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Efficient population transfer in modulation doped single quantum wells by intense few-cycle terahertz pulses

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Abstract. We demonstrate the direct observation of non-equilibrium intersubband dynamics in a modulation-doped multiple quantum well sample subject to intense few-cycle terahertz (THz) pulses. The transmission spectra show a distinct dependence on the incident THz field strength and contain signatures of a multitude of nonlinear effects that can be observed owing to the large THz-pulse bandwidth. We focus our attention on a case of transient nonlinear refractive index caused by the efficient transfer of electronic population from the ground state to higher-excited states of the quantum well sample. By comparing the experimental results with a one-dimensional finite-difference model going beyond the slowly varying envelope approximation, we prove that, depending on the pulse shape, the leading part of the intense pulse efficiently transfers electrons from the ground state to higher lying excited states. For weak electric fields and small-population transfer, the linear Lorentz model holds. For strong electric fields, up to 55 and 20% of the ground-state electrons are transferred to the first and second excited subbands, respectively, which could lead to the observation of the optical gain.

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1. Introduction

In recent years, terahertz time-domain spectroscopy (THz TDS) has found widespread application in the spectroscopy of elementary excitations and charge-carrier dynamics in bulk semiconductors and semiconductor heterostructures [1]. The main advantage over other spectroscopic techniques in the far-infrared spectral region, such as Fourier transform infrared spectroscopy, is the coherent generation and detection of electromagnetic pulses that enables the simultaneous and highly sensitive measurement of changes in the amplitude and phase induced by the sample under study. With photon energies in the range of a few tens of meV, THz TDS is the ideal tool for the investigation of intersubband dynamics in semiconductor quantum wells, where the number of quantized energy levels, their spacing and the transition dipole moments can be engineered at the growth stage [2].

In contrast to optical pump-probe experiments, that rely on interband transitions, THz TDS allows the undistorted investigation of the intersubband polarization with subcycle time resolution [3, 4]. For example, this technique has been used to directly observe the gain dynamics, such as gain clamping, inside an active quantum-cascade laser [5]. Furthermore, few-cycle THz pulses have been used successfully to coherently control the intersubband polarization [6, 7], which is a key ability for the observation of many fundamental quantum optical phenomena, such as Rabi oscillations [6, 8, 9], the Autler–Townes effect [10, 11] or electromagnetically induced transparency [12]. One very important ingredient for quantum optics and quantum information processing in general is the ability to prepare the system in a certain quantum state. The simplest example would be the controlled transfer of population from the ground state to any of the excited states, for example, to achieve a complete population inversion between two subbands. Apart from the pure scientific interest, such an inverted system may also be useful as a switchable THz gain medium in real world applications, such as THz amplifiers or Q-switches.

Despite the enormous theoretical interest in the efficient population transfer by pulsed coherent radiation [13–17], only a few experimental reports exist so far. Sherwin et al [18, 19] have demonstrated the undressing of collective excitations by a reduction of the associated depolarization shift using narrowband radiation from a free-electron laser with peak powers of the order of 1 kW. This renormalization of the transition frequency could be successfully explained by a THz-induced population transfer to the first excited subband [20], however,
no quantitative numbers were given. Eickemeyer et al [6] have measured a partial Rabi flop using narrowband mid-infrared pulses resonant with an intersubband transition in a multiple quantum well sample. Thereby, they achieved a transfer of approximately 30% of the ground-state electrons to the first excited subband. Later, Luo et al [8, 9] observed the signature of one and a half-Rabi cycles in the time-domain analysis of the re-emitted electromagnetic field from the same quantum well system using up to five times stronger mid-infrared pulses.

However, to the best of our knowledge, an experimental demonstration of efficient carrier transfer by coherent pulsed excitation in semiconductor quantum wells in the THz spectral region is still lacking. We have recently demonstrated the power-dependent renormalization of the intersubband transition frequency in a multiple quantum well sample under irradiation with intense few-cycle THz pulses [21]. Based on the relative absorption strengths of the three lowest intersubband transitions, we calculated that up to 44% of the ground-state electrons were transferred to the first excited and 8% to the second excited subband, respectively. However, these numbers only give the amount of transferred population averaged over the duration of the THz pulse and not the value which is left in the excited state after the pulse has passed.

