Coherent Manipulation of Phonon Emission Rates in Semiconductor Heterostructures

Xuedong Hu* and Walter Pötz
Department of Physics, University of Illinois at Chicago, 845 W. Taylor Street, Chicago, Illinois 60607
(Received 15 October 1998)

Quantum interference between electron-lattice and electron-light interaction allows coherent control of optical phonon emission rates in semiconductors. We use a microscopic theoretical analysis to show that the longitudinal-optical-phonon-mediated return rate of electrons into the lowest electron subband of a GaAs/AlGaAs heterostructure can be controlled by the phase of a microwave field which resonantly couples two upper electronic subbands of the heterostructure. For the structure investigated we predict a variation of the relaxation rate by a factor of 2 between 400 and 900 fs. This effect may be interpreted as quantum interference between a single-phonon emission process and a single-phonon–two-photon process. [S0031-9007(99)08923-1]

PACS numbers: 72.10.Di, 73.40.Gk, 78.45.+h, 78.66.Fd

Optical phonon scattering plays a fundamental role in the nonequilibrium carrier dynamics of semiconductors, influencing carrier transport, carrier thermalization, and optical properties [1]. For electrons in polar semiconductors, longitudinal-optical (LO) phonons frequently provide the dominant inelastic scattering mechanism. Optical phonon emission times are generally perceived as a given material property. Careful material selection, combined with structural design, is exercised to "engineer" LO phonon scattering in electronic devices. In this Letter we show that quantum interference between competing pathways can be used to coherently control LO phonon emission rates in electron intersubband transitions. Traditional coherent control has utilized quantum interference between single- and multiphoton absorption to control the symmetry of the final state and/or the absorption cross section ("reaction cross section") [2]. To the lowest order of Fermi's golden rule, the transition rate between an initial state $|i\rangle$ and a final state $|f\rangle$ is proportional to [3]

$$|\langle i|H_1 + H_2|f\rangle|^2, \tag{1}$$

where H_1 and H_2 may be, for example, the Hamiltonians for single- and two-photon absorption. The relative phase between H_1 and H_2 controls the interference term in Eq. (1) and offers a means to control the transition rate. In more general terms, when there are two or more competing quantum processes, their net effect does not equal the sum of the effects from the individual processes. This also pertains to perturbations which do not overlap in time, as long as they are separated by less than the characteristic phase breaking ("dephasing" or "decoherencing") time of the system [4]. Moreover, it is sufficient to be able to control the phase for either H_1 or H_2 as long as changing the phase of one perturbation does not appreciably influence the phase of the other. In terms of a density matrix description, the response of the system is determined not only by carrier distribution functions, i.e., the diagonal elements of the one-particle density matrix, but also inter(sub)band polarizations, represented by the off-diagonal density matrix elements [5].

In this paper we demonstrate that phonon emission rates in semiconductor heterostructures can be manipulated using quantum interference between single-phonon emission and a process which involves single-phonon emission combined with emission and absorption of a photon provided by an external microwave (mw) source. Indeed, the phase of the mw field allows substantial control of the net phonon intersubband emission rate.

Consider a double well (DW) of unequal well depth such that the lowest electron subband of the shallow well is near resonance with the second electron subband of the deep well. Such a structure is sketched in Fig. 1. A built-in or external electric field may be used to fine-tune or modify the subband splittings. Subband levels and electron wave functions in the growth direction of the system were determined within an envelope function model. Parabolic energy dispersion with equal effective masses of $0.067m_0$ were used for all three electron subbands. The $\pm$ subband splitting is adjusted to 10 meV. The subband doublet is positioned about 40 meV above the lowest electron subband of the DW. The ground state subband is well confined to within the deep wide well. Well widths are of the order of $\geq$10 nm. The barrier thickness is about 2 nm. This design allows efficient transfer of electrons between the doublet and the lowest subband by optical phonon emission on a time scale of about 500 fs. A coherent mw field resonantly couples the two subbands of the doublet. Initially, the lowest subband is populated by 10^{10} electrons per cm^2. This may be achieved, for

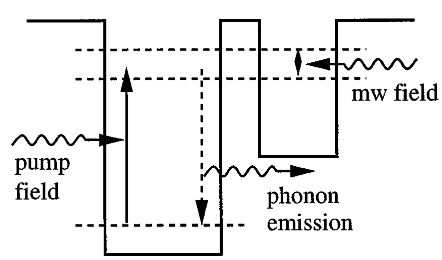

FIG. 1. Schematic representation of the electronic subband structure of the asymmetric GaAs-AlGaAs double well.

