invited

Coherent Control of Interband Transitions in Semiconductor Quantum Wells

Xuedong Hu and Walter Pötz

Physics Department
University of Illinois at Chicago, 845 W. Taylor St., Chicago, IL 60607, U.S.A.
Phone: (312) 996-5048, Fax: (312) 996-9016
E-mail:wap@uic.edu

1. Introduction

Coherent control has emerged as a tool to control fundamental processes in semiconductors, such as electrical current, optical absorption, and optical gain [1-4]. It is based on the fundamental principle of quantum interference between competing pathways from an initial state to a final state. As such, coherent control is limited to a time regime which is below the characteristic time of those competing channels over which one has no or only limited control, such as scattering processes, recombination and Auger processes, etc. As some of the latter can occur on a subpicosecond time scale, there are two essential ingredients for coherent control in semiconductors: precise microfabrication to tailor the electronic structure and, in most cases, subpicosecond laser spectroscopy. The former not only serves to select desired optical transition energies but also to engineer (dipole) matrix elements and scattering rates. In particular, unwanted transition channels should be suppressed as much as possible. To our knowledge, all coherent control experiments have relied on optical manipulation of inter(sub)band transitions. However, manipulation of absorption line shapes using Fano resonances has been reported by several groups [5,6].

In this invited paper we will briefly review our theoretical efforts to predict and evaluate possible coherent control schemes in semiconductor heterostructures, emphasizing some of our most recent results. We will show that, given suitable light sources, coherent control of optical absorption, optical gain, and coherent control of phonon emission can be achieved in relatively simple semiconductor heterostructures. One very versatile, yet simple, structure is a semiconductor double well which has been used to demonstrate coherent charge oscillations and quantum cascade lasing [7,8].

Two simple coherent control schemes are shown in Fig. 1. Fig. 1(a) shows the classical control scheme adopted from physical chemistry and is based on interference between single– and two–photon (in some cases, three–photon) absorption [9]. It is this scheme which

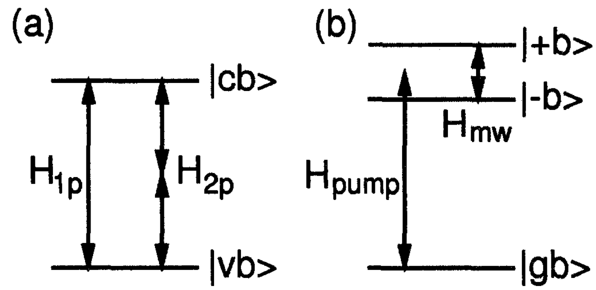

Figure 1: Schematic representation of two coherent control schemes: (a) traditional scheme using interference between single– and multi–photon absorption; (b) a coherent control scheme based on a three–level system. Horizontal bars and vertical arrows indicate electronic bands and light fields, respectively.

has been used to coherently control photocurrent in bulk GaAs and GaAs/AlGaAs quantum wells [1,2].

The second scheme is based on a three–level model (here three–subband model), consisting of a single level and a level doublet. The doublet is driven resonantly by a coherent cw light field. This scheme has originally been proposed as a scheme for lasing without inversion and state trapping [10]. More recently it has been shown that the *phase* of this driving field can be used to coherently control transitions between the singlet and the doublet [3,4]. Yet other experiments have utilized subpicosecond pump–probe schemes, whereby the first pulse sets up coherence ("interband polarization") in the system which influences the response to the second [11].

2. Theory

A truly microscopic theoretical picture of coherent control and its limitations in semiconductors must be derived from the full semiconductor many–body problem. There are two main approaches which have been used in this connection, the non-equilibrium Green's function approaches and the density matrix approach [12,13]. Both approaches have their merits and a combination of them proves to be most useful, both for an evaluation of validity of a particular approximation and to obtain certain approximations most efficiently [14]. The kinetic equations which we use are usually referred to as semiconductor Bloch equations or Boltzmann-Bloch equations. They use density matrix elements $f_{\alpha\beta}(\mathbf{k},t) \equiv \langle\beta\mathbf{k}|\rho(t)|\alpha\mathbf{k}\rangle$ to characterize the dynamics of single-particle ensemble averages, such as particle and current densities of the system. The density matrix elements obey first-order differential equations in time which, owing to particle-particle interactions, become nonlinear in $f_{\alpha\beta}(\mathbf{k},t)$. Light fields are included classically within the dipole approximation, but omitting the rotating-wave approximation. Many-body effects are included within the screened Hartree-Fock approximation to the self-energy. Calculations are limited to the low density regime of less than a few 10^{10} carriers per cm^2 to keep carrier-carrier scattering weak. The latter is important when dealing with schemes based on intersubband transitions.