The subject of the present paper is the demonstration of a highly efficient transfer of population to the first, second and even third excited subband of a modulation-doped multiple quantum well sample by a single few-cycle THz pulse with a peak electric field of up to 20 kV cm$^{-1}$. The measured electronic contribution to the refractive index of the sample for different values of the driving field is consistent with that of a simple Lorentz medium with a power-dependent strength of the three involved oscillators. We show that both the observed spectrum of the re-emitted radiation and the deduced refractive index are consistent with one-dimensional finite-difference simulations that treat the $N$-level quantum wells in the density matrix formalism. Based on these simulations, we find an almost complete depletion of the ground state during the duration of the driving pulse and demonstrate the inversion of population between the ground and first excited subband.

2. Experiment

Figure 1(a) shows a schematic of the experiment. Further details on the experimental procedure can also be found in [21]. In short, single-cycle THz pulses are focussed on the cleaved facet of a multi-quantum well sample with the electric field oriented parallel to the growth direction. The sample is mounted on a metallic holder with a 5 mm \times 500 \mu m sized aperture to block any bypassing light. After transmission through the 5 mm long sample, which serves also as a waveguide, the THz transient is detected using standard electro-optic sampling. By periodically applying a bias voltage of $-10$ V between an aluminium Schottky gate and Ge/Au Ohmic contacts at a rate of 500 Hz, the electrons can be entirely depleted from the quantum wells for every second THz pulse. This depletion modulation technique allows to selectively measure the electronic response to the strong THz field, $\Delta E = E_{0V} - E_{-10V}$ [3]. The amplitude of the modulation signal in the time domain is about 12% of the total signal transmitted through the depleted structure and can be measured with a signal-to-noise ratio (SNR) of 100:1. Part of this excellent SNR can be ascribed to the length of the sample. At the frequency of the first optical transition (1.5 THz), we achieve a peak modulation depth of 38%, which corresponds to an absorbance of $-\log(T) = 0.85$.

The SNR is further improved by the use of a dual colour detection scheme based on an Er-doped fibre laser (Menlo Systems) that provides both the seed pulses for a regenerative
Figure 1. (a) Scheme of the experiment. MQW: multiple quantum wells. (b) Level scheme of the 52 nm wide quantum wells. The relevant optical transitions are indicated by the arrows. The frequencies have been calculated taking into account the depolarization shift.

Ti:Sapphire amplifier (Spectra Physics), operating at a centre wavelength of 780 nm, and probe pulses for the electro-optic detection at 1560 nm. The setup is conceptionally similar to the one presented in [22]. A 300 µm thick, (110)-cut GaAs crystal is used as electro-optic medium, which exhibits a flat detector response up to 6 THz. The induced birefringence of the probe pulses is directly sampled at the 80 MHz repetition rate of the fibre laser using fast InGaAs photodetectors and a high-speed analogue-to-digital converter (National Instruments). Thus, the full 1 kHz repetition rate of the amplifier can be used, which automatically increases the SNR by $\sqrt{2}$ compared to a conventional THz TDS setup where the same pulse is used for generation and detection of the THz transients. In addition, the photodetectors used for the balanced detection are insensitive to stray light from the intense pump pulses.

The single-cycle THz pulses are generated by optical rectification in a 400 µm thick, large area GaP crystal [23]. To avoid saturation, the pump pulses with a duration of 130 fs and a maximum pulse energy of 3.3 mJ are expanded by two 2-inch lenses to a beam waist of 18 mm. The generated THz pulses have a bandwidth of 5.5 THz with the maximum peak electric field at the sample position being estimated to 20 kV cm$^{-1}$. This field strength has been determined from the electro-optic modulation signal without sample and taking into account the Fresnel transmission coefficients of the silicon windows of the optical cryostat.

