

## Coherent control of intersubband optical bistability in quantum wells

H. O. Wijewardane and C. A. Ullrich<sup>a)</sup>

Department of Physics, University of Missouri-Rolla, Rolla, Missouri 65409

(Received 29 December 2003; accepted 24 March 2004; published online 3 May 2004)

We present a study of the nonlinear intersubband (ISB) response of conduction electrons in a GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As quantum well to strong terahertz (THz) radiation, using a density-matrix approach combined with time-dependent density-functional theory. We demonstrate coherent control of ISB optical bistability, using THz control pulses to induce picosecond switching between the bistable states. The switching speed is determined by the ISB relaxation and decoherence times,  $T_1$  and  $T_2$ . © 2004 American Institute of Physics. [DOI: 10.1063/1.1751611]

Intersubband (ISB) transitions in semiconductor quantum wells take place on a meV energy scale and are therefore attractive for terahertz (THz) device applications.<sup>1</sup> Many ISB effects of practical interest occur in the nonlinear regime, such as second- and third-harmonic generation,<sup>2</sup> intensity-dependent saturation of photo-absorption,<sup>3,4</sup> directional control over photocurrents,<sup>5</sup> generation of ultrashort THz pulses,<sup>6</sup> plasma instability,<sup>7</sup> or optical bistability.<sup>8,9</sup> Inspired by the photoabsorption experiments by Craig *et al.*,<sup>4</sup> this letter presents a theoretical study of the optical bistability region in a strongly driven, modulation  $n$ -doped GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As quantum well. We demonstrate that ISB bistability can be manipulated on a picosecond time scale by short THz control pulses. This opens up opportunities for experimental study of optical bistability, which in the long run may lead to THz applications such as high-speed all-optical modulators and switches.

Most previous theoretical studies of nonlinear ISB dynamics were based on density-matrix approaches, in Hartree approximation<sup>10–12</sup> or using exchange-only semiconductor Bloch equations.<sup>13–15</sup> These studies showed that the collective ISB electron dynamics is strongly influenced by depolarization and exchange-correlation (xc) many-body effects.<sup>15</sup> We will account for these effects using time-dependent density-functional theory, which has the advantage of formal and computational simplicity.

We describe the conduction subbands in effective-mass approximation, where  $m^* = 0.067m$  and  $e^* = e/\sqrt{\epsilon}$ ,  $\epsilon = 13$ , are the effective mass and charge for GaAs. Initially, the conduction electrons are in the ground state, characterized by single-particle states of the form  $\Psi_{j\mathbf{q}_\parallel}^0(\mathbf{r}) = A^{-1/2} e^{i\mathbf{q}_\parallel \mathbf{r}_\parallel} \psi_j^0(z)$ , with  $\mathbf{r}_\parallel$  and  $\mathbf{q}_\parallel$  in the  $x$ - $y$  plane. The envelope function for the  $j$ th subband  $\psi_j^0(z)$  follows self-consistently from a one-dimensional Kohn–Sham (KS) equation,<sup>16</sup> with the ground-state density

$$n(z) = 2 \sum_{j, \mathbf{q}_\parallel} |\psi_j^0(z)|^2 \theta(\epsilon_F - E_{j\mathbf{q}_\parallel}). \quad (1)$$

Here,  $E_{j\mathbf{q}_\parallel} = \epsilon_j + \hbar^2 q_\parallel^2 / 2m^*$ , and  $\epsilon_j$  and  $\epsilon_F$  are the subband

and Fermi energy levels. We choose the electronic sheet density  $N_s$  such that only the lowest subband is occupied, in which case  $\epsilon_F = \pi \hbar^2 N_s / m^* + \epsilon_1$ .

Under the influence of THz driving fields, linearly polarized along  $z$ , the time-dependent states have the form  $\Psi_{j\mathbf{q}_\parallel}(\mathbf{r}, t) = A^{-1/2} e^{i\mathbf{q}_\parallel \mathbf{r}_\parallel} \psi_j(z, t)$ , with initial condition  $\Psi_{j\mathbf{q}_\parallel}(\mathbf{r}, t_0) = \Psi_{j\mathbf{q}_\parallel}^0(\mathbf{r})$ . In the absence of disorder and phonons, the time evolution of the envelope functions follows from a time-dependent KS equation

$$i\hbar \frac{\partial}{\partial t} \psi_j(z, t) = h(t) \psi_j(z, t), \quad (2)$$

with

$$h(t) = -\frac{\hbar^2}{2m^*} \frac{\partial^2}{\partial z^2} + v_{\text{dr}}(z, t) + v_{\text{conf}}(z) + v_{\text{H}}(z, t) + v_{\text{xc}}(z, t). \quad (3)$$