The set of generalized Boltzmann-Bloch equations is evaluated numerically. Density matrix elements are discretised in $k=|\mathbf{k}|$. Typically 60 mesh points are used for energy intervals of 0.15 eV within a given (sub)band. A cubic spline is used for interpolation. $\mathbf{k}$-integrals are evaluated using Gauss's or Romberg's method [15]. The computation of the time evolution is carried out within the interaction picture. The dynamics of the coupled free-particle Hamiltonian is treated exactly, whenever possible, and takes care of the high-frequency components. The slow time-dependencies from the external light fields and many-body effects are treated within a Runge-Kutta scheme [15].

If the light pulse duration approaches the inverse (LO) phonon period ($\approx 55fs$ in GaAs), memory effects become important for the electron-phonon interaction. Energy-conserving delta functions in the BB equations need to be replaced by convolutions in time [16]. For our present control schemes, however, a pulse duration of 200 fs proves to be ideal (note that for short pulses the spectral width of the pulse becomes comparable to a subband splitting characteristic of 10 nm heterostructures). Hence, phonon memory effects do not play a role in the coherent control processes which will be discussed here.

3. Coherent Control of Charge Oscillations in Semiconductor Double Wells

Coherent charge oscillations in semiconductors provided one of the first demonstrations that coherent carrier dynamics (transport) can be optically induced in semiconductors [7]. Based on scheme 1(a) and a subband doublet in the conduction band, the use of two coherent light fields allows control of the amplitude of charge oscillations and, consequently, the emission of THz radiation [17].

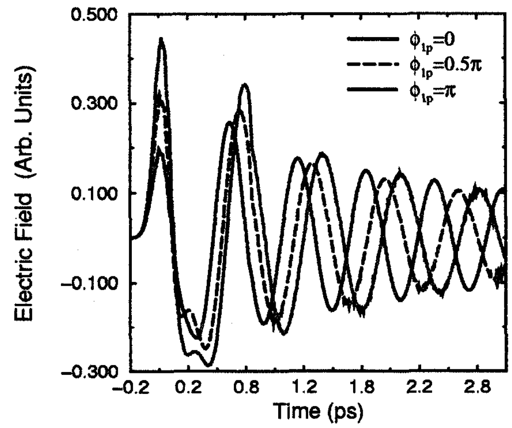

Figure 2: Emitted electric field versus time for ϕ_{2p}=0.

Following the original experiment, we consider an asymmetric GaAs-AlGaAs quantum well whose lowest two electronic subbands are tuned into resonance [7]. The first light field E_{1p}, is at resonance with the direct excitonic transition with the top heavy-hole subband, promoting single photon absorption and emission. A second, phase-matched light field E_{2p} has half the frequency of E_{1p}, thus, promoting two-photon absorption and emission between the top heavy-hole subband and the electron subband doublet. The relative phase between these two light fields determines the interference term between single- and two-photon absorption. Experimentally, E_{1p} can be obtained sending part of E_{2p} through a nonlinear crystal. Field intensities are such that the two pump pulses individually generate 0.5×10^{10} carriers per cm^2, corresponding to identical slits in Young's double slit experiment for best contrast in the "interference fringes".

In Fig. 2 we show the radiated electric field in the THz regime as a function of time for phase ϕ_{1p}=0, 0.5π, and π and ϕ_{2p}=0. Particularly in the onset of oscillations, clear evidence of the phase dependence is given. The latter arises from different exciton generation rates. A high generation rate, relative to the tunnel frequency, leads to a larger net amplitude of the ensemble of excitons as they tend to oscillate more closely in phase. Consequently, higher oscillation amplitudes also mean higher carrier densities and more carrier-carrier scattering. For phases with originally large charge oscillations there is more damping due to the carrier-carrier interaction.

