Rabi oscillations of 2D electrons under ultrafast intersubband excitation

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We investigate coherent nonlinear dynamics of 2D electrons under ultrafast intersubband excitation by mid-IR pulses. We include the effects of relaxation and dephasing, both homogeneous and inhomogeneous, as well as detuning within a non-Markovian equation to obtain temporal population redistributions. We show how, using a cross-correlation method, the effects of Rabi oscillations may be detected in this system, and briefly discuss other detection methods.

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The coherent dynamics and manipulation of two-level systems under ultrafast excitation dates back over three decades, where the majority of focus has been on atomic ensembles. During recent years, much attention has been given to intersubband effects in low-dimensional heterostructures. In particular, investigations of single quantum dots under ultrafast intersubband excitation, where dephasing times can be hundreds of picoseconds, has seen the experimental observation of Rabi oscillations, providing the basis for possible readout devices for excitonic quantum gates. Previously, interband Rabi oscillations under near-IR excitation had been observed in quantum well (QW) structures, albeit more highly suppressed by shorter dephasing times.

Following these interband investigations we investigate the possibility of Rabi oscillations between 1 and 2 conduction subbands in a QW under ultrafast mid-IR excitation. Advances in mid-IR femtosecond spectroscopic techniques have culminated with recent experimental results suggesting that ‘partial Rabi flops’ may be possible under relatively low pulse energies. Theoretical investigations of intersubband systems are therefore timely.

The occurrence of subband Rabi oscillations under relatively low pump energies in low-doped QWs is made possible due both to the suppression of the LO-phonon emission rate (assuming the interlevel energy is much greater than the optical phonon energy) and also the reduced efficiency of electron-electron scattering at low densities ($\sim 10^9 - 10^{10}$ cm$^{-2}$). The inclusion of the role played by dephasing mechanisms is crucial for any description of coherent intersubband optics. Whilst a complete treatment requires the full rigour of many-body calculations, we show in this paper that treating the different damping terms phenomenologically within a non-Markovian quantum kinetic scheme gives clear indications of the interplay between the various timescales involved, and provides insight into the possibility of coherent manipulation. Experimental observations show that carrier-carrier dephasing rates are effectively independent of carrier density at low concentration ($\sim 10^{10}$ cm$^{-2}$). We therefore restrict our study to the low-density regime where the assumption of constant dephasing is approximately valid.

After introducing the framework of the two subband model, we consider the balance of population redistributions to derive an integro-differential equation governing population dynamics under the influence of an ultrafast pulse. By considering the ideal limit, where dephasing and relaxation are neglected, we show that our model reduces to analytic expressions which essentially recover the area theorem. The effects of the various damping mechanisms are reintroduced in line with recent experimental measurements to investigate the degree of coherent behaviour which can realistically be observed. We extend our calculations to analyse a detection scheme based on cross-correlation absorption under two-pulse excitation, and briefly discuss a scheme based on THz emission from non-symmetric QWs. Such methods have previously been implemented for the near-IR interband pump case. We conclude with a brief discussion on the approximations we have made.

We describe the coherent dynamics of the system using a density matrix approach, evaluating the balance equation for an interlevel redistribution of population under a resonant intersubband pump. We consider the response of the system due to the interaction with an electric field $E_{s} \equiv [E \exp(-i\omega t) + c.c.] \omega t$ perpendicular to the QW plane. The dipole perturbation operator is: $\hat{\delta h}(t) = \langle ieE/\omega \rangle \hat{v}_{\perp} \omega t$, where $\hat{v}_{\perp}$ is the intersubband velocity operator and $\omega t$ is the form-factor of the pulse. We write the high-frequency component of the density matrix, $\hat{\rho}_{s} \exp(-i\omega t) + H.c.$, in the form:

$$\hat{\delta \rho}_{s} \simeq \frac{1}{\hbar} \int_{-\infty}^{0} dt' e^{i\omega t'} \left[ \int dt'' e^{i\omega t''} \frac{\delta h_{t}}{i\hbar} \right] e^{-(i/\hbar)\bar{h}t'}, $$(1)

where $\tau_{2}$, the dephasing time, describes the decay of the induced coherence, and $\bar{h}$ is the period-averaged Hamiltonian of the QW. The density matrix averaged over the period $2\pi/\omega$, $\bar{\rho}_{s}$, obeys the quantum kinetic equation

$$\frac{\partial \bar{\rho}_{s}}{\partial t} + \frac{i}{\hbar} \left[ H_{s}, \bar{\rho}_{s} \right] = \hat{G}_{t} + \hat{I}_{sc},$$

(2)

where $\hat{I}_{sc}$ is the collision integral. The time-dependent
The balance equations have the form:

\[ \dot{G}_t = \frac{1}{h} \int_{-\infty}^{0} dt' e^{i\omega t'/\tau} \left[ \hat{\Theta}_{t+t'} \hat{\rho}_{t+t'} + \text{H.c.} \right] + H.c., \]

where we have introduced the phototransition frequency \( \nu_r = (2eV/E_\hbar)^2 \tau_p \) with \( \tau_p \) the pulse duration. Factors which suppress the coherent response are described by the parameters \((\tau_{1,2})^{-1}\) and \(\gamma/h\), as well as detuning of the pulse from resonance \(\Delta \omega\).

