Excitonic-state trapping and quasiadiabatic population transfer in a two-band semiconductor

S. Hughes,* W. Harshawardhan,† and D. S. Citrin
Department of Physics, Washington State University, Pullman, Washington 99164-2814
(Received 10 March 1999)

By solution of the semiconductor Bloch equations in the presence of a frequency-modulated optical pulse, clear exciton trapping and quasiadiabatic population transfer in a two-band semiconductor is predicted. [S0163-1829(99)00647-5]

Driven by the pursuit to understand the differences and similarities between the semiconductor and atomic systems resonantly excited by laser pulses, the coherent nonlinear dynamics of direct-gap semiconductors has been vigorously investigated over the years. Moreover, with the ongoing advancement of short-pulse laser techniques and excellent semiconductors samples, classes of coherent dynamic phenomena have been discovered recently including, for example, coherent exciton control[1,2] and self-induced transmission.[3] Running in parallel have been discoveries in atomic optics such as population trapping using frequency modulated fields. A realization of such state trapping and its periodic redistribution in a two-level atom (TLA) was predicted in Ref. 4. In the past year, in combination with other well-known concepts such as adiabatic rapid passage, multiphoton resonances, and Landau-Zener transitions, trapping was experimentally demonstrated in a TLA [5]. For two-band semiconductors, however, trapping is scarcely expected due to Coulomb many-body complications and the valence/conduction band-continua of free-carriers. Surprisingly in this work we predict clear excitonic-state trapping (EST) and quasiadiabatic population transfer (QAPT) using multicolored excitation schemes.

For high-intensity ultrashort optical pulses, it is well established that Rabi oscillations of the population between two states can be seen in the temporal evolution of a TLA.[7] However, in the case of a sinusoidal frequency-modulated excitation, square-wave oscillations of the population—periodic state trapping—as well as more complicated and phase-dependent structures may appear. In semiconductors, the *two-level* model (as a first approximation to a two-band description) is considered inappropriate because Coulomb many-body interactions result in a renormalized Rabi energy and bandedge, and excitation-induced dephasing (EID). Nevertheless, self-induced transmission and multiple Rabi flopping on a semiconductor free exciton resonance have been recently reported in bulk[3] and quantum wells (QW's);[6] these measurements were successfully explained within the framework of the semiconductor Bloch equations. Naturally the question arises: Can one achieve population trapping dynamics in semiconductors using frequency-modulated or suitably-chirped broadband optical pulses? By employing, for example, a frequency-dependent 150-fs full width at half maximum (FWHM) irradiance sech pulse excited at the 1*s* exciton peak, we predict, *yes*, one can achieve EST and QAPT in a semiconductor, too. We present trapping dynamics in semiconductors akin to trapping in atomic systems.[4] We discuss in detail conditions to achieve EST and QAPT even when a broadband pulse excites both Coulomb-bound electron-hole (e-h) pairs (excitons) and free e-h pairs. The other well known trapping phenomenon in atomic systems is the coherent population trapping effect, which has been widely used *inter alia* in adiabatic population transfer, electromagnetically induced transparency, and velocity selective cooling. As pointed out recently by using adiabatic population transfer between heavy- and light-hole bands (to mimic a three-level atom),[8] the exact analog to the coherent-population-trapped state (dark state) used in atomic physics is not possible in semiconductors.