The sample itself consists of ten periods of symmetrically modulation-doped, 52 nm wide GaAs wells separated by 160 nm Al$_{0.3}$Ga$_{0.7}$As barriers grown on a 500 µm thick, semi-insulating GaAs substrate. It is mounted in an optical cryostat and is kept at 5 K throughout the entire experiment. At this temperature, the areal density of carriers per well is $n_{2d} = 2.55 \times 10^{10}$ cm$^{-2}$, as has been deduced from capacitance-voltage measurements. Figure 1(b) shows the level scheme of the quantum wells and the allowed transitions that are accessible with the bandwidth of the THz pulse. The transition frequencies have been calculated self-consistently and taking into account the depolarization shift [2]. Only the ground state is significantly populated and, hence, only the fundamental transition $|1\rangle \rightarrow |2\rangle$ is directly possible.

Figure 2 shows the obtained modulation spectra for different values of the incident THz peak field ranging from 0.8 to 12 kV cm$^{-1}$. The spectra have been obtained by taking the Fourier transform of the recorded modulation signals $\Delta E$ that are shown in the insets. There is a clear
Figure 2. Modulation spectra for different values of the incident THz field $E_{\text{THz}}$ obtained by taking the Fourier transform of the modulated THz transmission $\Delta E$ in the time domain (shown in the insets). (a) For weak fields ($0.8 \text{ kV cm}^{-1}$), only the transition $|1\rangle \rightarrow |2\rangle$ at 1.5 THz is visible. Compared with the absorption signal (black dotted line), a pedestal appears in the modulation spectrum as indicated by the arrows. (b) For a driving field of $4.5 \text{ kV cm}^{-1}$, the transition $|2\rangle \rightarrow |3\rangle$ starts to appear at 2.4 THz. The modulation spectrum acquires an asymmetric line shape as indicated by the arrow. (c) At $6.4 \text{ kV cm}^{-1}$ and above, a spectrally broad feature appears between the two lowest transitions (vertical arrow). (d) Around $12 \text{ kV cm}^{-1}$, the third transition $|3\rangle \rightarrow |4\rangle$ becomes visible at 3.3 THz.

dependence on the incident field strength, both in the time and frequency domain. As expected, only the transition from the ground state to the first excited state at 1.5 THz is visible for the lowest pump power. This is also confirmed by the normalized absorption spectrum, plotted as a black, dotted line in figure 2(a), which shows a single, narrow absorption peak centred at the same frequency. The absorption spectrum has been calculated via [24]

$$\alpha(\omega) \propto \Im \left\{ -i \frac{\Delta E(\omega)}{E_{-10 V}(\omega)} \right\},$$

(1)
where $i$ is the imaginary unit and $\Delta E(\omega)$ and $E_{-10\text{V}}(\omega)$ are the Fourier transforms of the modulation and reference signals, respectively. Compared with the absorption spectrum, there appears an additional contribution in the modulation spectrum to the left and right of the transition frequency (indicated by black arrows). As this feature is absent in the absorption spectrum, it has to be related to a modification of the real part of the refractive index. Around an incident field strength of 4.5 kV cm$^{-1}$ (figure 2(b)), the next higher transition $|2\rangle \rightarrow |3\rangle$ starts to appear at 2.4 THz. As all electrons are initially in the ground state, a significant portion has to be transferred to the excited state during the pulse duration. In addition, the line shape of the first transition becomes broader and asymmetric. This broadening is absent in the absorption spectrum (not shown). For 6.4 kV cm$^{-1}$, plotted in figure 2(c), a spectrally broad feature appears centred between the first and second intersubband transition frequencies (black arrow). Again, this feature is not reproduced by the absorption spectrum (not shown). A similar effect has been observed as a result of a ponderomotive current in the plane of the quantum well driven by the THz field under oblique incidence [24]. In this case, however, the THz field is polarized perpendicular to the quantum well plane and, thus, cannot couple to the in-plane motion of the carriers. Figure 2(d) shows the modulation spectrum for a field strength of 12 kV cm$^{-1}$. For such high fields, also the next higher transition from the second to the third excited level ($|3\rangle \rightarrow |4\rangle$) starts to appear around 3.3 THz. Again, the appearance of this transition requires the efficient transfer of population from the ground state to the first and second excited state. In principle, it is possible to infer the occupations of the quantum well levels from the ratios of the areas under the three involved absorption lines [21]. However, these values reflect only the effective occupation integrated over the pulse duration and sample length. Thus, the instantaneous occupations of the different levels are not directly accessible in the experiment. An alternative is to use computer simulations, which directly give insight into the microscopic parameters.