0031-9007/99/82(15)/3116(4)$15.00

example, by remote doping in the adjacent barrier layers. A subpicosecond pump pulse excites electrons from the lowest subband into the subband doublet and the return of electrons into the lowest subband (predominantly) due to LO phonon emission is monitored as a function of time.

For the study of this relaxation process we use a Markovian version of the Boltzmann-Bloch equations [6]. This is a well-justified approximation for the present analysis, as will be discussed in detail below. Within this approach and considering sufficiently low sample temperatures so that LO phonon absorption between the lowest subband and the subband doublet may be neglected, the rate of electron transfer from the upper subbands to the lowest subband due to spontaneous LO phonon emission is

$$\frac{d}{dt} f_{00}(\mathbf{k},t) = \frac{2\pi}{\hbar} \mathrm{Re}\Bigg\{ \sum_{\mathbf{q};\nu,\nu'=\pm} M_{0,\nu'}(\mathbf{q}) [M_{\nu 0}(-\mathbf{q}) \delta(e_0(\mathbf{k}) - e_\nu(\mathbf{k}+\mathbf{q}) + \hbar\omega_{\mathrm{LO}}(\mathbf{q})) f_{\nu\nu'}(\mathbf{k}+\mathbf{q}) \times (1 - f_{00}(\mathbf{k})) + M_{0\nu}(-\mathbf{q}) \delta(e_0(\mathbf{k}+\mathbf{q}) - e_\nu(\mathbf{k}) + \hbar\omega_{\mathrm{LO}}(-\mathbf{q})) f_{0\nu}(\mathbf{k}) f_{0\nu'}(\mathbf{k}+\mathbf{q})] \Bigg\}. \qquad (2)$$

Here $M_{0\nu}(\mathbf{q})$ denotes the electron-phonon matrix element. $f_{ij}(\mathbf{k},t)$ are the density matrix elements associated with single particle energy states $e_i(\mathbf{k})$, where i, j are band indices and $\mathbf{k}$ is the two-dimensional electron wave vector. The second set of terms on the right-hand side (rhs) of Eq. (2) is a quantum correction due to intersubband polarization between the lowest subband and one of the two doublet subbands. Here it is small because it is established essentially by the weak *pump* pulse. The first contribution on the rhs of Eq. (2) shows that the intersubband polarization f_{+-} of the subband doublet $\pm$ enters the otherwise standard expression for phonon emission, provided that *both* bands of the doublet couple to the ground subband. In analogy to the use of identical slits in Young's double slit experiment, equal coupling strength to both bands in the doublet leads to the best results for coherent control. For the present structure this condition implies small energy splitting for the subband doublet. Equation (2) identifies manipulation of $f_{+-}(\mathbf{k},t)$ as a means for coherent control of phonon emission for electron transitions between the upper bands and the lowest subband. Here we use a strong coherent mw field to resonantly couple the subbands and generate $\pm$ intersubband polarization. This makes the $f_{+-}(\mathbf{k},t)$ term the dominant quantum correction in Eq. (2). Nevertheless, all terms of Eq. (2) were included in our analysis. If the period of Rabi oscillations of $f_{+-}(\mathbf{k},t)$ induced by the mw field is longer than the LO phonon period, the phase of the mw field determines the electric field strength, and more importantly, the magnitude of $f_{+-}(\mathbf{k},t)$ intersubband polarization during the phonon emission process.

The LO phonon spectrum of such a system is quite complex and consists of modes which are largely confined to individual GaAs wells, the AlGaAs modes in the barrier regions, and interface modes. Using well widths $\geq$10 nm renders the interface modes of secondary importance to confined modes [7]. Because of the effective localization of the lowest subband wave function to within the wide well, barrier modes provide only small coupling to the doublet states. Hence, the dominant mode which couples the subband doublet to the lowest subband is a confined mode associated with the wide well [7]. This mode (with an energy of 36 meV) is included into our calculations for the LO-phonon induced intersubband transfer, as is intrasubband LO-phonon scattering. We account for the electron-electron interaction within the Hartree-Fock mean-field approximation. Moderate to low carrier densities are essential to keep electron-electron Coulomb scattering small and to allow significant buildup of $\pm$ intersubband polarization [8]. Our calculations which include electron-electron scattering show that this is indeed a valid approximation at present carrier densities and for times up to a few picoseconds [9].

For our numerical analysis we use a coherent cw mw field of an intensity of $\approx$1 MW cm^{-2}. The peak of the 200 fs Gaussian pump pulse arrives at $t = 0$ fs and excites electrons from the lowest subband into the subband doublet. This arrival marks the onset of net phonon emission and we measure the phase of the mw field relative to $t = 0$ fs.

