Ultrafast Adiabatic Population Transfer in p-Doped Semiconductor Quantum Wells

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The light-induced adiabatic population transfer of holes from the heavy-hole (hh) to the light-hole (lh) band in p-doped semiconductor quantum wells is investigated theoretically. The exact analog to the population-trapped state (PTS) used in atomic and molecular adiabatic population transfer does not exist in a semiconductor due to the continuum of transition energies and the dynamic light-induced shifts thereof. However, it is found that the population transfer only requires an approximate PTS condition to be fulfilled. As for a possible observation of the effect, the transient creation of a hh exciton resonance at the expense of the lh exciton is predicted. [S0031-9007(98)06870-7]

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In atomic and molecular 3-level physics, coherent optical techniques allowing for almost complete ultrafast adiabatic transfer of population between different molecular eigenstates have been developed and refined for many years [1–5]. It is possible to completely transfer population between two not-optically-coupled states by using a third state which is optically coupled to both the initial and the final state. The material is excited by two light pulses, one for each transition, with equal or similar detunings. This yields the so-called population-trapped state (PTS) or dark state, which is a superposition of the two not-optically-coupled states, and the weighting of the two states depends on the light-field amplitudes. The coherence between the two states is also called a Raman coherence, since the states are not optically coupled to each other. During the pulsed excitation the PTS changes according to the momentary field amplitudes. If the amplitude change is adiabatic, a system originally in the PTS remains in it. By delaying one of the pulses, a PTS can be created that coincides initially with one of the states of the 3-level system (the initial state). When the second pulse appears, the PTS is a superposition of the two states, and, when the first pulse is switched off before the second pulse, the PTS coincides in the end with the other state in the 3-level system (the final state). In this way, the population is transferred from one state to another. The transfer seems to work best in the so-called counterintuitive pulse delay scheme in which the first pulse corresponds to the optical transition that includes the final state and, therefore, to a transition between two empty states. In a 3-level system the exact resonance condition underlying the PTS is not necessary for the transfer to work. In the off-resonance case, a quasiadiabatic transfer solution can be obtained if there is a crossing of two of the three adiabatic eigenvalues of the dressed 3-level system. The crossing is essential, since it is necessary in order to have the states switched in the limit of vanishing light amplitudes (i.e., before and after the pulse sequence). We will publish more details on this elsewhere.

The physical processes in semiconductor quantum wells, which can be characterized in terms of a conduction (c) and two valence (v) bands [i.e., one heavy-hole (hh) and one light-hole (lh) band], are much more complex than in atomic 3-level systems, because of band structure effects and because of the Coulomb interaction which yields excitonic and plasma-induced many-body effects. Nevertheless, the existence of nonradiative coherences in semiconductors has been proven experimentally (see, e.g., [6]). Also, theoretical simulations predict the existence of the PTS in semiconductors [7]. In the following, we address the issue of light-induced population transfer in semiconductors, including the questions of (i) whether theory predicts this process to be possible in principle, (ii) if so, in what parameter regime does it work best, and (iii) how one can observe it. A semiconductor system suitable to study population transfer is a p-doped semiconductor quantum well. If the hh-lh splitting is sufficiently large for a given doping concentration and temperature, the p doping yields only heavy holes. In this case, the linear absorption spectrum does not exhibit a hh exciton because of the Pauli blocking. However, because of the absence of light holes, it may exhibit a lh exciton if the screening due to hh's is not too large. The transfer of holes from the hh to the lh band would result, ideally, in a reversal of excitons, i.e., in a linear spectrum that contains a hh exciton but not a lh exciton. For a related investigation of optical excitation of spatially indirect excitons in double quantum wells utilizing the counterintuitive pulse delay scheme see Ref. [8].

