Coherent Control of Light Absorption and Carrier Dynamics in Semiconductor Nanostructures

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It is shown theoretically that light absorption in semiconductor nanostructures can be manipulated by coherent microwave fields. In suitably designed double wells, amplitude and phase of a microwave field allow control of both light absorption and the dynamics, i.e., tunneling versus localization of electrons. Depending on the photon energy of the pump pulse, either enhancement or reduction of net absorption can be achieved. [S0031-9007(97)04379-2]

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Young's double slit experiment is not only a key experiment to demonstrate wave behavior but also provides the basis for coherent control of quantum-mechanical processes. Coherent control of optical excitation of atoms and molecules, as well as simple chemical reactions, has been demonstrated in the last decade [1,2]. Several schemes for lasing without inversion and state trapping have been proposed in atomic systems. They are based on quantum coherence between atomic levels which is mediated by some form of coupling, such as to a continuum (Fano resonance) or a microwave field [3,4]. Indeed, net photon gain without inversion from a beam of sodium atoms has recently been reported [5].

In solids, decoherencing times for charge carriers typically lie below 1 picosecond and thus are several orders of magnitude shorter than in atoms or molecules. Nevertheless, progress in subpicosecond laser spectroscopy has recently made possible first observation of optically induced coherent phenomena, such as the decay of spin polarization, charge oscillations in double wells, and Bloch oscillations in superlattices [6–9]. More recently, coherent control of photocurrent has been demonstrated [10]. A generation of revolutionary new optoelectronic solid-state devices can be envisioned to operate on the basis of the quantum interference principle.

While the Franz-Keldysh and (quantum-confined) Stark effect are well-known examples for manipulation of light absorption in solids by external electric fields [11,12], in this Letter we show that dc microwave (MW) fields can be used to *coherently* control light absorption and wave packet dynamics in semiconductor nanostructures. As a specific example we consider an asymmetric GaAs-AlGaAs double well (DW), as shown in Fig. 1. A static external electric field is used to control the energy separation between the two lowest electron subbands which, in addition, are coupled resonantly by a dc MW field. Using Young's double slit principle of two competing pathways, we consider optical transitions between the top hole subband $|H\rangle$ and the two lowest electron subbands $|-\rangle$ and $|+\rangle$. A pump pulse photon can be absorbed by transferring an electron from the top hole subband to either of the two electron subbands $|-\rangle$ and $|+\rangle$. The coupling between the latter depends both on amplitude and phase of the MW field. While the phase of the MW field is of no consequence for the dynamics of the two-level system $|-\rangle$ and $|+\rangle$, when probed by a "spectator level," such as the top hole subband $|H\rangle$, the phase of the MW field matters provided that the pump pulse duration is shorter than the period of Rabi oscillations between $|-\rangle$ and $|+\rangle$. As will be shown below, amplitude and phase of the latter allow control of the generation of electron-hole pairs (excitons) by a subpicosecond pump pulse.

Our theoretical analysis is based on a recently developed microscopic theory of coherence and phase breaking semiconductor multiband systems [13,14]. It incorporates phase breaking on a nonphenomenological basis which is essential for accurate assessment of the decay of interband polarization ("phase breaking") which is generated by the light fields during the excitation process. The dynamics of carriers in the DW is evaluated on the basis of Boltzmann-Bloch (BB) equations within a three subband model using the single-particle basis $|L\rangle$, $|R\rangle$, and $|H\rangle$, for left-well electron, right-well electron, and top-hole subband, respectively. The carrier-carrier Coulomb interaction is

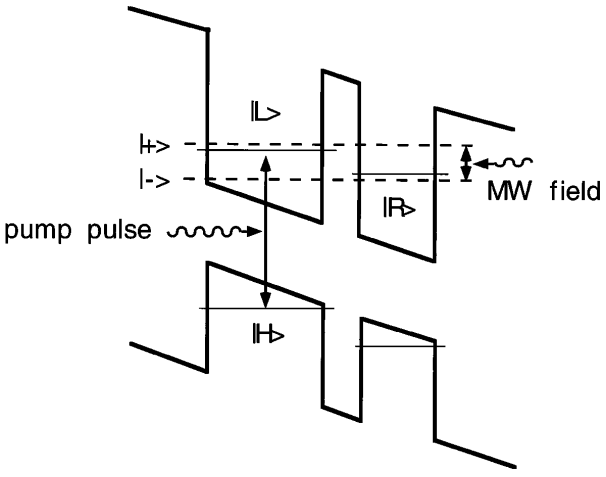

FIG. 1. An asymmetric double well in an external electric field. A dc microwave field couples the two electron subbands + and −. A pump pulse generates electron-hole pairs across the gap.