3. Finite-difference density matrix model

To simulate the interaction of the incident THz pulse with the quantum well sample, we have combined a one-dimensional finite-difference time-domain (FDTD) code for the electromagnetic fields with a density matrix approach for multilevel quantum systems. The sample is modeled as a homogeneous slab embedded in vacuum, whereby the background dielectric function of the GaAs substrate is included as a lossless dielectric with a constant refractive index of $n_0 = 3.6$. This is a valid simplification in our frequency range of interest. The computational space is bounded by Engquist–Majda absorbing boundary conditions, which are exact in one dimension [25, 26]. The cell size of the Yee grid is chosen as $\Delta z = 1 \mu$m which ensures a resolution of at least 10 grid cells per wavelength [27].

The quantum well is treated in a single-electron picture, i.e. collective effects such as the depolarization shift are not included. Furthermore, we neglect the electron momentum since the electric field is oriented perpendicular to the quantum well plane and the subband dispersion is assumed to be parabolic. In principle, collective effects have been shown to be important for the quantitative description of population transfer due to the time-dependent renormalization of the intersubband transition frequency [13–17]. However, in this case, the intersubband transition coincides with the maximum of the pump spectrum and shifts only by about 150 GHz [21]. The spectral energy density in this frequency range is approximately constant and, thus, we expect the simplified analysis presented in the following to correctly reproduce our experimental situation.
With the above mentioned simplifications, the quantum well can be described by a density matrix \( \rho \) whose time dependence is given by the von Neumann equation \([28]\):

\[
i \hbar \frac{\partial}{\partial t} \rho = [\mathbf{H}, \rho],
\]

where \( \mathbf{H} \) is the Hamiltonian of an \( N \)-level quantum system in an external field \( E_{ext} \). This approach allows to phenomenologically include elastic and inelastic scattering and yields a set of differential equations for the diagonal and off-diagonal elements of the density matrix, respectively \([29]\):

\[
\frac{\partial}{\partial t} \rho_{nm} = -\frac{i e}{\hbar} E_{ext} \left( \sum_k \mu_{nk} \rho_{km} - \sum_k \mu_{km} \rho_{nk} \right) - (\beta_{nm} + i \omega_{nm}) \rho_{nm}, \\
\frac{\partial}{\partial t} \rho_{nn} = -\frac{i e}{\hbar} E_{ext} \left( \sum_k \mu_{nk} \rho_{kn} - \sum_k \mu_{kn} \rho_{nk} \right) - \sum_{k \neq n} A_{nk} \rho_{nn} + \sum_{k \neq n} A_{kn} \rho_{kk}.
\]

Here, \(-e\) is the electron charge, \( \mu_{nm} \) is the transition matrix element, \( \omega_{nm} = 2\pi \nu_{nm} \) is the associated angular transition frequency, and \( A_{nm} \) is the rate of intersubband population transfer by phonon emission or impurity scattering, for example. This non-radiative carrier transfer adds to the pure dephasing, \( 1/\tau_{nm} \), leading to an effective dephasing rate

\[
\beta_{nm} = \frac{1}{\tau_{nm}} + \frac{A_{nm}}{2}.
\]

We have not included any additional particle loss rates as the total number of electrons in the quantum wells is conserved during the experiment. The density matrix is finally coupled to Ampère’s law via the macroscopic polarization density

\[
P = -ne \sum_{n,m} \mu_{nm} \rho_{nm},
\]

where \( n \) is the number of electrons per unit volume and the polarization of the electric field has been taken to be parallel to the transition dipole moment of the quantum well. Note that \( P \) is a real quantity as the density matrix is Hermitian, \( \rho_{nm} = \rho_{mn}^* \).

