Impact of femtosecond interband pumping on terahertz gain of quantum-cascade structures

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Abstract
Ultrashort terahertz pulses are a key prerequisite of terahertz time-domain spectroscopy. The generation of high-power terahertz pulses in quantum-cascade structures is limited by gain clamping, which can, however, be overcome for a short period of time using ultrashort interband pumping. In order to clarify this possibility, the terahertz gain of a quantum-cascade structure under irradiation by femtosecond pulses of various energies should be quantified. Using the density matrix and many-body theory, the influence of femtosecond interband pumping on the terahertz gain of quantum-cascade structures is characterized. The resulting dynamical gain spectra of a three-well GaAs/AlGaAs benchmark quantum-cascade structure show increasing terahertz gain up to an order of magnitude under pumping by 100 fs optical pulses with intensity in the active region 100 MW cm⁻². The gain switching process is characterized by a 1 ps rise time and 8 ps recovery time. The short rise and recovery times and large increment of the terahertz gain can be used to develop sources for terahertz time-domain spectroscopy.

Keywords: quantum cascade structure, interband optical pumping, THz pulse amplifier, density matrix, Liouville equation, gain clamping

1. Introduction
Nowadays, terahertz time-domain spectroscopy is considered to be a promising method for detecting explosives and chemical, biological, radiological and nuclear (CBRN) agents [1]. A key prerequisite for implementation of this detection technology is high-power broadband terahertz pulses. In [2], ultrafast gain switching in a quantum-cascade laser has been proposed as an approach for generating such pulses. The gain switching was implemented and demonstrated by embedding the Auston switch into the semiconductor structure. In this way, the authors used ultrashort electronic pulses generated in the Auston switch to implement gain switching. Another possible approach to achieve gain switching is the direct optical pumping of the active region of a quantum-cascade structure (QCS) by stimulating the interband optical absorption using visible and infrared ultrashort pulses. Here we estimate the feasibility of the approach proposed by applying density matrix theory to describe the optical response of a QCS under ultrafast interband optical pumping and highlight a possible experimental implementation of the approach proposed. The model is based on simplified density matrix theory [3], which describes intersubband electron transport and optical gain mediated by coherent electron tunneling and intersubband scattering.

2. Benchmark structure
As an amplifying medium for this benchmark study, we take the well-known three-well QCS operating in the terahertz frequency range [4]. Before any interactions occur with the
pump radiation, the QCS operates just above the threshold in continuous wave (CW) mode. This regime is characterized by the current density $J = 800 \, \text{A cm}^{-2}$, internal electric field of the active region $E = 12.5 \, \text{kV cm}^{-1}$ and lattice temperature $T = 10 \, \text{K}$. More information about the considered QCS can be found in [3–5]. In order to increase the gain coefficient temporarily, the QCS is irradiated by a femtosecond pump pulse characterized by a duration of 100 fs and a peak intensity $I = 1.455 \, \text{MW cm}^{-2}$, internal electric field $E_{\text{pump}} = 1.458 \, \text{eV}$ and a peak intensity.

The overlap integrals are shown in figure 2. The largest magnitudes of an overlap integral correspond to transitions between two top valence subbands $v_{1}$ and the lowest conduction subband $c_{1}$. The corresponding transitions energies are 1.455 eV and 1.458 eV. As illustrated in figure 1, these pairs of subbands belong to the injectors. The subband $v_{1}$ is strongly coupled to the higher laser subband $c_{2}$ in the active region via tunneling transitions. Therefore, increasing the electron density in the subband $v_{1}$ is accompanied by an increase in the population in the subband $c_{2}$, which belongs to the active region. What is also evident is that these transport channels are characterized by the smallest transition energies and are well separated from other interband transitions on the energy scale, giving rise to more precise control.

3. Mathematical description

3.1. Simplified density matrix

To describe the kinetics of the transport processes, we adopt density matrix theory. In what follows, we will use elements of the density matrix averaged over $k$-space, benefiting from the simplified mathematical description. The $k$-space averaging is based on the assumption that the electrons in each subband have a quasi-equilibrium distribution function and the intersubband transport processes, induced by the optical pumping, are much slower than the thermalization of electrons in the subbands [7]. The Liouville–von Neumann equation for the averaged density matrix reads:

$$\dot{\rho} = [H_{\text{coh}}, \rho] + H_{\text{inc}}$$

where:

$$H_{\text{coh}} = \begin{pmatrix}
E_{1} & \Omega_{12} & \Omega_{13} & \Omega_{14} & 0 \\
\Omega_{12} & E_{2} & \Omega_{23} & \Omega_{24} & 0 \\
\Omega_{13} & \Omega_{23} & E_{3} & \Omega_{34} & 0 \\
\Omega_{14} & \Omega_{24} & \Omega_{34} & E_{4} & 0 \\
0 & 0 & 0 & 0 & E_{p}
\end{pmatrix}$$

$$H_{\text{inc}} = \begin{pmatrix}
\Sigma(1) - \Sigma_{p} & \gamma_{12} & \gamma_{13} & \gamma_{14} & 0 \\
\gamma_{21} & \Sigma(2) - \Sigma_{p} & \gamma_{23} & \gamma_{24} & 0 \\
\gamma_{31} & \gamma_{32} & \Sigma(3) - \Sigma_{p} & \gamma_{34} & 0 \\
\gamma_{41} & \gamma_{42} & \gamma_{43} & \Sigma(4) - \Sigma_{p} & 0 \\
0 & 0 & 0 & 0 & \Sigma_{p}
\end{pmatrix}$$

$$Q = \begin{pmatrix}
\varrho_{11} & \varrho_{12} & \varrho_{13} & \varrho_{14} & 0 \\
\varrho_{21} & \varrho_{22} & \varrho_{23} & \varrho_{24} & 0 \\
\varrho_{31} & \varrho_{32} & \varrho_{33} & \varrho_{34} & 0 \\
\varrho_{41} & \varrho_{42} & \varrho_{43} & \varrho_{44} & 0 \\
0 & 0 & 0 & 0 & \varrho_{p}
\end{pmatrix}$$

where $E_{j}$ is the energy of the $j$th subband edge, $\Omega_{ij}$ is the Rabi frequency, $\Sigma_{\text{exc}}(j)$ is the scattering rate for the $j$th subband, $M$ is the set of subbands participating in in- and out-scattering events with regard to the subband $j$ and $\gamma$ is the dephasing rate.

The density matrix elements are directly related to the sheet density of electrons in the subband $N_{j}=\varrho_{j}/S$ and to the

**Figure 1.** Subband edges for the three-well QCS as a part of the terahertz pulse amplifier.

**Figure 2.** Overlap integral spectrum for the set of interband electron transitions.
quasi-microscopic polarization \( P_i = q_i / S \) between a pair of subbands with \( i \neq j \). Here \( S \) is the cross-sectional area of the quantum well layers. The prefix ‘quasi-’ is used to distinguish this value from the wave-vector-dependent microscopic polarization \( P_k \) and the macroscopic polarization \( P \), which also includes contributions from all subbands.

The optical response of a QCS is strongly affected by electron–impurity and electron–phonon scattering. We assume that the thermalization time in each subband is very small. In this case, intrasubband scattering can be described in terms of intersubband lifetimes averaged over the distribution function. The lifetimes are computed with Fermi’s golden rule using a series of approximations as discussed in [8].

3.2. Differential transmission spectra

In a possible experimental implementation, the pump and probe signals need to be distinguished. In a computational experiment, this can be done by extracting information for the probe signal from the time-dependent dielectric polarization of the medium [9]. This is implemented by a two-step computation [10]: in the first stage, the total contribution to the polarization from both the pump and the probe signals is computed; in the second stage, one computes polarization changes caused by the pump signal only. With these two polarizations \( Q_{ij}^{pp} \) and \( Q_{ij}^{pr} \), both depending on time, the absorption coefficient for the probe beam is computed as follows:

\[
\alpha_{p,pr}(\omega, \tau) = \frac{\omega}{Vcn} \text{Im} \left\{ \frac{3 \{ p_{23}^{pp}(t) \}}{3 \{ e^{pr}(t) \}} \right\}
\]

(5)

where:

\[
p_{23}(t) = d_{23}^p q_{12}^{pp}(t) + d_{23}^e q_{23}^{pp}(t) - d_{23}^e q_{12}^{pr}(t) - d_{23}^p q_{23}^{pr}(t)
\]

(6)

In pump–probe experiments, one usually measures the differential transmission spectra, which can be easily obtained using equation (5):

\[
\Delta \alpha(\omega, \tau) = \alpha_{p,pp}(\omega, \tau) - \alpha_{p,pr}(\omega)
\]

(7)

where \( \alpha_{p,pr}(\omega) \) is the absorption spectrum without the pump pulse.

