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Phonon sidebands of intersubband absorption in AlGaN/GaN high-electron-mobility transistors

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Abstract

Femtosecond two-color pump-probe experiments in the mid-infrared are performed on intersubband transitions in a GaN/$Al_{0.8}Ga_{0.2}N$ heterostructure. In these experiments, we observe around zero time delay distinct phonon sidebands, which disappear because of spectral diffusion with a time constant of 80 fs. The signal itself decays with a time constant of 380 fs. The independent boson model agrees very well with all observed results.

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1. Introduction

The energy of intersubband (IS) transitions in semiconductor heterostructures can be tailored in a broad range within a given material combination by changing the geometric dimensions. This approach is limited by the available band offsets, so that quantum cascade lasers [1–3], which use IS transitions for lasing, are working mostly in the mid- to far-infrared spectral range. A material combination that allows high IS energies is GaN/AlGaN. In this system, IS absorption at the communication wavelength has been demonstrated [4–6]. A problem with this material system is that often the IS linewidth is quite broad, which in a laser would lead to low gain. To find out whether these broad lines are caused by intrinsic properties of the material or by imperfections of a particular sample, we performed ultrafast two-color pump-probe measurements on the IS transition of a GaN/AlGaN heterostructure. It is found that the properties of the IS transition are determined to a large extent by the strong intrinsic electron–phonon coupling, even leading to the appearance of phonon sidebands of the IS line [7,8].

2. Experiment

We investigate a heterostructure grown by molecular beam epitaxy on a c-face sapphire substrate originally developed as a high-electron-mobility transistor [9]. The sequence of layers is shown in Fig. 1(a). At the interface between the thick, relaxed GaN layer and the AlGaN layer there is a large electric field, caused by the spontaneous polarization in the wurtzite structure and by the piezoelectric field due to the lattice mismatch between the layers. This field together with the conduction band offset between AlGaN and GaN leads to a deep triangular potential well for the electrons. The probability densities of the three lowest electron subbands [9] in this well are shown in Fig. 1(b). To achieve a good coupling of the incident beam to the intersubband dipole (polarized $\| z$) the sample is prism-shaped [Fig. 1(a)], which leads to a double pass of the optical beam through the well region. IS transitions of electrons from the $n = 1$ to the $n = 2$ subband lead to the

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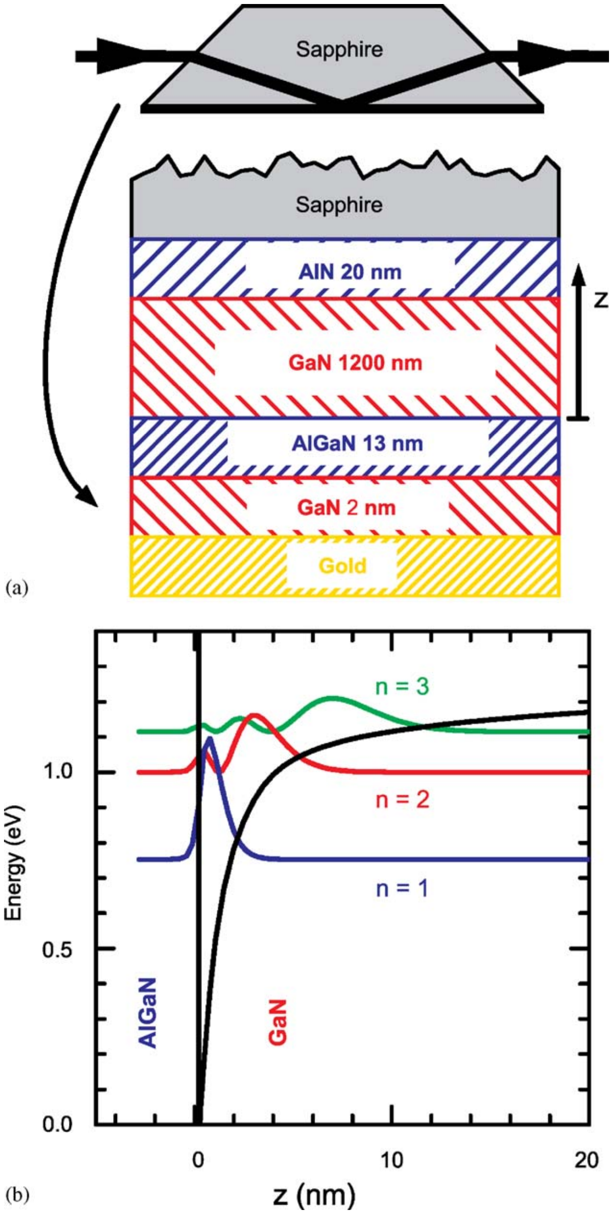

Fig. 1. (a) Cross section through the prism-shaped sample showing the sequence of layers. (b) Self-consistent calculation of the triangular potential well and the probability densities of the three lowest electron subbands $n = 1$, 2 and 3 near the GaN/$\mathrm{Al_{0.8}Ga_{0.2}N}$ interface.

broad IS absorption spectrum (symbols) shown in Fig. 2(a).