The finite-difference implementation of (3a) to (4) is based on a weakly coupled scheme where the density matrix is discretized at the same time steps as the magnetic field \([30]\). In one dimension, this leads to a decoupling of the equations. As a consequence, the Maxwell equations can be solved directly and a predictor–corrector step has to be used only for the material equations. To conserve positiveness of the density matrix, the material equations are split into a Hamiltonian part and a relaxation-nutation part, which can be solved separately \([31]\). The predictor–corrector step usually converges after three to five iterations to a relative precision of better than 0.001%.

As the THz pulse experiences significant dispersion upon transmission through the waveguide, the transient transmitted through the depleted sample, \( E_{-10\%}(t) \), is directly used as the input waveform for the FDTD simulation (similar to \([7]\)). The waveform obtained for an incident electric field of 6.4 kV cm\(^{-1}\) is shown in figure 3(a). This particular value of the driving field has been chosen as it represents an intermediate value showing contributions from all three intersubband transitions. To reduce the noise introduced by abruptly switching the electric field at the beginning of the simulation, a window function has been applied to the experimental data.
Figure 3. Comparison of experimental (blue) and simulated (orange) results for an experimental incident field strength of 6.4 kV cm$^{-1}$. (a) The experimental transmission signal is used as an input for the FDTD code. (b) Modulation signal in the time domain. (c) Modulation spectrum. (d) Absorption spectrum.

The transient is further normalized by the maximum field amplitude before scaling to the field strength used in the simulation.

By iteratively changing the quantum well parameters (transition frequencies, dephasing and population relaxation rates) and the incident field strength, an excellent agreement between the experimental data and the simulation results could be obtained. The results of such a simulation are shown in figures 3(b)–(d) for a peak amplitude of the simulated transient inside the sample of 1.6 kV cm$^{-1}$. The transition frequencies have been determined to be $\nu_{12} = 1.44$ THz, $\nu_{23} = 2.49$ THz and $\nu_{34} = 3.31$ THz. The best correspondence between the simulated and measured full-width at half-maximum (FWHM) line widths and relative amplitudes of the absorption peaks has been found for the following dephasing times and population decay rates: $\tau_{12} = 2$ ps; $\tau_{23} = 1$ ps; $\tau_{34} = 1.3$ ps; and $A_{31} = 3 \times 10^{11}$ s$^{-1}$. The population decay rate describes the non-radiative transfer of population from the upper to lower level. Its main effect is to reduce the relative amplitude of the absorption line associated to the $|3\rangle \rightarrow |4\rangle$ transition. The other decay times are longer than the interaction time between the THz pulse and quantum well system and, thus, do not play a role in the simulation. The transition matrix elements have been
Figure 4. Dependence of modulation spectra on THz peak field strength. The field is increasing from bottom to top. The curves have been offset for clarity. (a) Experimental results for an incident peak field from 0.8 to approximately 21 kV cm$^{-1}$ (from bottom to top: 0.8, 1.6, 2.4, 3.2, 4.0, 4.8, 5.6, 6.4, 7.2, 8.0, 9.6, 11.2, 12.8, 14.4, 16.0, 17.6, 19.2 and 21.1 kV cm$^{-1}$). (b) Simulation results for peak field strengths inside the sample ranging from 0.05 to 2.7 kV cm$^{-1}$ in steps of 175 V cm$^{-1}$.

taken from the solution to the Schrödinger equation for the quantum well: $\mu_{12} = 9; \mu_{23} = 10$; and $\mu_{34} = 11$ nm. The excellent agreement between simulated and measured data shows that the model is capable of reproducing the physics of the experiment.

The additional structure that is visible in the simulated spectra (figures 3(c) and (d)) is a consequence of the complicated shape of the input waveform and may obscure features related to the electron dynamics in the system. Thus, the following simulations have been performed using a model pulse that imitates the experimental transient:

$$E(t) = E_0 \cos(\omega_0 t + ct^2 + \phi_0) e^{-(4 \ln 2) t^2 / \Delta t^2}, \quad (5)$$

with $E_0$ being the peak amplitude, $\omega_0 = 2\pi \times 1.35$ THz the centre frequency, $c = 3 \times 10^{24}$ s$^{-2}$ the linear chirp, $\phi_0 = -1.3$ the phase offset and $\Delta t = 1.8$ ps the FWHM pulse length.

