Fano profile in the intersubband terahertz response of photoexcited GaAs/AlGaAs quantum wells

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Abstract. In our work we probe the conduction intersubband transition of an undoped GaAs/Al₀.₃₄Ga₀.₆₆As multiple quantum well via broadband terahertz pulses after resonant photoexcitation at the 1s heavy-hole exciton. The pump-induced change in the transmitted terahertz field shows a strong beating. In the frequency domain this results in an asymmetric Fano-like line shape for the intersubband resonance and an additional broad low-frequency peak. However, the total THz absorption shows only the single symmetric peak of the intersubband transition. In our microscopic theory these signatures unambiguously originate from the phase sensitive superposition of ponderomotive and terahertz intersubband currents.

1. Introduction
Terahertz (THz) spectroscopy on semiconductor heterostructures has revealed new insight in low-energy excitations and their dynamics. A microscopic analysis of these experiments has to take into account the true THz transitions, but also the so-called ponderomotive current. This current results from the classical, electric-field induced charge acceleration according to \( \hbar \dot{k} = -e E_{\text{THz}} \) [1] that causes a wiggling motion of the carriers. In recent experiments the influence of the ponderomotive current could only be seen rather indirectly [2, 3]. Here, we study the intersubband transition of multi quantum wells via THz spectroscopy and directly observe signatures of the ponderomotive current [4]. In the differential transmission spectra we measure a characteristic Fano-like line shape originating from the superposition of ponderomotive and true THz intersubband currents. Usually, asymmetric Fano line shapes result from the interference between a discrete level and a continuum of states [5, 6].

2. Experimental
Spectrally broadband THz pulses are generated in a 55 µm thin z-cut GaSe crystal (see Fig. 1(a)) by phase-matched difference frequency mixing [7] within ultrashort 12 fs optical pulses from a 78 MHz Ti:sapphire oscillator (Femtolasers: Femtosource Scientific sPro). The THz beam is focused on the multi quantum well (MQW) sample. The THz field has a strong component normal to the MQW plane to couple to the intersubband transition. Phase-matched electro-optic sampling [8] allows for field-resolved detection in a second 30 µm thin z-cut GaSe crystal.
The MQW sample consists of 60 8.2-nm-thick undoped GaAs quantum wells, separated by 19.6-nm-thick barriers of Al0.34Ga0.66As. It was prepared as a 38-degree wedged waveguide and an Al0.34Ga0.66As spacer layer of 300 nm thickness was grown on top for improved overlap of THz beam and MQWs. It was kept at a temperature of 6 K in a liquid He flow cryostat.

For photoexcitation (see Fig.1(b)) an optical interband pump pulse either from a spectrally narrow 2.5 ps or a broad-band 100 fs 78 MHz laser (both Spectra Physics: Tsunami) excites the MQWs resonantly at the 1s heavy-hole exciton around $h\omega_{opt} = 1.56$ eV. Only the first conduction subband is populated. The c1-to-c2 intersubband transition is then probed by THz pulses 25 ps after excitation. The photoexcited carrier density is around $2 \times 10^{10} \text{cm}^{-2}$ per quantum well. The transmitted THz transients with and without excitation are simultaneously measured. To this end both pump and THz probe beams are chopped at the same time at different frequencies. The first Lock-In amplifier locks on the modulation of the optical pump beam. Twice its signal yields the differential transmission $\Delta E(t)$, i.e. the pump-induced change in the transmitted THz field. The second Lock-In amplifier detects the mean value between transmission with and without excitation. The reference $E_{ref}(t)$ without excitation follows by subtracting $\frac{1}{2}\Delta E(t)$.

3. Results

After ps photoexcitation the time-resolved differential transmission $\Delta E(t)$ shows a strong beating (Fig. 1(c)(i)), superimposed on the dephasing of a reradiated THz-induced intersubband polarization [9]. The beating is connected to a broad low-frequency peak around 20 THz in the spectral amplitude $|\Delta E(\omega)|$ (Fig. 1(c)(ii)) while the intersubband resonance at 27.3 THz displays a Fano-like asymmetry with an undershoot at the low-frequency side.