Fig. 3 shows the maximum electric field versus ϕ_{1p} (full circles), as well as a cosine fit to the data. A period of 2π is clearly evident. Variation of ϕ_{2p}, not shown in Fig. 3, shows a period of π, confirming that this control process indeed arises from interference between single- and two-photon absorption.

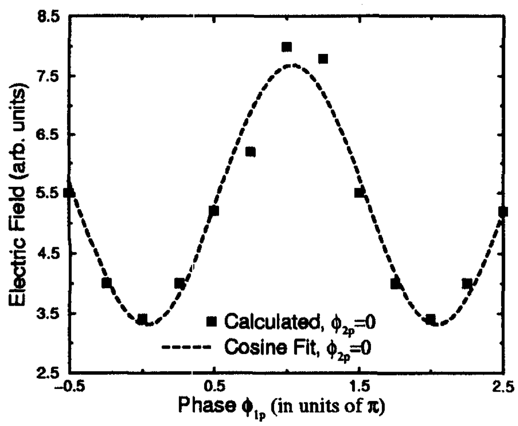

Figure 3: Amplitude of the emitted electric field versus phase ϕ_{1p} (full circles). Dashed line is a cosine fit to the data points.

4. Coherent Control of Phonon Emission in Intersubband Transitions

We will now give an example for control scheme 1(b). Again we use a double well to realize the desired subband structure. However, there are significant differences to the previous case. We consider only electronic subbands. Using two adjacent GaAs/AlGaAs wells of unequal depth, one can obtain a lowest subband whose wave function is well confined to within the deep (wide) well below a subband doublet which arises from hybridization of the lowest subband of the shallow (narrow) well and the second subband of the wide well. The lowest–subband/doublet separation is about 40 meV to allow for effective LO phonon coupling. Originally, the carrier density in the lowest subband is 10^{10} cm^{-2}. This may be achieved, for example, by remote doping. The upper doublet is initially empty. A 200 fs Gaussian pump pulse excites electrons from the lowest subband into the doublet. Eventually, electrons return into the ground subband, predominantly via LO phonon emission. The purpose here is to study whether a mw which resonantly couples the doublet subbands will affect the phonon emission process. To achieve this goal, a continuous–wave (cw) mw field about 1MW/cm^2 is used. In order to isolate coherent control of phonon emission from absorption modulation, see Ref. [3], we turn on the mw field with a time delay of 300 fs relative to the pump pulse. This ensures that electron–hole pair generation by the 200fs pump pulse occurs prior to the presence of the mw field. Both LO–phonon scattering and carrier–carrier Coulomb scattering is included here. In addition, we account for confined LO phonon modes only as well widths are $\geq$ 10nm.

Fig. 4 shows that the return rate into the subband can be controlled to a large degree by variation of the phase of the mw field, as soon as the mw field is turned on. Again, this is accomplished by controlling quantum interference between two processes, *i.e.*, LO phonon emission and mw photon emission and absorption. The latter establishes interband polarization in the subband doublet which influences LO phonon emission. Note that this control process works best for pump pulses whose duration is of the same order as the phonon emission time in the system (and, hence, is large compared to the lattice vibration period of about 55 fs associated with an LO phonon). Note also that here we have no direct control over when a phonon gets emitted, whereas in the previous example one has control over the phase of both pump fields.

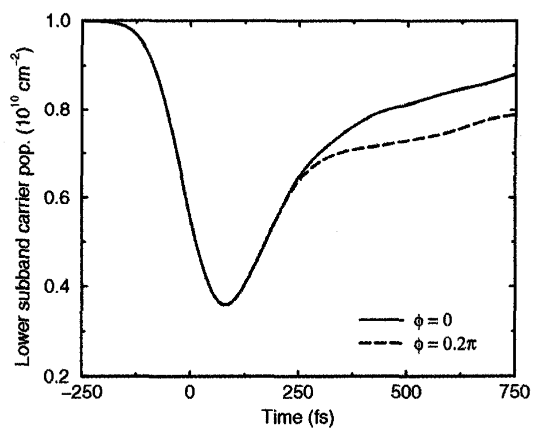

Figure 4: Carrier density in the ground subband $|gb\rangle$ versus time. Time zero marks the arrival of the peak of the pump pulse. ϕ denotes the phase of the mw field (relative to the time of arrival of the pump pulse).