Here,  $v_{\text{dr}}(z, t) = e\mathcal{E}z f(t) \sin(\omega t)$  describes the driving field, with electric field amplitude  $\mathcal{E}$ , frequency  $\omega$ , and envelope  $f(t)$ .  $v_{\text{conf}}(z)$  is the confining square-well potential, the Hartree potential follows from  $d^2 v_{\text{H}}(z, t) / dz^2 = -4\pi e^* n(z, t)$ , and we use the time-dependent local-density approximation for the xc potential:  $v_{\text{xc}}(z, t) = [de_{\text{xc}}(\tilde{n})/d\tilde{n}]_{\tilde{n}=n(z, t)}$  ( $e_{\text{xc}}$  is the homogeneous electron gas xc energy density). The time-dependent density  $n(z, t)$  follows by substituting  $\psi_j(z, t)$  in Eq. (1).

To account for disorder or phonon scattering, Eq. (2) is replaced by a density-matrix approach. We expand the first conduction subband as  $\psi_1(z, t) = \sum_{k=1}^{N_b} c_k(t) \psi_k^0(z)$ . The associated  $N_b \times N_b$  density matrix  $\rho$  has elements  $\rho_{kl}(t) = c_k^*(t) c_l(t)$  and initial condition  $\rho_{kl}(t_0) = \delta_{kl} \delta_{1k}$ . The time evolution of  $\rho$  follows from

$$i\hbar \frac{\partial \rho(t)}{\partial t} = [h(t), \rho(t)] - R, \quad (4)$$

with the relaxation matrix  $R_{kl} = \hbar[\rho_{kl}(t) - \rho_{kl}(t_0)]/T_{kl}$ . For simplicity,  $T_{kl} = T_1 \delta_{kl} + T_2(1 - \delta_{kl})$ , where  $T_1$  and  $T_2$  are phenomenological relaxation and decoherence times.

We consider a 40 nm GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As square quantum well with  $N_s = 6.4 \times 10^{10} \text{ cm}^{-2}$ , following Craig *et al.*<sup>4</sup> The lowest subband spacing is  $\epsilon_2 - \epsilon_1 = 8.72 \text{ meV}$ , and the ISB plasmon frequency is found from linear-response theory<sup>16</sup> as  $\hbar \omega_{\text{ISB}} = 9.91 \text{ meV}$ . The system has nine bound levels, and

<sup>a)</sup>Electronic mail: ullrichc@umr.edu

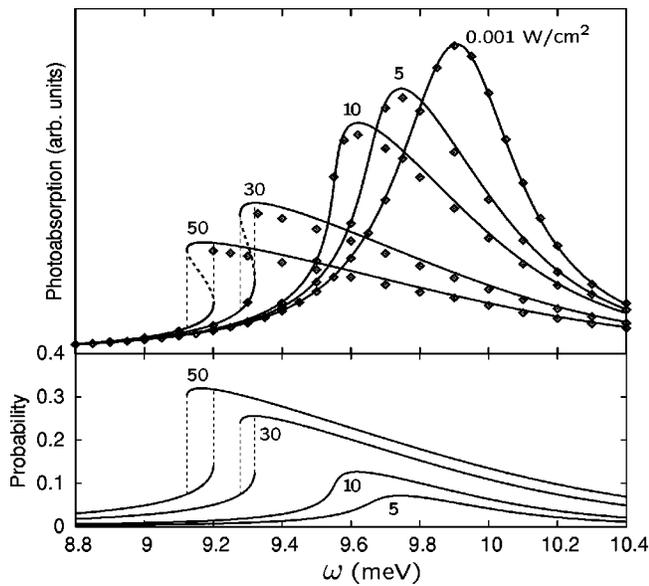


FIG. 1. ISB photoabsorption  $\sigma(\omega)$  and occupation probability  $p_2$  of the second subband for a 40 nm GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As quantum well with electron density  $6.4 \times 10^{10} \text{ cm}^{-2}$ , driven by THz fields with intensities as indicated. Lines: two-level rotating-wave approximation. Points: six-level density-matrix calculation.