The theory is based on equations of motion for the time-dependent optical polarization functions $P_{sj}(\vec{k})$ as well as the electron and hole distribution-coherence functions $f_{ss'}(\vec{k})$ and $f_{jj'}(\vec{k})$, respectively. The vector $\vec{k}$ is the two-dimensional in-plane wave vector, the electron

quantum numbers s, s' denote the spin-degenerate conduction bands with $s = \pm 1/2$, and the hole quantum numbers j, j' denote the two degenerate heavy-hole ($j = \pm 3/2$) and light-hole ($j = \pm 1/2$) bands. The equations of motion comprise the coherent Hartree-Fock (HF) contributions and incoherent dephasing and scattering contributions. If (as assumed in the following) biexcitonic effects are insignificant, the Hartree-Fock contributions to the nonlinear optical response constitute the ideal limit in the sense that optical pulses can, in principle, be shorter than typical incoherent scattering times, but the many-particle effects described by the Coulomb terms in the HF theory cannot be neglected on any time scale. Since typical femtosecond pulses do not quite reach this ideal limit, we have amended the HF theory with dephasing contributions and a self-consistent multiband relaxation rate model. Before discussing this model, we review briefly the basic HF contributions to the theory, which are similar to those used in [9], except we use a quasistatically screened HF theory to account for the effects of the p doping. The equation for $P_{sj}(\vec{k})$ reads ($\hbar = 1$)

$$i\frac{d}{dt}P_{sj}(\vec{k}) = \sum_{s'j'}\{[\delta_{ss'}\delta_{jj'}\varepsilon_k^s + \delta_{ss'}\mathcal{H}_{jj'}(-\vec{k})]P_{s'j'}(\vec{k}) - \Omega_{s'j'}(\vec{k})[\delta_{ss'}\delta_{jj'} - \delta_{jj'}f_{ss'}(\vec{k}) - \delta_{ss'}f_{jj'}(\vec{k})] + [\delta_{jj'}\Sigma_{ss'}(\vec{k}) + \delta_{ss'}\Sigma_{jj'}(-\vec{k})]P_{s'j'}(\vec{k})\} + i\frac{d}{dt}P_{sj}(\vec{k})\Big|_{\text{scatt}}, \quad (1)$$

and that for the hole distribution function is

$$i\frac{d}{dt}f_{jj'}(\vec{k}) = \sum_{j''}\{\mathcal{H}_{jj''}(\vec{k})f_{j''j'}(\vec{k}) - f_{jj''}(\vec{k})\mathcal{H}_{j''j'}(\vec{k})\} + \sum_{s}\{\Omega^*_{sj'}(-\vec{k})P_{sj}(-\vec{k}) - \Omega_{sj}(-\vec{k})P^*_{sj'}(-\vec{k})\} + \sum_{j''}\{\Sigma_{jj''}(\vec{k})f_{j''j'}(\vec{k}) - f_{jj''}(\vec{k})\Sigma_{j''j'}(\vec{k})\} + i\frac{d}{dt}f_{jj'}(\vec{k})\Big|_{\text{scatt}}. \quad (2)$$

A similar equation, but without the $\mathcal{H}$ contribution, governs $f_{ss'}(\vec{k})$. Here the electron energies are parabolic, $\varepsilon_k^s = \hbar^2k^2/2m_e + \Sigma_{\text{CH}} + E_G$, where E_G is the band gap of the undoped quantum well and Σ_{CH} is the Coulomb hole self-energy. The Luttinger Hamiltonian $\mathcal{H}$ consists of two 2×2 matrices (compare with [9]) with elements $H_{hh} = (\hbar^2/2m_0)(\gamma_1 + \gamma_2)k^2$, $H_{lh} = (\hbar^2/2m_0)(\gamma_1 - \gamma_2)k^2 + \Delta_{hh-lh}$, and, within an isotropic approximation,

$$c = -(\hbar^2/2m_0)\sqrt{3}\,[\gamma_2(k_x^2 - k_y^2) - 2i\gamma_3k_xk_y] \approx -(\hbar^2/4m_0)\sqrt{3}\,(\gamma_2 + \gamma_3)k^2.$$

Here γ_1, γ_2, γ_2 are the Luttinger parameters. The energy renormalizations are given by (with $a = s$ or j)

$$\Sigma_{aa'}(k) = -\sum_{\vec{q}''}V(\vec{q})f_{aa'}(\vec{k} + \vec{q}), \quad (3)$$

and the renormalized dipole energy is

$$\Omega_{sj}(k) = \vec{\mu}_{sj}\cdot\vec{E} + \sum_{\vec{q}}V(\vec{q})P_{sj}(\vec{k} + \vec{q}). \quad (4)$$