For our theoretical approach we assume the validity of the rotating-wave approximation and thus neglect the possibility of carrier-wave Rabi flopping.[9] We further assume a two-band QW where each e-h state within a certain bandstructure with a wave number $\mathbf{k}$, contributes to the total optical polarization $P=2A^{-1}\Sigma_{\mathbf{k}}d_{\mathrm{cv}}P_{\mathbf{k}}$, with d_{cv} the transition dipole matrix element. The SBE for the polarization functions are ($\hbar=1$) (Ref. 10) $\dot{P}_{\mathbf{k}}=-i\Delta_{\mathbf{k}}P_{\mathbf{k}}-i\Omega_{\mathbf{k}}(f^{e}_{\mathbf{k}}+f^{h}_{\mathbf{k}}-1)+\dot{P}_{\mathbf{k}}|_{\mathrm{corr}}$, where $\Delta_{\mathbf{k}}$ is the renormalized energy dispersion for a parabolic two-band semiconductor and $\Omega_{\mathbf{k}}$ is the generalized Rabi frequency, while $f^{e/h}_{\mathbf{k}}$ are the carrier distribution functions of the electrons and holes. The Coulomb interaction is treated here in a quasi-static approximation (single plasmon-pole).[10] Similarly, the carrier density can be calculated from the electron (or hole) population $N=2A^{-1}\Sigma_{\mathbf{k}}f_{\mathbf{k}}$, where $f_{\mathbf{k}}$ is calculated from $\dot{f}^{e/h}_{\mathbf{k}}=iP^{\star}_{\mathbf{k}}\Omega_{\mathbf{k}}-iP_{\mathbf{k}}\Omega^{\star}_{\mathbf{k}}+\dot{f}^{e/h}_{\mathbf{k}}|_{\mathrm{corr}}$. In addition to the terms that result from the time-dependent Hartree-Fock approximation, the carrier-carrier (CC) and carrier-phonon (CP) collisions drive the nonequilibrium distribution functions towards quasiequilibrium Fermi functions and yield optical dephasing. The CC collisions (Coulomb correlation terms) are calculated from the e-h Boltzmann equations including also a correlation field, nondiagonal dephasing, and polarization scattering.[11–13] The influence of CP interactions occurs over much longer time scales than those studied in this work and can be safely neglected.

We assume input optical pulses of the form $\mathbf{E}(\mathbf{r},t)=E(t)e^{-i[\omega_0 t+k_0 z+\phi(t)]}+\mathrm{c.c.}$, polarized in the plane of the QW, with all material parameters corresponding close to $In_xGa_{1-x}As/GaAs$ QW's (Ref. 14) and $\omega_0=\omega_x$ (1*s* exciton resonance). $In_xGa_{1-x}As/GaAs$ QW's are advantageous since with compressive strain one can increase the splitting of the heavy- and light-hole exciton and thus neglect the light-hole states, therefore validating the two-band model. In analogy with the atomic systems we modulate the input field by the phase factor $\phi(t)=M\sin(\Omega t)$, with M and Ω the index and frequency of modulation. For a TLA the resonant part of the

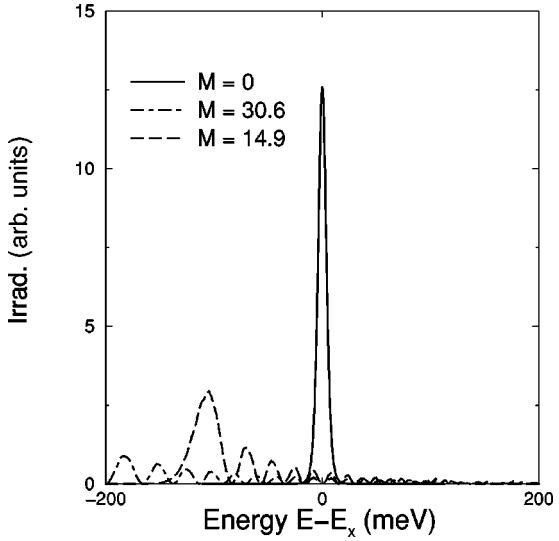

FIG. 1. Irradiance spectra for a frequency-modulated and unmodulated 150 fs optical pulse. Energy is in units of the exciton binding frequency.

interaction Hamiltonian vanishes if M is chosen such that the bessel function $J_0(M)=0$ and one is left with the nonresonant rapidly oscillating terms.[4] The solid-state community may recall that a somewhat similar criterion has been derived to realize dynamic localization in infinite lattices driven by a harmonic time-dependent electric field.[15,16]