To explain our experimental findings, the THz transmission of a quantum well positioned at $z = 0$ is computed ($z$ is the growth direction). The THz response follows from the wave equation

$$\left( \nabla^2 - \left( \frac{\omega^2}{c^2} \right)^2 \frac{\partial^2}{\partial t^2} \right) E(\mathbf{r}, t) = \mu_0 \delta(z) \frac{\partial}{\partial t} \left( J_A(t) + J_{THz}(t) \right)$$

($n_b$ is the background refractive index). Here a delta function is used since the quantum well width is much smaller than the THz wavelength. On the right hand side of the equation the induced current density contains $J_{THz}$, the current due to the intersubband transition, and $J_A$, the THz field induced ponderomotive motion of the excited carriers. The solution yields the differential transmission $\Delta E \propto J_A + J_{THz}$, which can be interpreted as a field reemitted by the current density that is directly proportional to the induced current [10].

$J_A$ is defined via $J_A = -\sum_\lambda (e^2 n_\lambda/m_\lambda) A_{THz}$ with the effective mass $m_\lambda$ and the carrier density $n_\lambda$ in band $\lambda$ ($\lambda$ includes both bulk-band index and subband index). The vector potential $A_{THz}$ of the THz probe pulse is given by $E_{THz} = -\frac{\partial}{\partial t} A_{THz}$. Thus, $J_A$ directly follows $A_{THz}$ with an
opposite phase. Since $J_A$ leads to a real-valued linear susceptibility $\chi_A(\omega) = -\omega_p^2/\omega^2$ (with the plasma frequency $\omega_p$), it does not contribute to the absorption $\alpha(\omega) \propto \omega \Im[\chi(\omega)]$.

The THz current is given by $J_{\text{THz}}(\omega) = \frac{i}{2} \sum_{\lambda,l',k} J_{\lambda,l',k}^\lambda p_{\lambda,l',k}$, with the quantization area $S$, the matrix elements $j_{\lambda,l'}^\lambda$ and the microscopic intersubband polarization $p_{\lambda,l',k}$ between subbands $l$ and $l'$ of bulk band $\lambda$. $j_{\lambda,l'}^\lambda = -\frac{i \hbar}{m^*} \int dz \xi_{\lambda,l}(z) \frac{\partial}{\partial z} \xi_{\lambda,l'}(z)$ is the intersubband matrix element with the conﬁnement wave function of the carriers $\xi_{\lambda,l}(z)$. $p_{\lambda,l',k}$ is computed microscopically with an equation-of-motion approach. Regarding only the c1-to-c2 transition, the sum is restricted to $\lambda = c$ and $l,l' \in \{1,2\}$. In analogy to the semiconductor Bloch equations [11], one finds for $p_{1,2,k}$: $i \hbar \frac{\partial}{\partial \tau} p_{1,2,k} \approx \left( \tilde{\epsilon}_{2,k} - \tilde{\epsilon}_{1,k} \right) p_{1,2,k}^\lambda - i \gamma p_{1,2,k} + \left( f_{c1}^1 - f_{c2}^2 \right) \left[ j_{2,1}^L A_{\text{THz}} - \sum_{q,k} V_{k-q} p_{1,2,q} \right]$, with the renormalized single-particle energies $\tilde{\epsilon}_{k}^\lambda$, the optically excited carrier distributions $f_{c1}^1$ in the conduction bands and the Coulomb matrix element $V_q$. Optically excited interband coherences have already decayed and do not contribute. All scattering effects are modeled by a phenomenological dephasing constant $\gamma$ to match the measured 440 fs decay.

We compute the single-particle energies via $k \cdot p$-perturbation theory. For the THz-response, we include the first two conduction subbands and the first heavy-hole and light-hole subband. The experimental THz reference transient serves as input for the time-dependent fields.