we take the lowest six to construct the density matrix ( $N_b = 6$ ). We use the ISB scattering times  $T_1 = 40 \text{ ps}$  and  $T_2 = 3.1 \text{ ps}$ , consistent with recent measurements of  $T_1$  and  $T_2$  for similar systems.<sup>17,18</sup>

To describe ISB photoabsorption, we propagate Eq. (4) in the presence of THz driving fields, switched on at  $t_0$  over a five-cycle linear ramp and then kept at constant intensity for several hundred picoseconds (ps). From the dipole moment  $d(t) = N_s \sum_{kl}^{N_b} \rho_{kl}(t) \int dz \psi_k^0(z) z \psi_l^0(z)$  we obtain the photoabsorption cross section (the dissipated power)  $\sigma(\omega) \sim \omega \int_{t_0}^{t_0+T} \cos(\omega t') d(t') dt'$ , where  $T$  is one cycle of the driving field  $v_{dr}$ . Following the switching on of the THz field,  $\sigma(\omega)$  fluctuates considerably from one cycle to the next, but eventually approaches a stable value as the transients settle down.

Figure 1 shows  $\sigma(\omega)$  calculated with our six-level density-matrix formalism, in close agreement with the two-level rotating-wave approximation<sup>10</sup> including xc. For low intensities,  $\sigma(\omega)$  is a Lorentzian with maximum at  $\omega_{ISB}$  and width  $2\hbar/T_2$ . Approaching the saturation intensity  $I_0 = c \epsilon^{1/2} \hbar^2 / 8\pi T_1 T_2 e^2 |\langle \psi_2^0 | z | \psi_1^0 \rangle|^2$ , the photoabsorption peak shifts to lower energies and changes shape (here,  $I_0 = 26.8 \text{ W/cm}^2$ ). The physical reason for this effect is that the depolarization shift  $\Delta = \hbar \omega_{ISB} - (\epsilon_2 - \epsilon_1)$  is proportional to the population difference  $p_1 - p_2$ . Initially,  $p_1 = 1$  and  $p_2 = 0$ . Strong driving fields lead to a decrease of  $p_1 - p_2$  because of transitions into the second and, eventually, higher levels, see the lower panel of Fig. 1. Since this population transfer is most efficient around the ISB resonance, the peak of  $\sigma(\omega)$  shifts more than the tails, leading to an asymmetric line shape.

For intensities over  $16 \text{ W/cm}^2$  we discover regions of optical bistability in  $\sigma(\omega)$ : the system responds in two different ways to one and the same driving field (the middle branch is unstable). The two states are characterized by density oscillations with different amplitudes and phases (upper