The interaction potential $V(q)$ is the statically screened Coulomb potential including the form factor according to the lowest-subband wave functions in an infinitely deep quantum well. The static screening function is taken as $\epsilon_q^{-1} = 1 - \omega_{pl}^2(q)/\omega^2(q)$, where $\omega_{pl}(q)$ and $\omega(q)$ are the two-dimensional plasmon frequency and plasma dispersion function, respectively [10]. The light pulses are denoted by $\vec{E}$. The dipole matrix elements $\vec{\mu}_{sj}$ contain the information of optical selection rules. The dipole matrix elements [11] are given by $\vec{\mu}_{1/2,3/2} = \sqrt{3}\vec{\mu}_{-1/2,1/2} = -\mu\vec{e}_+$ and $\vec{\mu}_{-1/2,-3/2} = \sqrt{3}\vec{\mu}_{1/2,-1/2} = -\mu\vec{e}_-$, where μ is the magnitude of the microscopic Cartesian dipole element. In the following, we will call the transitions from $j = 3/2$ and $j = -1/2$ to $s = 1/2$ "spin $+1$ transitions" (and this subset of bands "spin $+1$ subset") and the transitions from $j = -3/2$ and $j = +1/2$ to $s = -1/2$ "spin -1 transitions" (and this subset of bands "spin -1 subset").

For the scattering contribution to the $\dot{f}$ equations we use a relaxation time approximation (RTA) in which the band renormalization (shifts) are computed in a time-dependent and self-consistent fashion. In general, an RTA requires one to find, at each point in time, the Fermi distributions toward which the system would relax if no optical pulses were present. In the present case we assume that the spin $+1$ and spin -1 subsystems are not coupled by scattering processes since the spin flip times are typically in the ps rather than the fs regime [12]. Assuming, furthermore, that because of the high carrier density the fs-scattering processes are dominated by carrier-carrier scattering, the parameters characterizing the appropriate quasithermal equilibrium are (for each subsystem) the three band-offsets, two chemical potentials (one for the c band and one for the v bands), and the temperature. These parameters are uniquely and self-consistently determined by the zero-density band offsets, the two densities (electron and total hole density), and the total electron-hole kinetic energy. Note that the renormalization of the bands due to the occupation-dependent screened exchange interaction influences the relative shift of the hh and the lh band in a complicated way. Note also that the off-diagonal elements of the quasithermal hole distribution (denoted by the superscript "F" for Fermi) $f^F_{jj'}(\vec{k})$ ($j \neq j'$) do not vanish. This is

because we have written Eq. (2) in a basis that does not correspond to the true one-particle eigenstates of the system. A simple RTA of the form

$$\frac{d}{dt}\tilde{f}_{jj'}(\vec{k})\Big|_{\text{scatt}} = -\frac{1}{T_1}[\tilde{f}_{jj'}(\vec{k}) - \delta_{jj'}\tilde{f}^F_j(\vec{k})] \quad (5)$$

is possible only if the states labeled by j and $\vec{k}$ are the one-particle eigenstates. To indicate this, we use a "tilde" in Eq. (5), and the relation between $\tilde{f}$ and f is given by $\tilde{f}_{jj'} = \sum_{ii'} u^*_{ij} u_{i'j'} f_{ii'}$, where u_{ij} is the eigenvector matrix of $\mathcal{H}$ and we have suppressed the common index $\vec{k}$. Finally, in the scattering contribution to the equation for the optical polarization functions, we use a simple dephasing constant $1/T_2$ to simulate the effects of carrier-carrier scattering.

For the numerical investigation we use GaAs material parameters: $m_e = 0.067 m_0$ ($m_0 =$ electron mass in vacuum), $\epsilon_b = 12.7$ (background dielectric function entering the Coulomb potential), $\gamma_1 = 6.85$, $\gamma_2 = 2.1$, $\gamma_3 = 2.9$. We use a *hh*-*lh* splitting of 40 meV and a well thickness of 50 Å with infinite potential barriers. Unless otherwise noted, we use two 40 fs pulses (intensity FWHM), one left-handed circularly polarized pulse centered in time at $t = 0$ and in frequency at $\hbar\omega_0 = E_G - 180$ meV, and one right-handed circularly polarized pulse centered at $t = 40$ fs and $\hbar\omega_1 = E_G - 160$ meV. The peak amplitudes are $\mu E_0 = 212$ meV. Since the pulses have essentially no spectral overlap with the frequency region of the *hh* exciton, we can simulate the non-Lorentzian line shape of the optical transitions by choosing infinite or very long dephasing and relaxation times. The doping density is chosen to be 3.42×10^{11} cm^{-2} at a temperature of 20 K. We use, for simplicity, a time-independent screening model.