In the TLA it is further known that the two bare levels cross at the times $t_n=n\pi/2\Omega$ ($n=$ integer). The crossing of energy levels occurs quite naturally in the instantaneous frame of the modulated field. The population distribution at the crossings can be understood semiquantitatively by integrating numerically the time-dependent Schrödinger equation or by employing Landau-Zener theory.[4] For semiconductor the situation is much more complex, whereby the optical field provides coupling between two bands, and a modulated near-resonant field leads to coupling of higher spectral components of the field to larger $\mathbf{k}$ states in the band, and the field components below the resonance provide a nonresonant coupling to the excitonic transition. Moreover, the Coulomb interaction yields exciton and plasma induced many-body effects. These conditions prohibit us from having a simple set of criteria for trapping. Thus we propose here a novel spectral distribution for the field, which has its central frequency detuned far below the excitonic resonance, and, by strategic sweeping of the instantaneous carrier frequency, dynamical population trapping can be realized. As in the TLA case we do not necessarily have to choose M to be strictly a zero of the Bessel function.

For the modulation frequency we choose $\Omega=8$ meV unless stated otherwise. As mentioned above, for the *unmodulated* field, $E(t)$, we employ a 150-fs pulse excited at the $1s$ exciton peak. In Fig. 1, we depict the spectral irradiance of the unmodulated and frequency-modulated fields. With modulation, a much larger bandwidth can be obtained with a series of larger peaks extending far below the exciton resonance; the injected pulse profile is sufficiently broad to couple many different excitation modes. One recognizes that the dominant spectral components are well below the band edge. By increasing the index of modulation (M), the spectral width of the pulse increases. In contrast, without any modulation the spectrum of the excitation pulse is significantly narrower and peaked at the central frequency as shown in Fig. 4(b) (solid line; see later). Although such a modulation is difficult to obtain experimentally, one only requires about one approximate oscillation to obtain the appropriate energy level crossings. Indeed one can also achieve trapping phenomena using suitably chosen linear chirps (discussed below). Further, recent developments of free-electron lasers as well as terahertz solid-state emitters may be utilized in materials to achieve the desired frequency modulation in a suitably chosen nonlinear material.[17]

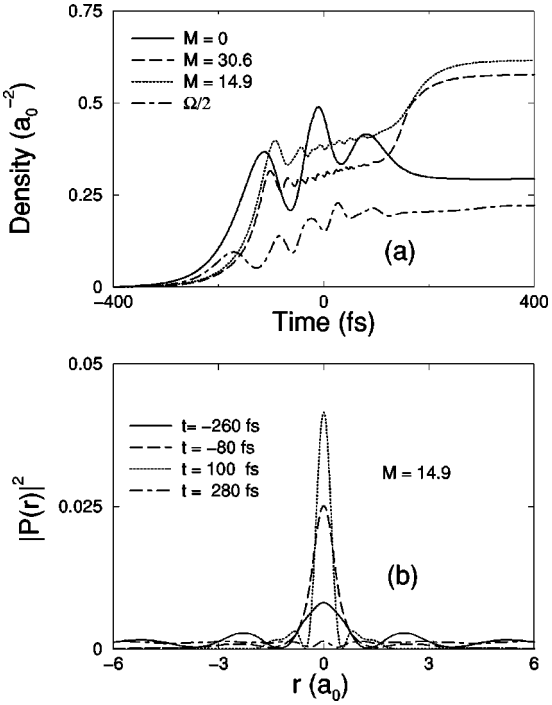

FIG. 2. (a) Pulse-induced carrier density (in units of inverse Bohr radius squared) as a function of time. The chain line corresponds to $M=14.9$ but with a frequency modulation of $\Omega/2$. (b) The polarization density corresponding to the dashed case in Fig. 2(a) at several snapshots.

For an optical pulse that has roughly an area (integral of its Rabi frequency over time) of 6π (~ 1 GW/cm^2), Fig. 2(a) shows the excitation-induced density for the QW optically excited at the $1s$ exciton resonance using (i) an unmodulated input pulse [the solid curve depicts the carrier-density showing the familiar Rabi flopping[7,18] of the density (complete inversion is not possible due to Coulomb processes)]; and (ii) a frequency-modulation of the input pulse for two different values of the index of modulation M: (the dashed and dotted curves are markedly different from the unmodulated case and display strong EST and ultrafast QAPT). The values of M were chosen in accord with the original work in atoms, while the pulse area was chosen to be large enough to drive the population into trapping–it is also close to values employed experimentally.[6] During the time interval from about -160 to 160 fs, the population remains trapped in the exciton level, eventually jumping out in the wake of the exciton-continuum quasi-adiabatic crossing. For the case of $\Omega/2$ and $M=14.9$ (chain curve), the trapping efficiency is much less as expected and QAPT does not oc-