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treated within the screened Hartree-Fock approximation to the self-energy, giving rise to both renormalization (exciton effects) and scattering (phase breaking) [14]. Both pump and MW field are treated classically using Hamiltonians of the form

$$H_{\text{pump}} = \sum_k \{a_p(t)\cos(\omega_p t)\hat{b}_L(k)^\dagger \hat{b}_H(K) + \text{H.c.}\}$$

and

$$H_{\text{MW}} = \sum_k \{a_{\text{MW}}\cos(\omega_{\text{MW}} t + \phi)\hat{b}_+(k)^\dagger \hat{b}_-(k) + \text{H.c.}\},$$

where $\hat{b}_\alpha(k)$ are single electron operators and a_i are the electron-light coupling matrix elements within the dipole approximation. These Hamiltonians are treated exactly in the kinetic equations and lead to non-Markovian carrier excitation. Note that we avoid the rotating-wave approximation for the light fields. While well justified for the pump pulse, the latter leads to some quantitative differences in the carrier dynamics when used for the MW field in the present case. Terms in the light-matter coupling which are quadratic in the vector potential lead to intraband coupling and are neglected here.

Numerical results shown in the figures have been obtained within a Runge-Kutta algorithm for an asymmetric DW consisting of a left 145 Å wide well and a right 100 Å narrow well, separated by a 25 Å $Al_{0.3}Ga_{0.7}As$ barrier. A static external field is used to lower the $|R\rangle$ electron subband below the $|L\rangle$ electron subband, ensuring that primarily the top hole subband is involved in the excitation process. The subband splitting is kept below the optical phonon energies of the structure.

A 100 fs Gaussian pump pulse of typically 10^6 W/cm^2 peak intensity excites electron-hole pairs across the main energy gap. The intensity amplitude of the dc MW field is varied from zero to about 8×10^6 W/cm^2. Its phase ϕ is measured relative to the time ($t = 0$) of maximum pump pulse intensity. Figure 2 shows the absorption spectrum, i.e., the number of *e-h* pairs generated as a function of photon energy of the pump pulse, relative to the nominal energy gap. Here, the subband splitting is 25 meV. A profound modification of the absorption profile by the MW field is evident. For zero MW field, solid line, the main absorption occurs at the direct exciton peak, about 15 meV below the nominal band edge. The indirect exciton peak is much weaker and, due to the spectral width of the pump pulse, is buried beneath it, as discussed previously [14]. A resonant microwave field of about 1×10^6 W/cm^2 (Rabi frequency $\approx 2 \times 10^{13}$ Hz) and phase $\phi = 0$, dash-dotted line, shifts and broadens the absorption peak, leading to reduced absorption at the direct exciton peak and increased absorption below the exciton peak. At higher MW intensity two peaks appear, dotted line, giving rise to significant enhancement of "below-bandgap" absorption, while an absorption minimum occurs near the position

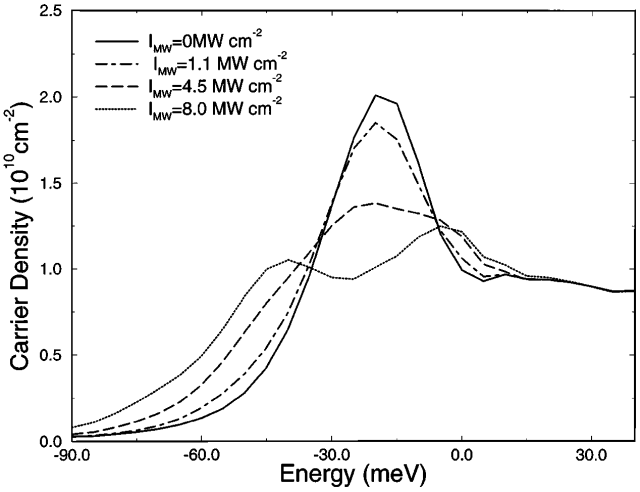

FIG. 2. Absorption profile of a double well versus pump photon energy, relative to the gap energy, for an electron subband splitting of 25 meV.

of the original exciton peak. A second, smaller peak emerges above the original direct exciton peak.