4. Results

4.1. Terahertz field-dependent modulation spectra

Figure 4 shows a comparison of measured and simulated modulation spectra for increasing THz field strengths. In the experiment (figure 4(a)), the incident peak field strength is varied from 0.8 to approximately 20 kV cm$^{-1}$ from bottom to top. In order to get a good correlation between the experimental results and numerical model, the simulated field strength inside the sample had to be varied in the range from 0.05 to 2.7 kV cm$^{-1}$ (see figure 4(b)). The discrepancy between the values for the experimental and simulated electric field strengths is due to several reasons. Firstly, the electric field strength in the experiment refers to the peak value of the incident single-cycle pulse in vacuum, whereas the value in the simulation refers to the dispersed pulse in the substrate. Secondly, the field inside the sample, that effectively interacts with the quantum well
electrons, will be lower in the experiment due to the finite coupling efficiency from free space to the waveguide and the limited spatial overlap of the waveguide mode with the quantum wells. In the simulations, however, the entire electric field interacts with the electrons.

As can be seen in figure 4, the FDTD simulations are able to reproduce correctly the field dependent appearance of the three lowest intersubband transitions and especially the occurrence of the asymmetric line shapes and broad spectral feature between the two transitions \(|1\rangle \rightarrow |2\rangle\) and \(|2\rangle \rightarrow |3\rangle\). This is a further argument against the ponderomotive current as origin of the Fano-like line shapes in our case, because this term is not included in the FDTD code [24]. As these features do not appear in the absorption spectra, they are caused by a coherent modification of the real part of the refractive index. Likewise, these effects have to be related to the intersubband transitions.

4.2. Electronic contribution to the refractive index

To gain more insight into the underlying processes, we have calculated the real part of the refractive index for both experimental and simulated data following [1]. Under the assumption of low absorption, the electronic contribution to the refractive index is given by

$$\Delta n(\omega) = \frac{\phi(\omega)c_0}{\omega L},$$

where \(L = 5\) mm is the sample length, \(c_0\) the vacuum speed of light and

$$\phi(\omega) = \text{arg} \left( \frac{E_{0V}(\omega)}{E_{-10V}(\omega)} \right)$$

the phase of the complex transmission coefficient. From the shape of \(\Delta n\), one can directly infer the shape of the modulation spectra. These consist, essentially, of the refractive index change induced by the intersubband transitions weighted by the input spectrum. Thus, a constant, non-zero value of \(\Delta n\) would show up in the modulation signal as an exact replica of the input spectrum.

The results for different values of the incident field strength are displayed in figure 5 as dots. There is an excellent agreement between the experimental and simulated values for \(\Delta n\). The solid lines are results of a least squares fit of a sum of three Lorentzians

$$\Delta n_{\text{model}}(\omega) = \Re \left\{ \left( 1 + \sum_{k=1}^{3} \frac{\Lambda_k}{\omega_k^2 - \omega^2 + i \Gamma_k \omega} \right)^{1/2} \right\} - 1,$$

where \(\Lambda_k\), \(\omega_k\) and \(\Gamma_k\) are amplitude, position and width of the Lorentzian oscillators, respectively. Equation (8) corresponds thereby to the contribution of the three involved intersubband transitions to the linear refractive index [28]. The amplitude factor includes the oscillator strength of the transition and the occupation difference of the upper and lower quantum well state averaged over the pulse duration.

For weak driving fields, as shown in figures 5(a) and (e), only the first transition at 1.5 THz is active. The line shape of \(\Delta n\) is perfectly reproduced by a single Lorentzian oscillator. For increasing field strengths, the next higher transition eventually becomes visible (figures 5(b) and (f)). Apart from the additional resonance line, the main effect of the second oscillator is to add an offset to the first oscillator. This causes an increasing asymmetry of the line shape of the first transition in the modulation spectrum, while the shape of the absorption peak is
Figure 5. Electronic contribution to the refractive index (dots) and least-squares fit of a Lorentz model (solid line) for both experimental (a–d) and simulated data (e–h) at different peak field strengths: (a) 0.8 kV cm$^{-1}$, (b) 4.5 kV cm$^{-1}$, (c) 6.4 kV cm$^{-1}$, (d) 21 kV cm$^{-1}$, (e) 0.05 kV cm$^{-1}$, (f) 0.9 kV cm$^{-1}$, (g) 1.8 kV cm$^{-1}$ and (h) 2.7 kV cm$^{-1}$. The quality of the fit degrades for increasing pump powers.