In Fig. 1 we show the temporal density response of the six bands involved for the idealized case of infinite T_1 and T_2. Except for an adiabatic transient population of the *c* bands, we see that the initial populations of the *hh* bands get transferred to the *lh* bands. Although the pulses have been chosen to simulate the conventional PTS-based transfer configuration for the spin $+1$ subset shown in Fig. 1(a), it apparently works as well for the spin -1 subset [Fig. 1(b)]. The fact that the transfer works at all in the semiconductor indicates clearly that the existence of an exact PTS is not necessary for the process to work, and, therefore, it is not surprising that it may work simultaneously for both spin subsets. In Fig. 2 we show the final density after the transfer as a function of $\hbar\omega_0$ with otherwise unchanged parameters for the spin $+1$ subset and, for comparison, quasi-cw results for an ideal 3-level system, in which the PTS is at $\hbar\omega_0 = E_G - 200$ meV. The large spectral overlap of the 40 fs pulses underlying Fig. 2(a) is not present in 2(b) and, hence, not a crucial factor in the transfer. As for the semiconductor, Fig. 2(a) shows a striking resemblance to the 3-level system despite the continuum of transition energies involved, the dynamic band gap shift, and the dynamic change of excitonic

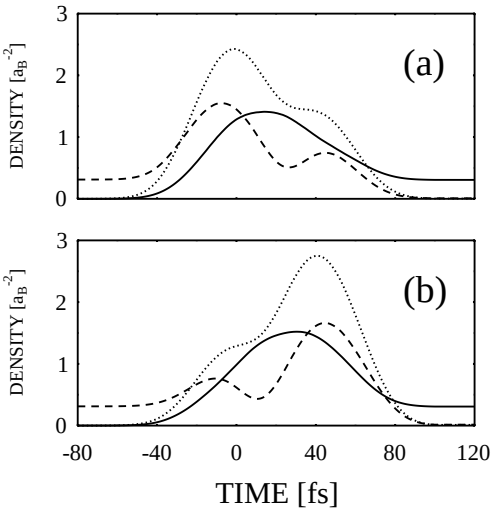

FIG. 1. Density vs time for the heavy-hole density (dashed line), light-hole density (solid line), and conduction band density (dotted line). The unit length is $a_B = 135$ Å. In (a), $s = 1/2$ and $j = 3/2$ and $-1/2$, respectively, and in (b) $s = -1/2$ and $j = -3/2$ and $+1/2$. Here dephasing and relaxation have been omitted.

Coulomb correlations (cf. Fig. 4). Calculations including finite dephasing and relaxation times yield, in general, a weakening of the transfer and an increase of final electron population. However, this does not necessarily mean that such incoherent processes destroy the actual population transfer mechanism, because the momentum states involved in the transfer are generally different from the ones involved in the dephasing-induced absorption process. This is especially true in the large detuning limit

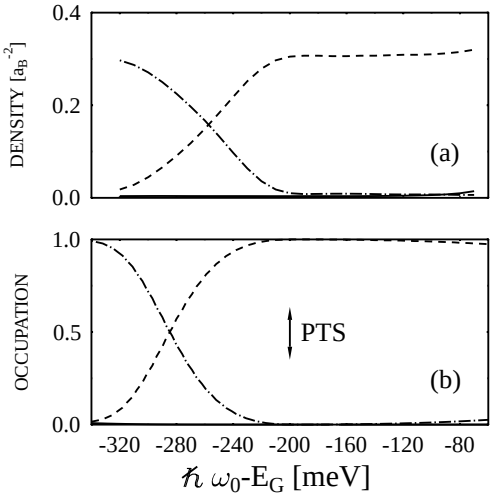

FIG. 2. (a) Final density of the $j = 3/2$ *hh* (dashed-dotted line) and the $j = -1/2$ *lh* (dashed line) population vs center frequency $\hbar\omega_0$ of the first pulse. The second pulse is centered 160 meV below E_G. (b) Corresponding results for a 3-level system with one transition at E_G and one transition 40 meV above E_G. "PTS" marks the population-trapped state.