The efficiency of the MW field is determined by its phase, as is evident from the following two figures which were obtained for a subband splitting of 10 meV and MW peak pump intensity of 2.8×10^5 W cm^{-2}. Figure 3 shows the number of *e-h* pairs as a function of time for a pump pulse tuned in resonance with the direct exciton (15 meV below the conduction band edge) and phase $\phi = 0$ (solid line), $\phi = \pi/4$ (dot-dashed line), and $\phi = \pi/2$ (dashed line), in comparison to the result without MW field (dotted line). The variation of generated *e-h* pairs versus MW phase is shown in Fig. 4. It can be seen that variation of phase between zero and $\pi/2$ allows

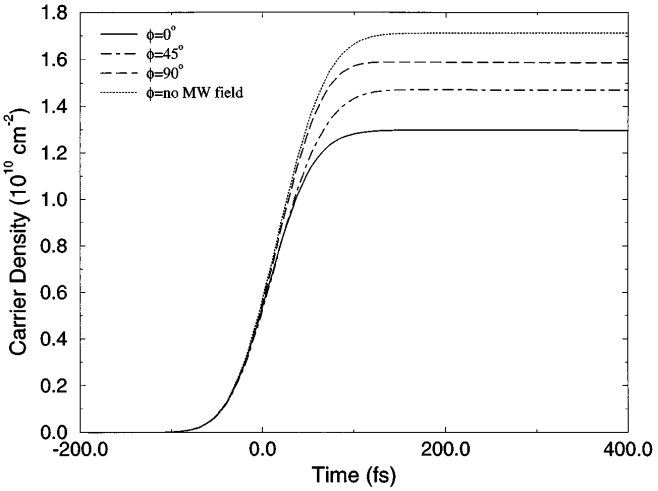

FIG. 3. Number of photogenerated electron-hole (*e-h*) pairs versus time for an electron subband splitting of 10 meV and MW intensity 2.8×10^5 W cm^{-2}. Solid line: MW phase $\phi = 0$; dot-dashed line: MW phase $\phi = \pi/4$; dashed line: MW phase $\phi = \pi/2$; dotted line: without MW field.

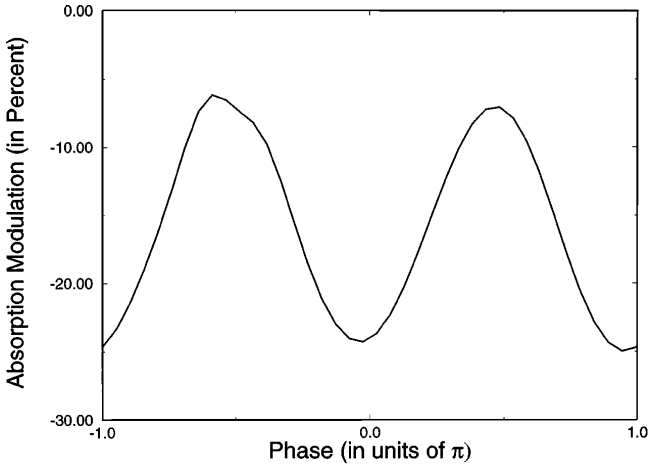

FIG. 4. Absorption modulation due to phase variation of the MW field in percent relative to absorption without MW field.

variation of absorption by about 20%, while the maximum reduction in absorption is about 25% in the present case.

Figure 5 displays the number of photogenerated carriers in the left well versus time, demonstrating that the presence of a MW field significantly influences the dynamics and distribution of carriers between left and right well. In the absence of the MW field (dotted line), most of the electrons are generated in the left well and, due to the separation of electron subbands, charge oscillations are weak. With the MW field present, electrons are more evenly distributed between left and right well, i.e., both direct and indirect excitons are generated. Furthermore, charge oscillations, whose amplitude and temporal evolution is controlled by the phase of the MW field, are induced between left and right well. While damping of charge oscillations is evident in Fig. 5, on a picosecond time scale and for present carrier concentrations, scattering processes are found to have negligible influence on the present effects. At higher carrier concentrations we find substantial broadening of the absorption peaks due to carrier screening of exciton features. If the pump pulse duration is increased the phase sensitivity of the effect disappears, while a dependence on the amplitude of the MW field persists. Similarly, reduction of pump pulse duration enhances phase sensitivity.