not affected. As the second oscillator gains strength due to the increasing number of electrons populating the upper quantum well levels (figures 5(c) and (g)), the entire part of $\Delta n$ to the left of the second transition is raised above the zero line. As mentioned previously, this causes the appearance of a spectrally broad feature in the modulation spectrum, which is essentially
Figure 6. Simulated temporal evolution of the relative level occupations for different peak field strengths in the sample: (a) 0.05 kV cm$^{-1}$, (b) 0.9 kV cm$^{-1}$ and (c) 2.7 kV cm$^{-1}$. (d) Same as (c) but without chirp. The normalized instantaneous THz electric field is shown for reference as grey line.

There is a clear dependence of the observed refractive index on the incident THz field strength which is due to a coherent transfer of electrons to higher lying states during the duration of the THz pump pulse and the connected modification of the linear refractive index responsible for the phase delay experienced by the very same THz pulse. To confirm this hypothesis, we have calculated the temporal evolution of the relative level occupations of the multilevel quantum well at the entrance facet of the sample. These are given by the diagonal elements of the density matrix, $\rho_{ii}$, and are labelled $|1\rangle$ through $|4\rangle$, respectively. The results of these calculations are plotted in figure 6 for three different values of the incident electric field strength along with a replica of the incident pulse spectrum. For the highest field strengths, shown in figures 5(d) and (h), respectively, also the effect of the third optical transition appears in the refractive index change. This additional oscillator again introduces an offset to the first and second oscillator. While the Lorentzian oscillator model (8) perfectly reproduces both experimental and simulated datasets for low driving fields, the quality of the fit to the first transition strongly degrades with increasing THz pump power. For the highest driving field, the quality of the fit to the second transition peak is also affected.

4.3. Population dynamics

There is a clear dependence of the observed refractive index on the incident THz field strength which is due to a coherent transfer of electrons to higher lying states during the duration of the THz pump pulse and the connected modification of the linear refractive index responsible for the phase delay experienced by the very same THz pulse. To confirm this hypothesis, we have calculated the temporal evolution of the relative level occupations of the multilevel quantum well at the entrance facet of the sample. These are given by the diagonal elements of the density matrix, $\rho_{ii}$, and are labelled $|1\rangle$ through $|4\rangle$, respectively. The results of these calculations are plotted in figure 6 for three different values of the incident electric field strength along with
the instantaneous electric field of the THz pulse (grey line). In all four cases, all the electrons are initially in the ground state.

Figure 6(a), which has been calculated for a peak field of 0.05 kV cm⁻¹, corresponds to the refractive index modification shown in figure 5(e). In this case, the simple Lorentzian oscillator model adequately describes the refractive index change induced by the quantum well electrons. The driving field is weak enough that all electrons remain in the ground state. Thus, the refractive index modulation just shows a single Lorentz oscillator corresponding to the transition from the ground state to the first excited state.

When the field strength is increased, a pronounced effect on the subband populations becomes apparent. Figure 6(b) shows the population for a field amplitude of 0.9 kV cm⁻¹ corresponding to figure 5(f). As the population relaxation rate from the first excited state to the ground state has been set to zero, the decrease of population in level |2⟩ (and the increase in level |1⟩) is related to the coherent driving of the system. The fast oscillatory features overlaying the population curves are due to the instantaneous electric field associated to the high-frequency carrier and would not be observable in the usually adopted rotating wave approximation. After the pulse has passed, most electrons (about 72% versus 27% and 1% for the first and second excited state) reside in the ground state and, therefore, the linear Lorentz model still gives a good approximation to the observed refractive index. Thereby, the amplitude $\Lambda_k$ of the Lorentzian oscillators is determined by the occupation numbers averaged over the pulse duration. As the population of the third level relaxes back to the ground state at a rate of $3 \times 10^{11}$ s⁻¹ (see above), we have chosen to use the fractional occupation numbers at a time of 7 ps. At this time, the electric field of the pulse does no longer drive any significant coherent transfer, while the influence of the population relaxation is still negligible.