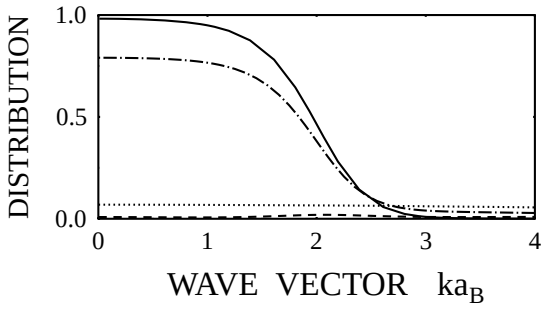

FIG. 3. Distribution function before and shortly after the population transfer. Solid line: distribution of hh ($j = 3/2$) before and lh ($j = -1/2$) after the transfer according to Fig. 1 (the distributions are essentially identical). The electron distribution is zero. For the case with dephasing and relaxation ($T_1 = T_2 = 600$ fs) the final lh distribution is shown as a dash-dotted line, and the final electron and hh distributions are shown as dotted and dashed lines, respectively.

considered here. Figure 3 shows the distribution functions before and after the transfer according to Fig. 1(a) as well as those for the case where incoherent processes are included. Whereas without incoherent processes the hh distribution before the transfer is practically the same as the lh distribution after the transfer, incoherent processes reduce slightly the number of light holes in the low-momentum region and create hh's and electrons spread out over a very large momentum region (in the case shown these distributions go to zero at about $k = 40a_B^{-1}$).

The general guidelines for the optimum parameter regime can be summarized as follows: the pulses should be detuned from the band gap only as much as necessary to avoid spectral overlap between the pulses and the interband absorption. Larger than necessary detuning would require stronger pulses for the effect to work, and therefore increase, in reality, the undesirable two-photon absorption.

We discuss briefly a possible way to measure the transfer. If one simply takes a linear absorption spectrum with linearly polarized light immediately after the transfer and compares it with a linear absorption spectrum taken before the transfer, one obtains, ideally, only a lh-exciton resonance before the transfer and only a hh-exciton resonance after the transfer. Of course, this ideal picture will be modified due to the plasma effects (screening, relaxation, etc.), but, as Fig. 4 shows, it is not completely destroyed by them. Note that the plasma-induced blueshift of the exciton resonances seen in Fig. 4 should not be taken as quantitative prediction because it is well known that it depends sensitively on the details of the screening model.

In summary, we have presented a theoretical analysis of population transfer in p-doped semiconductor quantum wells and identified a parameter regime that should allow for successful experimental verification of the transfer mechanism in the future.

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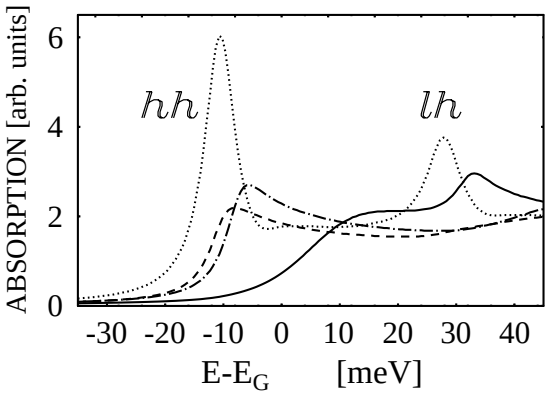

FIG. 4. Linear optical absorption spectra, including heavy-hole and light-hole exciton resonances: undoped quantum well (dotted line), doped quantum well before (solid line) and after the adiabatic transfer [dash-dotted line: according to Fig. 1; dashed line: dephasing and relaxation ($T_1 = T_2 = 600$ fs) included]. In all cases, the dephasing time used in the calculation of the linear spectra is $T_2 = 200$ fs.

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