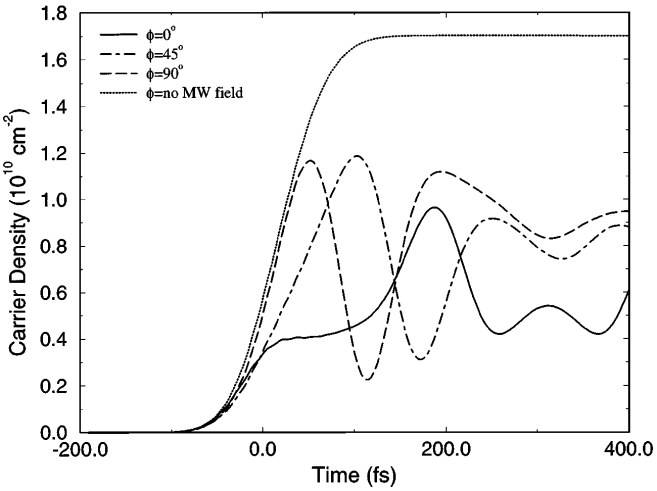

FIG. 5. Number of photogenerated electrons in the left (wide) well versus time for an electron subband splitting of 10 meV and MW intensity 2.8×10^5 W cm^{-2}. Solid line: MW phase $\phi = 0$; dot-dashed line: MW phase $\phi = \pi/4$; dashed line: MW phase $\phi = \pi/2$; dotted line: without MW field.

The present results can be explained qualitatively within a driven two-level system and the electronic properties of the system. In the absence of the MW field, the pump pulse predominantly couples the hole subband of the wide well to the upper $(+)$ subband which, due to the applied bias, is a superposition of the lowest left well subband and a small admixture of the right well subband. Thus, electrons (direct excitons) are generated predominantly in the left (wide) well. The resonant MW field hybridizes the lower $(-)$ and upper $(+)$ electron subband transferring oscillator strength from the upper state to the lower, allowing significant formation of indirect excitons. This explains qualitatively the features of Fig. 2 and the final distribution of electrons within the wells, Fig. 5. The phase sensitivity can be understood by recalling that, within the RWA, a resonantly coupled two-level system undergoes Rabi oscillations between its uncoupled eigenstates, which here are the two lowest subbands of the double well, renormalized and coupled by Coulomb effects. The phase of the MW field determines during which part of this cycle the pump field arrives at the sample. Thus, as the pump pulse duration exceeds the Rabi frequency of the MW field, phase sensitivity is lost, while renormalization effects due to the MW field persist. This picture is qualitatively correct also when the counter-rotating MW field contribution is included. However, the counter-rotating contribution introduces higher frequency contributions to the simple picture of Rabi oscillations in the carrier dynamics, as evident in Fig. 5. An alternative picture which explains the phase sensitivity is obtained by inspection of the photon Boltzmann-Bloch equation which takes into account phase coherence (interband polarization) in the carrier system [14,15]. It can be shown that the MW field controls (the sign of) the $+/-$ interband polarization and, hence, the absorption process.

In summary, we have demonstrated that amplitude and phase of a coherent microwave (MW) field can be used to control the near-bandgap light absorption of semiconductor nanostructures. For a pump pulse duration below the Rabi period, proper structural engineering allows almost complete control of the final carrier concentration by the phase of the MW field between the MW-free absorption and the optimum case in the presence of the MW field. Simultaneously, the dynamics of charge carriers can be manipulated by a MW field. A resonant MW field enhances charge oscillations (charge transfer) between

quantum wells whose amplitude can be controlled by the MW phase. The intensity of the MW field controls the final ratio of the number of direct versus indirect photogenerated excitons. The carrier-carrier Coulomb interaction limits the effects predicted here to low and moderate carrier densities which ensure the presence of excitonic features, as well as comparatively long decoherencing times.

It should be pointed out that the present system resembles the atomic V-scheme predicted to give rise to "state trapping" [4]. However, here we consider effects directly on a subpicosecond pump pulse and exploit many-body effects associated with the carrier-carrier Coulomb interaction in electron subbands. The physical processes discussed here are not limited to double wells. A variety of intersubband transitions in semiconductor nanostructures can be controlled in this fashion. In particular, the use of overlapping subpicosecond pump and MW *pulses* allows coherent control of (nonadiabatic) charge transfer between quantum wells, similar to experiments in atomic and molecular physics [16].

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