For ps excitation (Fig. 1(c)(ii)) the theoretical result (red line) as the absolute value of the computed current density, i.e. the differential transmission $|\Delta E(\omega)|$, is in excellent agreement with the measurement (shaded area). In the case of broadband fs excitation the low-frequency peak and the Fano-like asymmetry of the intersubband resonance are enhanced (Fig. 2(a), measured: black dashed line; computed: red solid line). These features are also affected by the THz probe spectrum. Shifting its center away from the previous 23.5 THz (Fig. 2(a) inset, black line) to 26.5 THz (inset, grey line) decreases the broad low-frequency peak while the asymmetry of the resonance increases (Fig. 2(a), measured: grey dashed line; computed: blue dotted line). In Fig. 2(b) the total current density for the low-frequency THz probe spectrum (red solid line) is broken up in its individual contributions, i.e. the ponderomotive current density $J_A$ (black dotted line) and the intersubband current $J_{\text{THz}}$ (green dashed line). $J_A$ follows mainly the THz probe spectrum, while $J_{\text{THz}}$ contains the intersubband resonance weighted with the THz probe.
spectrum. The observed Fano-like line shape in the differential transmission results from a phase sensitive superposition of $J_A$ and $J_{THz}$: $|I_{total}| = |J_A + J_{THz}|$. Below the intersubband resonance both contributions partially compensate each other whereas they interfere constructively above. Thus, the resonance is narrowed at the low-frequency side and broadened at the high-frequency side, leading to an asymmetric Fano-like line shape. By shifting the THz probe spectrum mainly $J_A$ is affected and shifted accordingly, explaining the observed behavior in Fig. 2(a). However, there is no Fano signature in the THz absorption $\alpha(\omega) = 2\text{Im}[-i\Delta E(\omega)/E_{ref}(\omega)]$ (Fig. 2(c), shown for the low-frequency THz spectrum).

In the case of fs excitation we saw an enhanced ponderomotive contribution. The fs pulses are spectrally broader than the linewidth of the 1s exciton resonance and thus they are partially (46%) transmitted through the quantum well film and absorbed in the GaAs substrate. Since the ponderomotive current $J_A$ is proportional to the carrier density, $J_A$ is increased, but not $J_{THz}$. Consequently the carrier density in the wells plus substrate is 1.85 times larger than in the wells alone, which can be modeled by increasing $J_A$ relative to $J_{THz}$ by 1.85.

It would be interesting to check whether the ponderomotive contribution could be seen in standard Fourier transform infrared (FTIR) spectroscopy while modulating the carrier density, e.g. by photoexcitation. Since FTIR spectroscopy detects only intensities one would record a differential intensity change $\Delta I(\omega)$ of the transmission without and with excitation, respectively, of the form $\Delta I(\omega) = I_{\text{without exc.}}(\omega) - I_{\text{with exc.}}(\omega) \propto |E_{\text{ref}}(\omega)|^2 - |E_{\text{ref}}(\omega) + \Delta E(\omega)|^2$. In the case of fs photoexcitation Fig. 2(d) presents the simulation of $\Delta I(\omega)$ based on our field-resolved data for $E_{\text{ref}}(t)$ and $\Delta E(t)$. It turns out that the ponderomotive contribution has vanished almost completely compared to a field-resolved measurement.

4. Summary

In summary, we have observed strong Fano signatures in the differential transmission spectra of broadband THz pulses probing an undoped GaAs/AlGaAs multiple quantum well after resonant photoexcitation. In our microscopic theory, the Fano-like line shape of the intersubband resonance is unambiguously attributed to a phase sensitive interference of the sharp intersubband resonance with the effective continuum provided by the light-matter interaction through the ponderomotive contribution. Finally, the applied field-resolved method is better suited than intensity-based measurements for revealing the ponderomotive influence to the THz response.

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