Figure 2 shows the population of the lowest subband as a function of time. The solid curve shows the situation without mw field. After the initial excitation by the pump pulse, electrons return to the lowest subband by LO phonon emission with a time constant of about 470 fs. A slight modulation for times $>$200 fs with the tunneling period between the two wells is indeed due to the tunneling of electrons. Transfer into the lowest subband is more likely when an excited electron is in the wide well allowing a direct transition than when it is in the narrow well.

When a coherent mw field couples the subband doublet the rate of transfer depends on its phase. Transfer into the lowest subband, i.e., LO phonon emission, can be either accelerated or slowed down when compared to the mw-free case. The control seen in Fig. 2 is entirely due to adjusting the phase of the mw field. In fact, there are two effects. One is a modulation of absorption of pump pulse photons by the phase of the mw field. This effect has been predicted earlier for excitations across the main band gap in heterostructures [10]. Second, the transfer rate (slope in Fig. 2) depends on the phase of mw field.

One may speculate that the latter effect may be solely a consequence of the first effect which leads to a variation in the number of excited carriers. However, this is not the case due to the moderate carrier densities involved in the transitions. We can demonstrate this clearly by a model calculation in which the mw field is turned on with a time delay of 200 fs relative to the pump pulse. During the excitation of electrons into the upper subbands, there is practically no mw field present to set up $\pm$ intersubband polarization. The results are shown in Fig. 3. Note that, as expected, the initial dynamics is almost identical for all mw phases. However, as soon as the mw field builds up $\pm$ intersubband polarization, the phonon emission rate clearly displays a significant phase dependence.

While Figs. 2 and 3 were obtained by using Markovian phonon emission terms and treating the e-e Coulomb interaction with the Hartree-Fock mean-field approximation, we also performed calculations which included e-e Coulomb scattering in this multisubband system. These calculations are significantly more time-consuming. However, they have confirmed that our simpler approach is well justified and does not invalidate our conclusions. As expected from previous calculations and comparison with experiment [11], electron-electron scattering cannot destroy coherence (here the $\pm$ intersubband polarizations) on a time scale below 1 ps for present carrier densities.

Figure 4 shows the effective carrier relaxation rate as a function of mw phase ϕ_{mu}. This parameter was obtained by fitting an exponential to the decay of carrier densities shown in Fig. 3. Substantial variation of the phonon emission rate between about 400 fs and 1 ps is obtained. The period of phase dependence is π, as compared to 2π in conventional coherent control processes where the phase of the light field responsible for single-photon absorption is varied [2]. This suggests a simple interpretation of this effect in terms of a quantum interference process between single-phonon emission and a single-phonon emission process in conjunction with a mw photon emission and absorption process; see Fig. 1. The two-photon process carries a $2\phi_{\mathrm{mw}}$ dependence into the transition rate.

Note that a recent experiment which reports coherent control of phonon emission in *bulk* GaAs operates in a much shorter time regime, using 15 fs optical pulse to influence the phonon emission dynamics [12]. These pulses are not only significantly shorter than the average LO phonon emission time but also shorter than the corresponding period of lattice vibration which, for bulk GaAs LO phonons, is about 100 fs. Hence, a Markovian picture using energy-conserving delta functions for phonon emission is *not* valid in their scheme, which has a charge transport analog in the intracollisional field effect envisioned to occur in submicron electronic devices [13]. In the current study we have considered a scheme that operates on a time scale which is about an order of magnitude longer. The mw period and the Rabi period of $f_{+-}(\mathbf{k}, t)$ are about 400 fs, the pump pulse duration is 200 fs, and the phonon intersubband emission rate is about 500 fs. The period of lattice vibration of the relevant confined mode in our structure is about 100 fs. As the time scale for the "buildup" of the energy-conserving delta function is determined by the inverse phonon frequency, the use of a Markovian version of electron-phonon scattering terms, Eq. (2), is well justified. Note also that the maximum Stark shift induced by the mw field is less than 1 meV and is thus negligible when compared to intersubband splittings and the LO phonon energy.

In summary, we have shown that quantum interference processes can be used to coherently control effective phonon emission rates in semiconductors. In our present scheme using an asymmetric double quantum well considering of a deep and a shallow GaAs/AlGaAs well, this is accomplished by the phase of a coherent mw field which resonantly couples a pair of electron subbands from which the transition into the ground subband occurs. The phase of the mw field controls the intersubband polarization between the upper two subbands which, in turn,

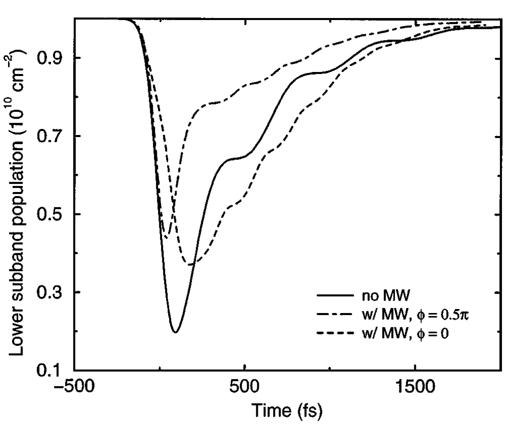

FIG. 2. Lower subband carrier population as a function of time in the presence of a cw mw field.