cur, since the crossing occurs just once and the on-resonance field components are not sufficient to create the trappinglike feature; however, even in a regime where the pulse irradiance is negligible there is some evidence for small density changes at around 360 fs. In all cases, there are signatures of phase interference due to coherent carrier evolution along different interfering pathways and EID. We would like to point out that to obtain such good Rabi flopping one must include EID at a microscopic level that includes both nondiagonal dephasing and polarization scattering;[11] these contributions from CC scattering reduce the interband optical linewidths of the higher **k** states and thus limit the higher-energy continuum occupations in comparison to the pure dephasing (essentially a relaxation-time approximation) treatment.[12] Physically, this is important since the leading edge of the pulse, which is detuned below the exciton resonance, prepares the system for the excitation of real population. A state and energy-independent dephasing time would result in erroneous large dephasing of the initially virtual excitation and therefore suppress the QAPT and EST. Polarization scattering is also known to result in a transfer of oscillator strength from the continuum to the exciton.[13]

To highlight the difference between the Rabi flopping and trapping we depict in Fig. 2(b) the polarization density (wave-packet), $|P(\mathbf{r},t)|^2$ with $\mathbf{r}=\mathbf{r}_e-\mathbf{r}_h$ at various temporal snapshots.[19] During the period -260 to 100 fs the population becomes strongly trapped in the exciton state indicated in the figure by a high probability of finding the electron and hole at the same relative position. However at the later time of 280 fs, after the density increases rapidly (we mention that this increase is almost steplike in the absence of dephasing), the excitonic probability *decreases* substantially and the wave packet speads out significantly, demonstrating that the population is no longer trapped. This drammatic spreading of the wave packet arises due to the modulation and the resulting crossing of carriers into the continuum. The above picture sheds much more light on the trapping scenario than, for example, $f_{\mathbf{k}}^{e,h}$ or **k**, which do not clearly distinguish between Coulomb-bound excitons and free carriers in the continuum. The spatial polarization dynamics at $\mathbf{r}=0$ may in fact be probed experimentally using conventional four-wave mixing techniques. We observe here a method to control, coherently, the excitonic wave packet by tailoring the conditions of the energy level crossing. To probe the entire spatial dynamics one would need to, e.g., couple a THz field with the optical field; this would involve a significantly more complex analysis. We also mention that to direcly detect density changes, experimentally pulse-propagation studies are very difficult and one should, for example ,use the technique reported recently in Ref. 6. This allows for the detection of density changes via simple differential transmission changes of a probe pulse.

The aforesaid phenomena beg a more physically intuitive explanation: Adding a frequency-modulation to the excitation field is equivalent to modulating the energy separation between the ground and exciton/continuum states. In Fig. 3 we show, schematically, the various crossings of energy levels that lead to quasiadiabatic transfer of population at these crossings. The crossings ''*B–D*'' transform into anticrossings (solid lines) due to the coupling with the effective Rabi field. In contrast the crossing at ''*A*'' is unaffected due

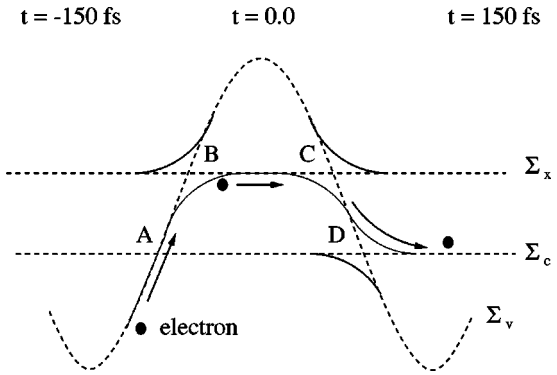

FIG. 3. Schematic of the energy level crossing that results from modulation of the field, where ''*A–D*'' represent the various crossings. The arrows depict the evolution of the population (large black dots).

