Rabi Flopping in a Two-Level System with a Time-Dependent Energy Renormalization: Intersubband Transitions in Quantum Wells

Adriano A. Batista* and D. S. Citrin

School of Electrical and Computer Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332-0250, USA

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We obtain pulse-driven Rabi oscillations guided by a generalization of the rotating-wave approximation to include, in the optical-Bloch equations, two-level systems with a time-varying transition energy. We achieve this by using chirped pulses with the central frequency given by the time-varying transition energy. Using this approach, we predict Rabi oscillations in intersubband transitions in a two-subband n-type modulation-doped quantum well by taking into account the time-dependent intersubband energy-gap renormalization due to depolarization-shift effects. We obtain Rabi oscillations for $j\pi$ ($j = 0, 1, 2, \ldots$) pulses in the presence of dephasing.

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It is well known that electromagnetic pulses with a slowly varying amplitude at resonance with a two-level quantum mechanical system give rise to coherent oscillations in the populations of the two levels known as Rabi oscillations [1]. When the pulse area $\mathcal{A}(z) = \mu \int_{-\infty}^{\infty} F_0(z, t)\, dt$, with $F_0(z, t)$ the slowly varying envelope of the pulse, is an even multiple $j = 2m$ of π, the pulse propagates without decay in a single-resonance two-level atomic medium. On the other hand, for $j = 2m + 1$, $j\pi$ pulses cause population inversion and are rapidly attenuated. This occurs only if the pulse duration is shorter than the homogeneous depopulation and decoherence times T_1 and T_2, respectively, and also any dephasing time due to inhomogeneous broadening; detuning has to be negligible as well on this time scale. These results were obtained by McCall and Hahn [2,3]. The Rabi-flopping concept is the framework on which virtually all subsequent theory and experiment on coherent optical pulses interacting with two-level atoms [4,5], one of the most fundamental problems of quantum optics, are based. It has found large experimental success and broad technological applications also. Furthermore, Rabi oscillations are a prototypical example of a nonperturbatively driven transition in a two-level system, and thus serve as a model for transitions beyond Fermi's golden rule. As such, Rabi oscillations involve dynamics on a time scale shorter than the dephasing time associated with coupling to a bath, which in solid-state systems is far from typical. The ability to produce Rabi oscillations in a given system, moreover, indicates whether that system is a candidate for more elaborate coherent manipulation, such as in coherent control experiments or for quantum computing.

In this Letter, we aim to extend the validity of these results to n-type modulation-doped semiconductor quantum wells (QWs). We apply our theory here to intersubband transitions in a two-subband symmetric QW with only homogeneous broadening and energy-gap renormalization due to depolarization shift. With a slight modification, this approach can also be applied to interband transitions. In the following, we show that rather substantial Rabi oscillations can be achieved in intersubband transitions in QWs, but only if the time-dependent renormalization of the subband splittings is taken into account.

Semiconductors provide only an approximate realization of a two-level system. Even in the absence of carrier-carrier interactions, a two-band semiconductor presents a set of two-level systems with natural frequencies corresponding to the energies of allowed vertical transitions. The introduction of carrier-carrier and carrier-phonon interactions, together with the accompanying optical nonlinearities, produce even further departures from the two-level system. Nevertheless, phenomena closely allied with two-level Rabi flopping have been observed. It has already been shown experimentally in semiconductors that Rabi oscillations can occur [6,7]. More recently in heterostructures, π pulses have been used to perform a single-electron coherent turnstile in quantum dots under a dc bias [8]. Theoretical studies of Rabi oscillations for interband transitions have also already been made [9–12]. Although they took into account the effect of many-body interactions in the semiconductor-Bloch equations by setting up the pulse frequency with detuning with the unrenormalized energy gap, this frequency was fixed in time and the pulse areas that achieved the best results were not multiples of π. The general plan was to choose simple pulse shapes and vary the area to achieve the deepest Rabi flopping subject to this constraint. Suppose the energy gap $\omega(t) = E_1(t) - E_0(t)$ in fact varies with time. How can one find an effective π pulse for this system? We show that one can use a pulse that is suitably chirped (time-dependent carrier frequency) to track self-consistently the renormalized subband splitting in time induced by the pulse itself. We begin quite generally with a quantum mechanical two-level system whose levels depend upon time. We then justify this ansatz with detailed numerical calculations for the modulation-doped n-type QW.