In this calculation carrier-carrier scattering is included to show that it does not eliminate the mw phase dependence of the phonon emission rate. A detailed analysis of this phase dependence of the phonon emission rate shows that variations of the effective interband relaxation time between 400 fs and 1 ps are obtained with mw intensities of 1 MW cm^{-2}. For a cw mw field our study predicts coherent control of both absorption as well as LO phonon emission. Moreover, the period of modulation is π, rather than 2π as in Fig. 3. This allows a simplified interpretation of the present effect. A transition from one of the upper subbands to the ground–state subband can occur either directly via LO phonon emission or via LO phonon emission in conjunction with emission and absorption of a mw photon. For this second–order process in the mw field to provide an effective pathway into the ground subband a high mw intensity is required.

5. Summary and Conclusions

In semiconductors coherent control can be used to manipulate fundamental processes, such as light absorption, optical gain, tunneling, and phonon scattering. The control schemes presented here are hoped to provide stimulation for experimental verification and to serve as examples of how future ultrafast (*i.e.*, subpicosecond) semiconductor devices may operate on the principle of quantum interference. There is little doubt that it will not be practical to build devices whose control is performed by high–intensity external mw fields, as in the second example. However, more practical alternatives may be provided by the use of built–in mw microcavities or Fano–resonance–based control schemes which are controlled by electric fields.

Acknowledgment: We would like to thank the U.S. Army Research Office for financial supporting of this work, as well as W. A. Schroeder for discussions regarding experimental aspects.

References

1. E. Dupont, P. B. Corkum, H. C. Liu, M. Buchanan, and Z. R. Wasilewsky, Phys. Rev. Lett. **74**, 3596 (1995).

2. A. Hachè, Y. Kostoulas, R. Atanasov, J. L. P. Hughes, J. E. Sipe, and H. M. van Driel, Phys. Rev. Lett. **78**, 306 (1997).

3. W. Pötz, Phys. Rev. Lett. **79**, 3262 (1997); X. Hu and W. Pötz, Phys. Stat. Sol. (b) **204**, 350 (1997).

4. X. Hu and W. Pötz, Appl. Phys. Lett. **73**, 876–878 (1998).

5. H. Schmidt, K. L. Campman, A. C. Gossard, and A. Imamoglu, Appl. Phys. Lett. **70**, 3455-3457, (1997).

6. J. Faist, F. Capasso, C. Sirtori, K. W. West, and L. N. Pfeiffer, Nature **390**, 589–591 (1997).

7. K. Leo, J. Shah, E. O. Göbel, T. C. Damen, S. Schmitt–Rink, W. Schäfer, and K. Köhler, Phys. Rev. Lett. **66**, 201 (1991); H. G. Roskos, M. C. Nuss, J. Shah, K. Leo, D. A. B. Miller, A. M. Fox, S. Schmidt-Rink, and K. Köhler, Phys. Rev. Lett. **68**, 2216 (1992).

8. J. Faist, F. Capasso, D. L. Sivco, A. L. Hutchinson, and A. Y. Cho, Science **264**, 553 (1994).

9. See, *e. g.*: R. J. Gordon, Annual Reviews of Physical Chemistry **48**, 595 (1997).

10. M. Scully, S. Y. Zhu, and A. Gavrielides, Phys. Rev. Lett. **62**, 2813 (1989).

11. A. P. Heberle, J. J. Baumberg, and K. Köhler, Phys. Rev. Lett. **75**, 2598 (1995).

12. J. Rammer and H. Smith, Rev. Mod. Phys. **58**, 323 (1986).

13. See, *e. g.*: W. H. Louisell, *Quantum Statistical Properties of Radiation*, (J. Wiley, New York, 1990).

14. W. Pötz, Phys. Rev. B **54**, 5647 (1996); U. Hohenester and W. Pötz, Phys. Rev. B **56**, 13177–13189 (1997).

15. W.H. Press, S.A. Teukolsky, W.T. Vetterling, and B.P. Flannery, *Numerical Recipies* (Cambridge University Press, Cambridge, 1986).

16. X. Hu and W. Pötz, in *Proc. Int. Workshop on Coherent Control*, Chicago, May 1998, edited by W. Pötz and W. A. Schroeder, (Kluwer Publishing), in press.

17. W. Pötz, Appl. Phys. Lett. **72**, 3002–3004 (1998).