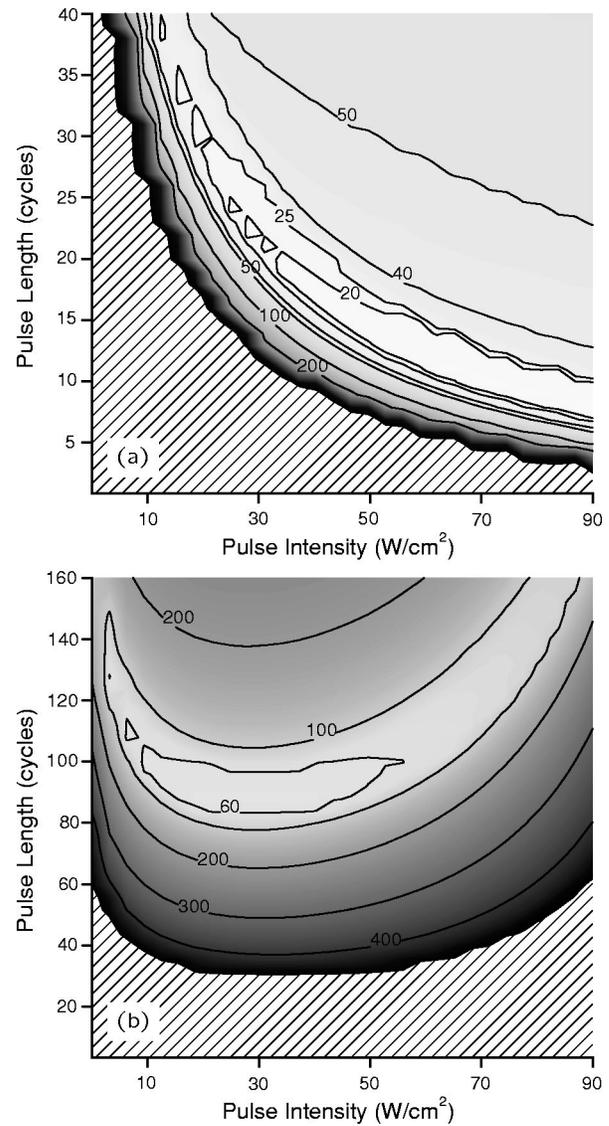


FIG. 2. Intensity and length of THz control pulses for switching between the lower and upper ISB bistable states at  $I = 30 \text{ W/cm}^2$  and  $\omega = 9.3 \text{ meV}$  driving field (0.445 ps per cycle). Contour labels indicate the switching times in ps. No switching occurs in the hatched regions. (a) Up-switch, (b) down-switch (control pulses are phase shifted by  $\pi$ ).

branch: close to  $\pi/2$ ; lower branch: around  $\pi/4$ ), and different level populations.

Whether the system will be in the upper or lower bistable state depends on its history. A hysteresis-like behavior is observed at fixed intensity and under adiabatically slow frequency changes of the driving field:<sup>12</sup> Entering the bistability region from the low-frequency side, the system continues on the *lower* branch and then jumps up at the end of the bistability region. Entering from the other side, the system follows the *upper* branch. The required continuous, adiabatic frequency tuning of THz driving fields is difficult to realize in experiment, and of little practical use in exploring ISB optical bistability for potential applications. Instead, it would be desirable to switch rapidly between the two bistable states.

In the following, we demonstrate coherent control of ISB optical bistability by short THz control pulses. This method is both rapid and robust, and lends itself for experimental implementation. Figure 1 suggests that a switch from

the lower to the upper bistable state requires a transfer of population into the second level. The necessary energy can be rapidly injected into the system by a short THz pulse. On the other hand, we expect switching down from the upper to the lower state to be inherently slower, since upper-level population needs time to relax.

We consider a driving field with  $I=30 \text{ W/cm}^2$  and  $\hbar\omega = 9.3 \text{ meV}$  (0.445 ps per cycle) in the middle of the bistable region, such that the system is in the lower bistable state. At  $t_1$ , we apply a short pulse with the same frequency and *in phase* with the driving field, and with a trapezoidal pulse envelope: linearly turned on and off over five cycles, constant in between over  $N_c$  cycles (the precise pulse shape is not essential). We calculate  $d(t)$  and  $\sigma(\omega)$  as earlier, using our density-matrix formalism. After transients and other disturbances induced by the pulse have subsided, the system either slowly returns to the lower state, or converges towards the upper bistable state, in which case we define a “switching time” as that time after  $t_1$  when  $\sigma(\omega)$  is converged to within 5% of its final value. To study the reverse process, we prepare the system in the upper bistable state and then apply similar control pulses, *phase shifted by  $\pi$*  with respect to the driving field. After the transients have settled, the system either remains in the upper state, or goes down to the lower state.

Control pulses of various intensity and length  $N_c$  were tested to determine the conditions for successful up- or down-switching, and to find those pulses that induce the fastest switching. The results are summarized in Fig. 2. The hatched areas indicate the unsuccessful pulses: in the case of up-switching, they do not have sufficient energy to promote enough electrons to the upper level, and in the case of down-switching, they are too short to allow enough electrons to relax down.

The limiting curve in Fig. 2(a) separating the successful from the unsuccessful pulses corresponds to pulses of energy  $1.87 \times 10^{-10} \text{ J/cm}^2$ . The shortest switching times of 15–20 ps are achieved for somewhat higher pulse energy, around  $3.3 \times 10^{-10} \text{ J/cm}^2$ . No systematic effort was made to further optimize the control pulses. A physical limit for switching speed is set by the decoherence time  $T_2$  (here 3.1 ps), which governs the decay of transients.

Down-switch from the upper to the lower ISB bistable state requires longer control pulses, at least 30 cycles. The associated switching times are limited by the ISB relaxation time  $T_1$  (here 40 ps). We find that the shortest down-switching times of 50–60 ps are achieved by control pulses with intensity not far from the driving field,  $30 \text{ W/cm}^2$ . Due to the phase difference  $\pi$ , these control pulses effectively

suppress the driving field, giving electrons time to relax from the upper level.

