dependent density of a resonantly excited two-level system exhibits oscillations (Rabi flops) between the dipole-coupled states. In semiconductors, however, there is no real "on-resonant" condition since the exciton involves a large number of different k states, each of which has a slightly different detuning from the pump pulse. Different detunings imply different Rabi frequencies of the individual states, which might lead to pronounced interference effects, eventually even completely destructive interference, and the absence of Rabi oscillations. However, as Fig. 2(a) shows, when excited at or above the exciton resonance, the semiconductor carrier density clearly exhibits temporal oscillations. Moreover, the number of Rabi flops is basically twice that expected for a two-level system. This feature will be analyzed below. Here we



just note that a finite carrier density remains in the system after the pulse is gone. Figure 2(a) also demonstrates that the amplitude of the Rabi oscillations is significantly reduced already for the relatively small detuning of $-0.4E_b$, where $E_b=2.85R^*$ [$-1.4E_b$ below the gap, long-dashed curve in Fig. 2(a)]. The oscillations occur only as small variations on an envelope which basically exhibits the adiabatic following character known from discrete-level systems, i.e., situation (iii) mentioned above. The density remaining after the pulse is very small in comparison to the cases of resonant excitation.

To study the influence of damping and to further illustrate the role of the many-body Coulomb effects, we plot in Figs. 2(b) and 2(c) the results for resonant excitation with a 3π pulse for different dephasings using the full semiconductor Bloch equations [Fig. 2(b)] and for the cases without exchange terms [solid line, Fig. 2(c)] and without all Coulomb terms [short-dashed line, Fig. 2(c)]. Furthermore, we show the system response with and without the exchange terms when the pump is tuned one binding energy below the exciton resonance as dash-dotted and long-dashed curves in Fig. 2(c), respectively. We see in Fig. 2(b) that the most pronounced effect of increasing dephasing is an increasing carrier density left in the system when the pump pulse is gone. This is fully expected since finite dephasing leads to a broadening of the exciton resonance and, hence, to increased spectral overlap between pump-pulse and exciton absorption peak. With increasing overlap more and more real absorption occurs, so that the density dynamics becomes a superposition of the coherent and incoherent features, with the incoherent feature being a monotonous increase roughly proportional to the time integral over the pulse. More surprising is the difference between the results shown as solid lines in Figs. 2(b) and 2(c). The solution without the exchange terms [Fig. 2(c)] exhibits 1.5 density oscillations, exactly as expected for the case of a two-level system under these conditions. However, as mentioned above, the solutions of the full semiconductor Bloch equations [Fig. 2(b)] show twice as many flops,

FIG. 2. (a) Excited carrier density, $n=2\sum_k f_k$, in units of a_B^{-2}, where a_B is the exciton Bohr radius for bulk GaAs, for various excitation frequencies: $\hbar\omega_0=E_g-E_b$ (solid line), $\hbar\omega_0=E_g-1.4E_b$ (long dashed line), $\hbar\omega_0=E_g-0.6E_b$ (short-dashed line). Inset: Pump field $\mu E(t)/R^*$ as a function of time for a pulse with an area of 2.2π. (b) Carrier density for resonant excitation with a 3π pulse without dephasing (solid line), with a dephasing time $\hbar/T_2=0.2R^*$ (long-dashed line) and $\hbar/T_2=0.1R^*$ (short-dashed line), respectively. (c) Solid line: computed carrier density without dephasing and exchange terms for otherwise the same conditions as the solid line in (b). Short-dashed line: same conditions as the solid line but $V\equiv 0$. Large detuning limit ($1E_b$ below the exciton resonance) with (dash-dotted line) and without (long-dashed line) the exchange contributions.

demonstrating clearly that the field renormalization $\sum VP$ is of the same magnitude as μE. Hence, the exchange terms are extremely important under resonant excitation conditions. The omission of these contributions even removes the physically required restriction for the distribution function f to vary only between 0 and 1. The short-dashed line in Fig. 2(c) shows the completely unrealistic results when we omit all Coulomb terms in the Bloch equations. For comparison we also studied resonant excitation of a bulk semiconductor (not plotted) and found the same approximate doubling of the number of Rabi flops with and without the exchange terms. The results at $-E_0$ detuning in Fig. 2(c) demonstrate the transition to the adiabatic following regime. Already for this relatively small detuning, the exchange contributions have almost no influence on the dynamics, which becomes qualitatively similar to that of an atomic system, as already assumed in our earlier analysis of the nonresonant excitonic Stark effect.[2,4,10]

In conclusion, we presented an analysis of the complete semiconductor Bloch equations. We demonstrate the significance of the many-body Coulomb effects for the case of resonant excitation. For detunings below the exciton resonance we identify transient ultrafast adiabatic following of the semiconductor polarization and of the carrier density, allowing Stark shift, bleaching, and full recovery during the pump excitation. The system kinetics is almost completely determined by the light field and the semiconductor specific many-body contributions are unimportant in this case.

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[12]We make this approximation mainly to simplify the numerical evaluation; it is not crucial for the presented results. Moreover, electron and hole distributions are always equal for the model case of equal electron and hole masses. Even for real semiconductors $f_e \cong f_h$ for nonresonant fs excitation because of the adiabatic following. It is also a reasonable approximation for the investigated case of resonant excitation, as long as the pulse is present and shorter than the characteristic T_1 times.

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