Before we resort to numerical solution of Eq. (7), we consider analytic solutions in the limiting case \(\tau_{1,2} \to \infty\), \(\gamma \to 0\), and \(\Delta \omega \to 0\), (i.e., \(\Phi_t \to 1\)). Eq. (7) can then be transformed into the second order differential equation

\[ \frac{d^2 \Delta n_t}{dt^2} - \frac{1}{w_t} \frac{d\nu_r}{dt} \frac{d\Delta n_t}{dt} + \frac{\nu_r}{\tau_p} \frac{w_t^2}{w_t^0} \Delta n_t = 0, \]

where \(w_t^0 = \sqrt{\nu_r \tau_p} \int_{-\infty}^{t/\tau_p} dwz_w \), which we illustrate in Fig. 1. As expected, population redistributions are solely determined by the area of the incident pulse: \(A_p = \sqrt{\nu_r \tau_p} \int_{-\infty}^{t/\tau_p} dwz_w \). In Fig. 1a we plot the solution as a function of time for both secant and Gaussian pulses each with duration \(\tau_p\) (FWHM). As illustrated in panels (i) and (ii), which correspond to secant pulses with \(A_p = \pi \) and \(2.5\pi\) respectively, population can be distributed among the two levels according to the area theorem. For fixed values of \(\nu_r\) and \(\tau_p\), the area under the Gaussian pulse is slightly less than that under the secant pulse. The final redistribution of population, shown in Fig. 1b as a function of the secant pulse area, \(\sqrt{\nu_r \tau_p}/\alpha_s\), where \(\alpha_s = 2 \text{sech}^{-1} [0.5] \), confirms this point.

The condition for population inversion, \(A_p = \pi\) can be obtained by a secant pulse, with duration \(\tau_p = 100\) fs, having a free space intensity per unit area of 5.22 MW/cm\(^2\). In order to achieve maximum coupling of the transverse field to the intersubband dipole, we consider a 45° prism integrated onto the sample. With this geometry, approximately 33% of the incident power can be coupled to the transition. We assume parameters for a typical GaAs QW, of width 85 Å, corresponding to an interlevel energy of \(\hbar \omega \approx 100\) meV. Both sets of parameters are typical of recent experiments.

To study the effects of the various damping processes, we solve Eq. (7) numerically using a form of Picard iteration. We take as example a secant 1.6\(\pi\)-pulse, and consider the effects of the parameters, \(\tau_1, \tau_2, \Delta \omega^{-1}\), and \(\sqrt{\nu_r}/\gamma\), setting each in turn to \(\tau_p\) with the other three set to the limiting values as outlined above. The results are shown in Fig. 2. LO-phonon emission \((\tau_1)\) causes slow relaxation of the population back to the ground state after the pulse has passed. In contrast, the effect of a large detuning as well as broadening, both inhomogeneous and homogeneous \((\sqrt{\nu_r}/\gamma, \tau_2)\), is to cause suppression of the Rabi flop during the pulse. Clearly the potential for coherent manipulation depends on the interplay between these processes.
FIG. 1: (a) Temporal population redistribution, $\Delta n_t$, for the ideal case ($\tau_\perp \to \infty$, $\Phi \to 1$) due to secant (thick solid curve) and Gaussian (dashed curve) pulses of duration $\tau_p$ (FWHM). The pulse profiles are shown as the thin solid (secant) and dotted curves (Gaussian). Panels (i) and (ii) correspond to pulse areas (for secant pulse) $A_p = \pi$ and 2.5$\pi$ respectively. (b) The area theorem is recovered as final distribution is plotted against secant pulse area ($1.193(\sqrt{e/\tau_p})$) for secant (solid curve) and Gaussian (dashed curve) profiles.

To include realistic estimates of these parameters, we take experimentally determined values of coherence times for a typical sample [8, 9]. Four-wave-mixing measurements provide a value for the homogeneous broadening corresponding to an electron-electron scattering time of the order 320 fs. LO-phonon emission times are estimated to be between 1-4 ps. As a conservative estimate we take $\tau_\perp = 1$ ps and calculate the system response to a 100 fs (FWHM) pulse tuned to the $\to 2$ transition at $E = 100$ meV, and allow for a detuning of up to 12 meV. In Figs. 3a,b we show the population redistributions due to these parameters. Although complete inversions are suppressed, we estimate that up to 70-80% of the initial distribution can be coherently excited to the upper level with a resonant $\pi$-pulse: Fig. 3a (i). Effects of pulse area can still be seen in the population 1 ps after the maximum of the pulse intensity. However the question remains as to whether these density flops can be detected experimentally. We now consider two possible scenarios.

Following the interband example [5], we calculate cross-correlation functions comparing changes in pulse modification due to transmission through unexcited and weakly excited samples. With the induced current density written in the standard form: $J_t = (2e/L^2) \sum_{j,j'} \langle j' | p | \delta \rho_t | j \rangle$, we can use Eq. (1) to express the induced current through $\Delta n_t$. The absorbed power, $P_t = J_t E_t$ where the overline means the average over the period, is written in the form:

$$P_t = \frac{(2e v_{\perp} E)^2}{h\omega} \left| w_t^0 \int_{-\infty}^0 dt' w_{t'} e^{t'/\tau_\perp} \cos \Delta \omega t' \Delta n_{t+t'} \right|^2.$$  

(10)
The transmitted intensity of a single pulse is thus given by: $I_s = I_t - P_t$, where $I_t \propto |E_t|^2$. $I_d$, the transmitted intensity of an identical pulse after weak excitation of the sample by a prepulse, is similarly extracted from Eqs. (7)(10) using the two-pulse form-factor, $w_t \to w_{t+\tau}^+ +$


In conclusion, we have developed a simple description of Rabi oscillations between subband levels in low-doped QWs. We have considered the population dynamics of a two-level system under ultrafast mid-IR radiation in a non-Markovian quantum kinetic model. Our calculations, based on realistic pulse and material parameters, show that coherent manipulation of the order of $10^6$ electrons (depending on area of excitation) should be possible under relatively low-power excitations.

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