In summary, we have simulated coherent control of ISB optical bistability in quantum wells, using THz control pulses to switch rapidly between the bistable states. Switching times are in principle limited only by the intrinsic relaxation and decoherence times. Interestingly, a shorter relaxation time  $T_1$  (i.e., more scattering) implies faster switching. More realistic simulations would replace  $T_1$  and  $T_2$  with microscopic, intensity-dependent scattering theories, including finite temperatures and a detailed modeling of the absorption profile and waveguide geometry of the device. In the search for THz devices and applications, nonlinear ISB effects such as bistability merit further exploration.

The authors acknowledge support from the donors of the Petroleum Research Fund, administered by the ACS, and from the University of Missouri Research Board.

- <sup>1</sup> *Intersubband Transitions in Quantum Wells I*, edited by H. C. Liu and F. Capasso, Semiconductors and Semimetals Vol. 62 (Academic, San Diego, 2000).
- <sup>2</sup> J. N. Heyman, K. Craig, B. Galdrikian, M. S. Sherwin, K. Campman, P. F. Hopkins, S. Fafard, and A. C. Gossard, Phys. Rev. Lett. **72**, 2183 (1994).
- <sup>3</sup> F. H. Julien, J.-M. Lourioz, N. Herschkorn, D. Delacourt, J. P. Pocholle, M. Papuchon, R. Planel, and G. Le Roux, Appl. Phys. Lett. **53**, 116 (1988).
- <sup>4</sup> K. Craig, B. Galdrikian, J. N. Heyman, A. G. Markelz, J. B. Williams, M. S. Sherwin, K. Campman, P. F. Hopkins, and A. C. Gossard, Phys. Rev. Lett. **76**, 2382 (1996).
- <sup>5</sup> E. Dupont, P. B. Corkum, H. C. Liu, M. Buchanan, and Z. R. Wasilewski, Phys. Rev. Lett. **74**, 3596 (1995).
- <sup>6</sup> J. N. Heyman, R. Kersting, and K. Unterrainer, Appl. Phys. Lett. **72**, 644 (1998).
- <sup>7</sup> P. Bakshi, K. Kempa, A. Scorupsky, C. G. Du, G. Feng, R. Zobl, G. Strasser, C. Rauch, C. Pacher, K. Unterrainer, and E. Gornik, Appl. Phys. Lett. **75**, 1685 (1999).
- <sup>8</sup> M. Seto and M. Helm, Appl. Phys. Lett. **60**, 859 (1992).
- <sup>9</sup> M. I. Stockman, L. N. Pandey, L. S. Muratov, and T. F. George, Phys. Rev. B **48**, 10966 (1993).
- <sup>10</sup> M. Załuzny, Phys. Rev. B **47**, 3995 (1993).
- <sup>11</sup> B. Galdrikian and B. Birnir, Phys. Rev. Lett. **76**, 3308 (1996).
- <sup>12</sup> A. A. Batista, P. I. Tamborenea, B. Birnir, M. S. Sherwin, and D. S. Citrin, Phys. Rev. B **66**, 195325 (2002).
- <sup>13</sup> D. E. Nikonov, A. Imamoglu, L. V. Butov, and H. Schmidt, Phys. Rev. Lett. **79**, 4633 (1997).
- <sup>14</sup> A. Olaya-Castro, M. Korkusinski, P. Hawrylak, and M. Yu. Ivanov, Phys. Rev. B **68**, 155305 (2003).
- <sup>15</sup> J. Li and C. Z. Ning, Phys. Rev. Lett. **91**, 097401 (2003).
- <sup>16</sup> C. A. Ullrich and G. Vignale, Phys. Rev. B **58**, 15756 (1998); Phys. Rev. Lett. **87**, 037402 (2001).
- <sup>17</sup> J. N. Heyman, K. Unterrainer, K. Craig, B. Galdrikian, M. S. Sherwin, K. Campman, P. F. Hopkins, and A. C. Gossard, Phys. Rev. Lett. **74**, 2682 (1995).
- <sup>18</sup> J. B. Williams, M. S. Sherwin, K. D. Maranowski, and A. C. Gossard, Phys. Rev. Lett. **87**, 037401 (2001).