In GaAs/AlGaAs QWs, the conduction subbands are quite parabolic with little mass dispersion; therefore the dependence on k of the transition energy and the momentum matrix elements are safely neglected. This is an aid to the treatment, since models based on the optical-Bloch equation will, to a large extent, be applicable to the intersubband transitions. From the two-level subspace of solutions of the Schrödinger equation, we find the amplitudes a and b for the lower and upper levels, respectively, obey $i\dot{a}(t) = \mu F(t) b$ and $i\dot{b}(t) = \omega(t) b + \mu F(t) a$, where $\omega(t)$ is the time-dependent energy difference of the levels, μ is the dipole moment, and $F[t) = (F_0 e^{-i\int_0^t \omega(s)\,ds} + F_0^* e^{i\int_0^t \omega(s)\,ds}]$ is the incident electric field ($\hbar = 1$). If we make the transformation $b(t) = \tilde{b}(t) e^{-i\int_0^t \omega(s)\,ds}$, we obtain $i\dot{a}(t) = \mu F(t)\tilde{b} e^{-i\int_1^t \omega(s)\,ds}$ and $i\dot{\tilde{b}}(t) = \mu F(t) a e^{i\int_0^t \omega(s)\,ds}$. We now suppose that the electric-field amplitude depends on time, i.e., $F_0 = F_0(t)$ is the slowly varying pulse envelope. The rotating-wave approximation (RWA), in the frame whose rotation is given by $\exp[-i\int_0^t \omega(s)\,ds]$, is applied to the foregoing and results in $i\dot{a}(t) = F_0^*(t)\mu\tilde{b}$ and $i\dot{\tilde{b}}(t) = F_0(t)\mu a$. One notes that these are the same equations resulting in Rabi flops when a two-level system with a constant energy gap is driven by a slowly varying pulse envelope at resonance. Namely, if the $j\pi$ pulse for a time-independent level splitting is $F(t) = F_0(t)\exp(-i\omega t) +$ c.c., then for $\omega(t)$ the corresponding effective $j\pi$ pulse is $F(t) = F_0(t)\exp[-i\int_0^t ds\,\omega(s)] +$ c.c..

For the sequel, it will prove useful to restate the former in the language of the density matrix. Begin with the free-carrier density-matrix equations $\dot{\Delta} = 4\mu\,\mathrm{Im}\,\sigma F(t)$ and $\dot{\sigma} = i\omega(t)\sigma - i\mu\Delta F(t)$, where Δ is the population difference between upper and lower levels and σ is the off-diagonal element of the density matrix in the basis of the two levels. The driving field is chosen to be $F(t) = F_0(t) e^{-i\int_0^t \omega(s) ds} +$ c.c. Then, as above, using the transformation $\sigma = \tilde{\sigma} e^{i\int_0^t \omega(s) ds}$ followed by our RWA ansatz, we obtain $\dot{\Delta} = -2i\mu(\tilde{\sigma} - \tilde{\sigma}^*)F_0(t)$ and $\dot{\tilde{\sigma}} = i\mu\Delta F_0(t)$, where without loss of generality we assume the phase dependence of $F_0(t)$ to be constant in time. By integrating the above equations, we obtain $\Delta(t) = \Delta_0\cos[2\mu\int_0^t F_0(s)\,ds]$, where Δ_0 is the equilibrium population difference between the bottom and the upper level. The polarization is given by $P(t) = -N\mu\Delta_0\sin[2\mu\int_0^t F_0(s)\,ds]\sin\int_0^t \omega(s)\,ds$, which is a simple generalization of the polarization from the usual Rabi flops in two-level systems [5]. Once again, this discussion shows that the envelope for a $j\pi$ pulse, with its high frequency varying in time, is the same as in the case where the levels are time independent. This will now be applied to Rabi oscillations in intersubband transitions.