to the weak coupling of the $v \leftrightarrow c$ transition on the leading edge of the pulse. The modulation of the dressed energy levels results from transforming into a frame corresponding to the instantaneous field frequency, and are given as $\Sigma_c = -[\Delta_{\mathbf{k}}^2 - \Delta_x^1]$, $\Sigma_x = 0$ and $\Sigma_v = -[\Delta_x^1 + M\Omega\cos(\Omega t)]$, for the electrons in the conduction band (c), excitonic state (x), and electrons in the valence band (v), respectively. One can of course depict the diagram in reverse for the holes. Here, Δ_x^1 $(\Delta_{\mathbf{k}}^2)$ denote the detuning of the central laser field frequency from the excitonic (continuum) energy levels. The coupling of the field to the $v \leftrightarrow x$ and $v \leftrightarrow c$ transitions transforms these crossings into *avoided crossings*. Initially the system will be off resonance and far from any crossing; as the modulation changes, the population is swept though resonance and it evolves quasi-adiabatically into the excitonic state where it displays the trapping feature. The system further encounters two closely spaced crossings, resulting in an enhanced step-like transfer of population into the continuum. The arrows in Fig. 3 indicate one possible temporal path (high probability) along which the electrons may evolve ($\Delta_{\mathbf{k}}^2$ is fixed for simplicity). The detuning term $\Delta_{\mathbf{k}}^2$-Δ_x^1 is the difference between the continuum energy levels and frequency of the the $1s$ exciton peak. One should keep in mind however that the above model is grossly simplified and many-body effects, included in our numerical results, will complicate things substantially; however our quantitative theoretical study is in fairly good agreement with the above level crossing model.

The essential difference in the trapping criterion between the atomic case and semiconductor case lies in the frequency content of the exciting field. In the atomic case the trapping-like phenomenon with nearly complete inversion results from *correlated sideband excitation* of the atom.[20] The frequency components of the modulated field excite the atom symmetrically about the atomic resonance, resulting in trapping. In the semiconductor, a symmetric frequency content of the excitation does not lead to the desired trapping due to its nonsymmetric coupling to the band structure of the semiconductor; effectively the high-frequency components of the modulated pulse selectively excite the continuum of states resulting in large carrier generation, thus washing out the trapping. We circumvent this problem by employing a modulated pulse with its predominant frequency content away from the band edge.

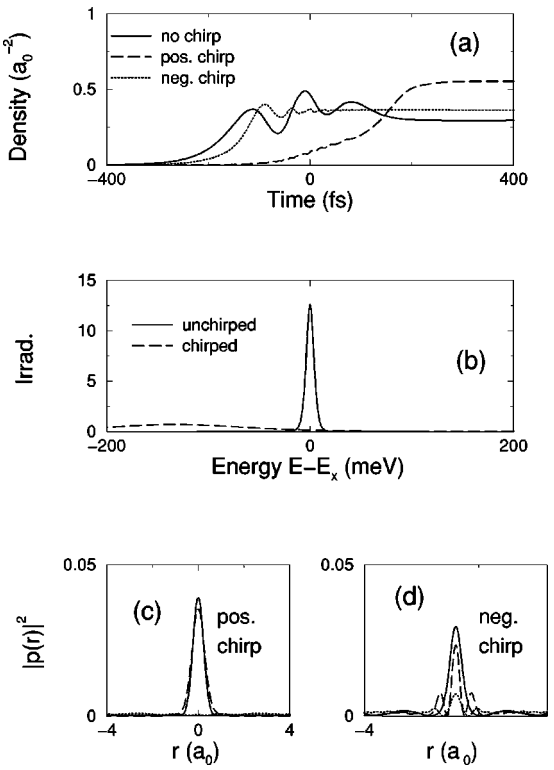

FIG. 4. (a) Pulse-induced carrier density as a function of. (b) Corresponding input pulse spectra for (a). (c) The polarization density for the positive chirp corresponding to the dashed case in (a) at several snapshots (see text). (d) As in (c) but for negative chirp.