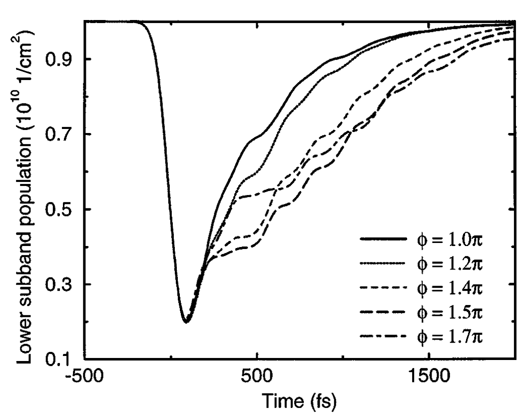

FIG. 3. Lower subband carrier population as a function of time when the cw mw field is turned on with a time delay of 200 fs relative to the pump pulse.

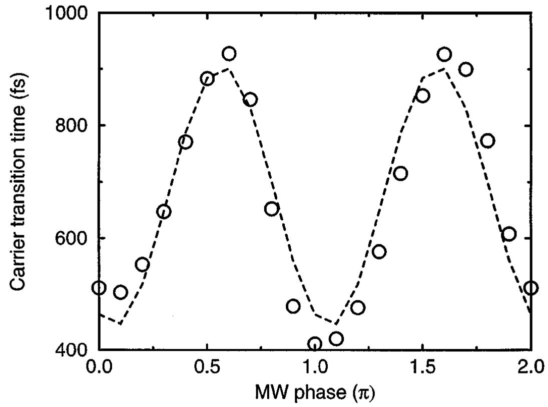

FIG. 4. Phonon emission rate as a function of mw phase. The mw field is turned on with a time delay of 200 fs relative to the pump pulse to ensure equal initial carrier distributions.

influences LO-phonon-induced transitions between the subband doublet and the ground state subband. Our calculations predict that phonon emission rates associated with this intersubband transition can be varied by as much as a factor of 2, i.e., between about 400 fs and 1 ps in the present structure. This significant effect should clearly be observable once coherent mw fields of sufficient intensity become available. The first promising results in building such light sources have been published recently [14].

We thank the U.S. Army Research Office for financial support of this work.

*Present address: Department of Physics, University of Maryland, College Park, MD 20742-4111.

[1] See, for example, K. Seeger, *Semiconductor Physics* (Springer, Berlin, 1973).
[2] See, for example, R. J. Gordon, Annu. Rev. Phys. Chem. **48**, 595 (1997).
[3] See, for example, J. J. Sakurai, *Modern Quantum Mechanics* (Addison-Wesley, Redwood City, 1985), p. 332.
[4] A. P. Heberle, J. J. Baumberg, and K. Köhler, Phys. Rev. Lett. **75**, 2598 (1995).
[5] W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1990).
[6] X. Hu and W. Pötz, in *Coherent Control in Atoms, Molecules, and Semiconductors*, edited by W. Pötz and W. A. Schroeder (Kluwer, Dordrecht, 1999).
[7] B. K. Ridley, Phys. Rev. B **39**, 5282 (1989); Rep. Prog. Phys. **54**, 169 (1991).
[8] 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).
[9] X. Hu and W. Pötz, in Proceedings of the International Workshop on Computational Electronics, Osaka, 1998 (unpublished).
[10] W. Pötz, Phys. Rev. Lett. **79**, 3262 (1997); X. Hu and W. Pötz, Phys. Status Solidi (b) **204**, 350 (1997).
[11] W. Pötz, Phys. Rev. B **54**, 5647 (1996).
[12] M. U. Wehner, M. H. Ulm, D. S. Chemla, and M. Wegener, Phys. Rev. Lett. **80**, 1992 (1998).
[13] J. R. Barker and D. K. Ferry, Phys. Rev. Lett. **42**, 1779 (1979).
[14] R. A. Kaindl, D. C. Smith, M. Joschko, M. P. Hasselbeck, M. Woerner, and T. Elsaesser, Opt. Lett. **23**, 861 (1998).