The resonant intersubband THz response of modulation-doped n-type QWs is known to be highly nonlinear [13]. As electrons undergo transitions between the two subbands, the band bending is modified dynamically. Other nonlinearities include time-dependent Pauli blocking and excitation-induced dephasing. The result is that the intersubband plasmon exhibits a significant nonlinear response. Our approach is to apply the time-dependent Hartree approximation to describe the intersubband response to a strong THz field. This approach has been used [14,15] to describe the experimentally observed nonlinearities in QWs [16,17]. The time-dependent Hartree approximation with cw THz driving is valid for wide QWs at moderate to high carrier densities in materials where mass dispersion can be safely neglected [18]. For pulse durations of several picoseconds, the linewidths are narrow enough that one can assume the results are close enough to those of cw THz driving. The cases we concentrate on fall well within this range of validity. As such, exchange and exchange-correlation effects are expected to be small for the purposes of this study. We use the forgoing treatment of a two-level system to obtain effective $j\pi$ THz pulses. Having obtained these pulses, we use them in numerical computations based on the time-dependent Hartree approximation to calculate $\omega(t)$ and thus the THz response of the QW. Although in Ref. [7] the possibility of a time-dependent band gap was discussed in the context of interband Rabi flopping, the excitation levels were always sufficiently low that these effects did not play a major role.

The treatment of the nonlinear THz intersubband response of QWs is discussed in depth elsewhere [19,20]. Here, we quote the essential results. We have shown that the time-dependent Hartree approximation can be conveniently restated in terms of the density matrix [19,20]. The density-matrix equations for intersubband transitions in a symmetric QW are given by

$$\begin{aligned} \dot{\Delta} &= -\gamma_1(\Delta - \Delta_0) + 4\,\mathrm{Im}\,\sigma[\mu F(t) + \alpha\,\mathrm{Re}\,\sigma], \\ \dot{\sigma} &= i\omega_{10}\sigma - \gamma_2\sigma_{10} - i\Delta[\mu F(t) + \alpha\,\mathrm{Re}\,\sigma] \\ &\quad - i\alpha\beta\sigma(\Delta - \Delta_0)/4, \end{aligned} \tag{1}$$

where ω_{10} is the Hartree self-consistent QW intersubband energy gap, $\gamma_1 = 1/T_1$ and $\gamma_2 = 1/T_2$, and α and β are constant coefficients due to Coulomb interactions numerically calculated from the Hartree approximation, which are defined in Ref. [20]. [They are roughly $\propto N_s L_{\mathrm{QW}}$, where L_{QW} is the QW width.] Within the RWA we can recast Eq. (1) in the free-carrier form if we take $\omega(t) = \omega_{10} - \alpha\Delta/2 - \alpha\beta(\Delta - \Delta_0)/4$. With this information we can design pulses on demand. This approach could in principle be applied to more complex systems, such as to intersubband transitions with mass dispersion, exchange interaction, and electron-electron scattering, and to interband transitions; in such cases, we merely utilize the appropriate form of $\omega(t)$. The two-level model should be accurate provided the renormalized energy does not depend too strongly on k (since this dependence generates inhomogeneous broadening).

We now present numerical results of the action of appropriately designed $j\pi$ pulses on n-type modulation-doped QW's. The GaAs/AlGaAs double QW structure used for our calculations is 310 Å wide and 200 meV deep with one barrier of 50 Å in width and 50 meV in height, as shown by the inset in Fig. 1. The QW intersubband energy ω_{10} is about 1.4 THz (5.7 meV). We take $T_1 = 0.7$ ns, $T_2 = 6.6$ ps ($\gamma_2 = 0.1$ meV), and the temperature $T = 4$ K. In all cases, the pulse envelopes $F_0(t)$ are Gaussian. The Eqs. (1) were integrated using the fourth-order Runge-Kutta method with 2048 steps per cycle of drive.