4. Results and conclusions

Solving equation (1), one obtains the time-dependent electron sheet densities and quasi-microscopic polarizations. As a general feature, the system kinetics are characterized by an almost immediate increase of the electron population in the subband (1) due to optical absorption and subband (2) due to strong tunneling coupling to the subband (1). At the same time, the subband populations in (3) and (4) increase more slowly due to the weak coupling to the subbands (1) and (2) via phonon-assisted scattering events. Such a difference in the kinetic rates results in a temporary increase of the population inversion between the subbands (2) and (3). As a result, the increase in the gain can be observed due to the appearance of additional polarization.

The optical response of the benchmark structure in the pump–probe configuration is shown in figure 3. Each time point in the figure represents the gain spectrum computed for some defined time delay and carrier frequency of the pump signal. The characteristic shows that interband pulse pumping produces a maximal increase of the gain at a time delay of \( \approx 1 \) ps, which is caused by the finite tunneling rate from the injector to the active region. For the pump peak intensity \( I = 50 \, \text{MW cm}^{-2} \), the gain coefficient at the maximum is eight times larger than its magnitude when there was no pumping. The gain recovery time is \( \approx 8 \) ps, as can be seen from figure 3.

For the Auston switch configuration of [2], the maximal gain is reached nearly 60 ps after the Auston switch is turned on, while a QCS with interband optical pumping has a maximal gain after 1 ps. Moreover, the gain recovery after reaching this maximum is \( \approx 90 \) ps for the Auston switch configuration, while in direct optical pumping the recovery time is a low 8 ps, as can be deduced from figure 3.

4.1. Possible device implementation

The theoretical results obtained and discussed in the preceding sections show that there are no fundamental obstacles for gain switching in a terahertz QCS via ultrafast optical pumping. The benchmark structure can be pumped by Ti: sapphire ultrafast oscillators, as the transition energies used for gain switching correspond to the wavelengths \( \lambda = 852.12 \) nm and \( \lambda = 850.37 \) nm, which are within the tuning range. However, thinking about a possible device implementation, we can identify one hindrance: the coupling of the ultrashort pumping pulse to the QCS. As we discussed in the previous sections, the wavelength of the pumping pulse is chosen to provide good excitation of the carriers in the conduction subband (1). Thus, the optical pulse is expected to be heavily absorbed. The absorption depth, or equivalently the penetration depth, can be estimated from the Beer–Lambert–Bouguer law. Using absorption \( \alpha = 1000 \, \text{cm}^{-1} \), which is easily achieved in our case, we can estimate the penetration depths for incident intensity levels of \( \exp(-2) \) and 0.01 to be \( D_{1/2} = 20 \, \mu \text{m} \) and \( D_{0.01} = 46 \, \mu \text{m} \), respectively. These values are rather small in comparison to the typical cavity length of edge-emitting lasers.
Thus, the schemes used in [11, 12] are impractical in our case, and their straightforward implementation is likely impossible.

However, the estimated penetration depths are much larger than the active region thickness. The ratio of the penetration depth to the thickness of one cascade (395.5 Å [3]) is 505 and 1163 for the $D_{-0.01}$ and $D_{0.01}$ cases, respectively, which significantly exceeds the typical number of cascades (20–50). Thus, normally incident optical pumping could be appropriate. This pumping scheme can be realized by etching a suitable window in the ridge contact or by using transparent electrodes [13–15]. Also the pumping beam could be injected through the side surfaces of the waveguide ridge.

5. Conclusion

The obtained results evidence that the interband optical pumping of a QCS by ultrashort pulses can be used to increase the terahertz gain, avoiding the formation of ultrashort electronic bursts by an Auston switch and the delayed response of a QCS due to the transport of non-equilibrium electrons to the active region of the QCS. The considered benchmark three-well QCS in a GaAs/AlGaAs material system demonstrates a terahertz gain that increases by up to an order of magnitude under pumping by 100 fs optical pulses with an intensity in the active region of 100 MW cm$^{-2}$. The gain switching process is characterized by a 1 ps rise time and 8 ps recovery time. Thus, the ultrafast interband optical pumping could be utilized in QCSs to amplify terahertz ultrashort pulses for terahertz time-domain spectroscopy. Since there is no fundamental limitation for the implementation of this method, its application will be determined only by the technical possibilities for the realization of the efficient coupling/guiding of the pump radiation to the active region of a QCS.

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