When the system is driven strong enough such that the effect of the THz electric field on the level occupations can no longer be considered a weak perturbation, the Lorentz model will eventually break down. Figure 6(c) shows the calculated level occupations for a driving field of 2.7 kV cm⁻¹. After the pulse has passed, only 24% of all electrons reside in the ground state, while 55%, 20% and 1% of the electrons have been transferred to the first, second, and third excited states, respectively. This ultrafast build-up of an inversion of population between the ground state and the first excited state leads to a time varying sign of the electronic susceptibility associated with the first optical transition $|1⟩ \rightarrow |2⟩$. While the first part of the THz pulse experiences loss, the second part experiences optical gain at the transition frequency (1.5 THz). This dynamical change from a normal to a gain medium within the pulse duration manifests as a non-Lorentzian line shape of the (averaged) refractive index, as is shown in figures 5(d) and (h). For this power level, the effect of the population relaxation between levels $|3⟩$ and $|1⟩$ is clearly visible.

Because the observed population transfer is a coherently driven process, its dynamics and the final number of transferred electrons are strongly dependent on the magnitude of the electric field and exact shape of the driving pulse. As has been mentioned above, the THz pulses used in our experiment are initially single-cycle pulses. By passing through the sample, the pulses are dispersed and finally acquire the temporal shape that has been used in the simulations so far. Hence, the electrons experience a different driving field at different positions along the waveguide. Despite the fact that the simulations are in excellent agreement with the experimental results, it is worth investigating the influence of the pulse shape on the achievable population transfer. Figure 6(d) shows the calculated populations for a single-cycle THz pulse having the same spectral width and amplitude as the one used in figure 6(c).
This is achieved by setting $E_0 = 5.3 \text{ kV cm}^{-1}$, $c = 0$, $\phi_0 = 0$ and $\Delta t = 0.47 \text{ ps}$ in (5). This pulse shape corresponds to the single-cycle THz pulse that enters the sample when we assume that no additional absorption occurs inside the waveguide. Comparing figures 6(c) and (d), we find that both chirped and unchirped pulses lead to qualitative similar population dynamics. Owing to the shorter pulse length of the unchirped pulse, the coherent transfer occurs on a shorter time scale and is less subject to dephasing and loss and, thus, more electrons can be transferred. At a time of 5.5 ps, just after the pulse has passed, only 8% of all electrons are in the ground state, while the majority, 71%, occupies the first excited state. The population inversion reaches a value of $w = \rho_{22} - \rho_{11} \approx 0.6$ which corresponds to a pulse area of approximately $0.7 \pi$. Thus, the system undergoes almost a complete half-Rabi cycle. A limiting factor for the inversion is the additional transfer of electrons to higher subbands. The second and third excited state show a fractional occupation of 20% and 1%, respectively. Note, that the simulations do not account for the renormalization of the transition frequencies caused by changing occupations. However, as discussed above, we do not expect this to have a significant effect in this case due to the broad (and flat) spectrum of the THz pulses. Therefore, the final occupation numbers achieved experimentally can be assumed to lie in between the values obtained for the two limiting cases plotted in figures 6(c) and (d), respectively.

5. Summary

In summary, we have demonstrated the direct observation of the nonlinear propagation of intense few-cycle THz pulses through a modulation-doped multiple quantum well sample. The modulation spectra could excellently be reproduced by one-dimensional finite-difference simulations that included the quantum wells via the density matrix formalism. For low-field strengths, the electronic part of the refractive index is perfectly modeled by a sum of three Lorentzians corresponding to the three lowest intersubband transitions. For increasing field strengths, however, the quality of the fit strongly degraded. We could attribute this behaviour to the build-up of population inversion between the ground state and the first excited state during the pulse duration. Finally, we could demonstrate the efficient transfer of 55% and 20% of the population from the ground state to the first and second excited states, respectively. This result holds great promise for further quantum optics experiments, as well as for applications such as a THz amplifiers or THz $Q$-switches.

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