In Fig. 1, we show that the usual π pulses do not work properly in inverting the populations once the time-dependent depolarization-shift effects, expressed via the nonlinear terms of $\omega(t)$, become relevant. Namely, we employ naive π pulses, as shown in Fig. 1(a). Frames (b) and (c) show the inversion for various electron densities N_s neglecting and including dephasing, respectively. At very low doping levels ($N_s = 0$ in the figure, but in practice $\sim 10^9$ cm^{-2}), the π pulse achieves population inversion; however, at moderate densities of $\sim 10^{10}$ cm^{-2}, the degree to which the system has been inverted is strongly degraded. Above $N_s \sim 7 \times 10^{10}$ cm^{-2}, the upper subband becomes populated, and the pulse has little effect on the inversion.

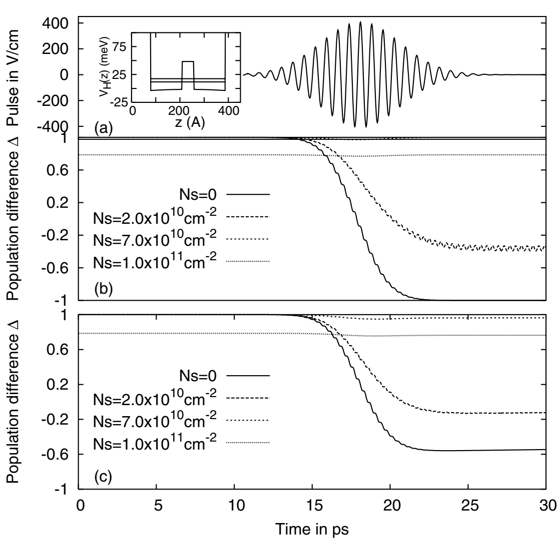

FIG. 1. (a) π pulse with a fixed carrier frequency at resonance with the time-independent subband splitting ω_{10}. The inset shows the QW potential in the conduction band together with the two relevant states. (b) The population dynamics in the absence of dephasing. No population inversion is achieved when depolarization effects are relevant, i.e., above $N_s \sim 10^{10}$ cm^{-2}. (c) Same as in (b), but with a dephasing time of $T_2 = 6.6$ ps.

We now see if including the time-dependent renormalization of the subband splitting within the pulse achieves a higher degree of inversion. In Figs. 2(a) and 2(b), we see that our prescription requires a strongly chirped pulse (the blueshift of the initial frequency is due to depolarization shift, which changes sign when there is inversion; the fast small-amplitude oscillations in $\omega(t)$ indicate that the RWA is not perfect, but as long as these oscillations are relatively small the RWA is a good approximation). The illustrated pulse is an example for $N_s = 7 \times 10^{10}$ cm^{-2}. One can see that our method for generating π pulses works quite effectively in frames (c) and (d) (without and with dephasing) at densities $> 10^{10}$ cm^{-2}, where the unchirped-pulse effectiveness in achieving inversion becomes severely degraded. Note that the pulse duration is slightly longer than T_2 so that dephasing places a limit on the greatest inversion that can be obtained.

The pulses employed above have peak fields of 400 V/cm. Such large-amplitude pulses are difficult to obtain; however, smaller peak fields imply longer pulse duration for a given value of $\mathcal{A}$. Nevertheless, it is of interest to see the degree to which the technique might be