Next, we employ linear-chirped input pulses to discern if some of the same qualitative trapping features (EST and QAPT) can also be obtained using suitably chosen chirp parameters. To best immitate the frequency-modulated spectrum, we choose a frequency chirp of the form: $\omega \rightarrow \omega_x - 120\ \text{meV} + a_c t$ with a_c being ± 0.5 meV/fs. $E_0(t)$ is the same as before. The chirped-pulse spectrum along with the unchirped spectrum is displayed in Fig. 4(b). The pulse-induced density with positive a_c is shown by the dashed line which, although does not show signs of trapping, does show fast oscillations in the density as well as a rapid increase in the density near the crossing time. For the negative a_c, strong excitonic trapping is again achieved and seems to maintain its trapped state since no further crossing takes place. This is again clear from the simplified zero-order energy-level picture with appropriate crossing of the $1s$ exciton state, ground crystal state, and continuum states. The slope and sign of the chirp (a_c) gives us a handle to selectively suppress or enhance the excitonic transition. The trapping and population transfer are highlighted in Figs. 4(c)–4(d) which show the wave-packets at the times -80 fs (solid curve), 100 fs (dashed curve), and 280 fs (dotted curve) for both the negative and positive chirp pulse excitation. Population transfer is not expected for the negative-chirp case as the zero-order energy level never cross.

In conclusion, we predict the possibility of exciton trapping and quasiadiabatic population transfer in a two-band semiconductor using frequency-modulated optical pulses. It is clear that the trapping feature arises out of an interplay of the dominant excitonic resonance and the excitation by a sufficiently *strong, broadband off-resonant pulse*, with a weak yet broad spectral content to excite the continuum of states. The population redistribution results from the crossing of energy levels. Besides being an intriguing theoretical study, our results are timely with recent advances in frequency-modulated spectroscopy techniques and the observation of multiple Rabi flopping on free exciton transitions.

This work was supported by the National Science Foundation Grant No. DMR9705403 and by the Office of Naval Research.

*Present address: Department of Physics, University of Surrey, Guilford, Surrey, GU2 5XH, United Kingdom. Electronic address: steve.hughes@surrey.ac.uk

†Present address: Department of Physics, Illinois State University, Normal, IL 61790-4560.

[1] A. P. Heberly *et al.*, Phys. Rev. Lett. **77**, 2598 (1995).
[2] D. S. Citrin, Phys. Rev. Lett. **77**, 4596 (1995).
[3] H. Giessen *et al.*, Phys. Rev. Lett. **81**, 4260 (1998).
[4] G. S. Agarwal and W. Harshawardhan, Phys. Rev. A **50**, 4465 (1994).
[5] M. W. Noel *et al.*, Phys. Rev. A **58**, 2265 (1998).
[6] A. Schülzgen *et al.*, Phys. Rev. Lett. **82**, 2346 (1999).
[7] See, for example, L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1995).
[8] R. Binder and M. Lindberg, Phys. Rev. Lett. **81**, 1477 (1998).
[9] S. Hughes, Phys. Rev. Lett. **81**, 3363 (1998).
[10] M. Lindberg and S.W. Koch, Phys. Rev. B **38**, 3342 (1988).
[11] A. Knorr *et al.*, Chem. Phys. **210**, 27 (1996).
[12] S. Hughes *et al.*, J. Opt. Soc. Am. B **49**, 754 (1997).
[13] T. Rappen *et al.*, Phys. Rev. B **49**, 10 774 (1994).
[14] Reduced mass, $\mu = 0.035\ m_e$; $1s$ excitonic binding energy, $E_{1s} \approx 12$ meV; and Bohr radius (2D), $a_0 = 75$ Å.
[15] D. H. Dunlap and V. M. Kenkre, Phys. Rev. B **34**, 3625 (1986).
[16] M. Holthaus, Phys. Rev. Lett. **69**, 351 (1992).
[17] G. von Plessen (private communication).
[18] I.I. Rabi, Phys. Rev. **51**, 652 (1937).
[19] W. Harshawardhan and G. S. Agarwal, Phys. Rev. A **55**, 2165 (1997).
[20] P.K. Lam and C.M. Savage, Phys. Rev. A **50**, 3500 (1994).