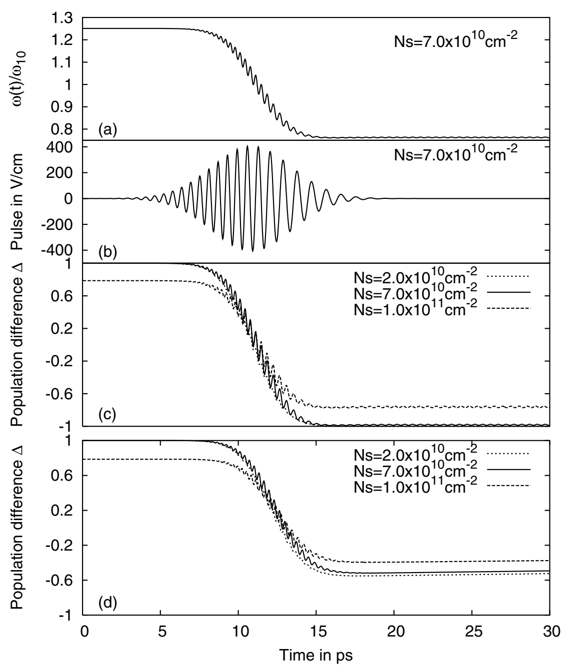

FIG. 2. Frames (a) and (b) show an effective π pulse accounting for the time-dependent renormalized subband splitting. Note the strong down chirp resulting in a decrease in carrier frequency over the pulse duration. This example is for electron density $N_s = 7 \times 10^{10}$ cm^{-2}. The pulse shape is Gaussian and the fast frequency varies according to $\omega(t)$ for the QW as given in the text. (a) The frequency dependence and the (b) electric field of the effective π pulse. Frames (c) and (d) show the inversion in the absence and presence of dephasing (with $T_2 = 6.6$ ps), respectively.

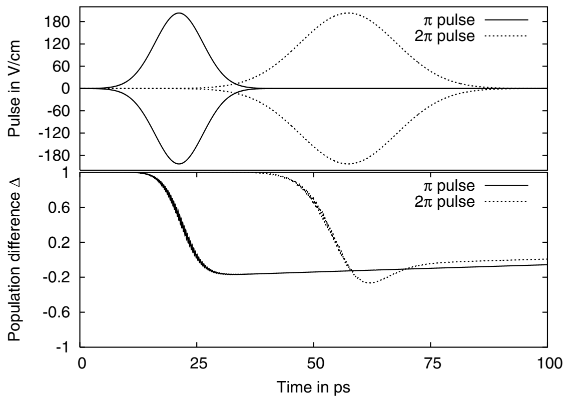

FIG. 3. Upper frame shows effective π and 2π pulse envelopes with peak electric fields of 190 V/cm; the lower frame shows the inversion. In particular, the weak oscillation in the curve in the lower frame for the 2π pulse indicates a partial Rabi flop. Here $N_s = 7.0 \times 10^{10}$ cm^{-2} and $T_2 = 6.6$ ps.

useful for pulses with smaller peak fields. We choose the value of 190 V/cm; chirped pulses with these amplitudes are likely to be obtainable using optical rectification of ultrafast optical pulses in suitably designed nonperiodically poled lithium niobate [21]. In Fig. 3, we consider the action of optimized effective π and 2π pulses with a peak amplitude of 190 V/cm. In this case, the π-pulse duration is $\sim$25 ps $> T_2$. Because of dephasing, the π pulse does not achieve complete inversion, nor does the 2π pulse return the system to the lower level; however, the inversion does show the signature of π- and 2π-pulse action. Clearly, there is a tradeoff between peak field and pulse duration.

In conclusion, we have generalized the concept of Rabi flopping to the case where the level separation varies with time sufficiently slowly that a simple extension of the RWA may be applied. In particular, we consider the case where the level splitting depends self-consistently on the optical pulse. We show how to obtain the self-consistent effective $j\pi$ pulses leading to population oscillations. The method is applied to intersubband Rabi flopping in modulation-doped n-type QWs, where the time-dependent subband-energy renormalizations may be substantial. We find that deep Rabi flopping can be achieved even in the presence of dephasing for moderate doping provided the peak fields are on the order of 200 V/cm. More broadly, the technique may be helpful to generate Rabi oscillations in other systems exhibiting time-dependent level renormalizations, and may find application in coherent control. Very recently, some phase-resolved nonlinear-response experiments detected intersubband Rabi oscillations in QWs in the tens of THz [22]; in principle they should be able to study the effects of many-body interactions in the coherent evolution of polarization when they use wider QWs with doping concentrations similar to ours.

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*Present address: Instituto de Física, Universidade de Brasília, CEP: 70919-970, Brasília-DF, Brazil.

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