For our time-resolved experiments, independently tunable pump and probe pulses are generated at 1-kHz repetition rate by difference-frequency generation in 1-mm (for the pump) and in 0.5-mm (for the probe) thick GaSe crystals [10] between signal and idler of two synchronized optical parametric amplifiers. The pulse lengths of the bandwidth-limited pump and probe pulses are 100 and 50 fs, respectively. After interaction with the sample, the probe pulses are spectrally dispersed in a monochromator (3 meV resolution) and detected by a liquid-nitrogen-cooled InSb detector.

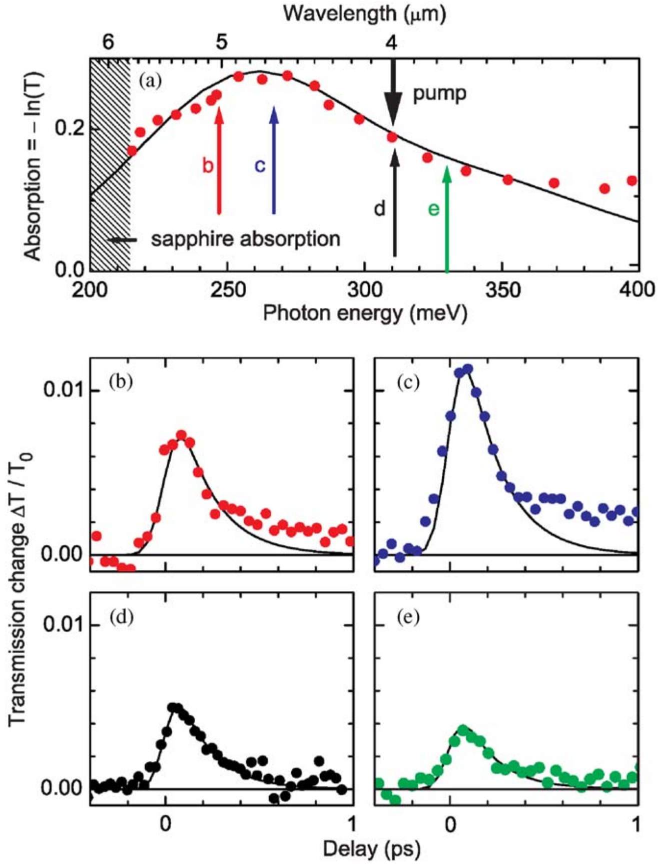

Fig. 2. (a) Measured (symbols) and calculated (solid line) absorption spectrum at a temperature of 300 K. The broad absorption band is due to transitions of electrons from the $n = 1$ to the $n = 2$ subband in the triangular potential well shown in Fig. 1(b). (b)–(e) Time transients from two-color mid-infrared pump-probe experiments for several probe energies [see arrows in (a)] and a pump energy of 310 meV.

3. Results

Transients from an extensive series of time-resolved experiments with pump and probe pulses at different frequencies within the IS absorption band are shown in Fig. 2(b)–(e). The transmission change $\Delta T/T_0 = (T - T_0)/T_0$ is plotted as a function of the delay between pump and probe (T, T_0: transmission with and without excitation, respectively). For all spectral positions of pump and probe the signal (circles) rises within our time resolution of 50 fs. Afterwards it shows a biexponential decay with a fast decay time of 80 fs and a slow one of 380 fs. The amplitude ratio of these two components depends on the spectral position.

Transient IS spectra obtained with a photon energy of the pump of 355 meV are shown in Fig. 3. Around time delay zero we observe two pronounced peaks of nonlinear transmission (spectral holes) separated by the energy of the GaN longitudinal optical phonon $\hbar\omega_{\mathrm{LO}} = 92\,\mathrm{meV}$. After 100 fs, these spectral features are nearly washed out. For delays larger than 200 fs, the transient spectrum has a

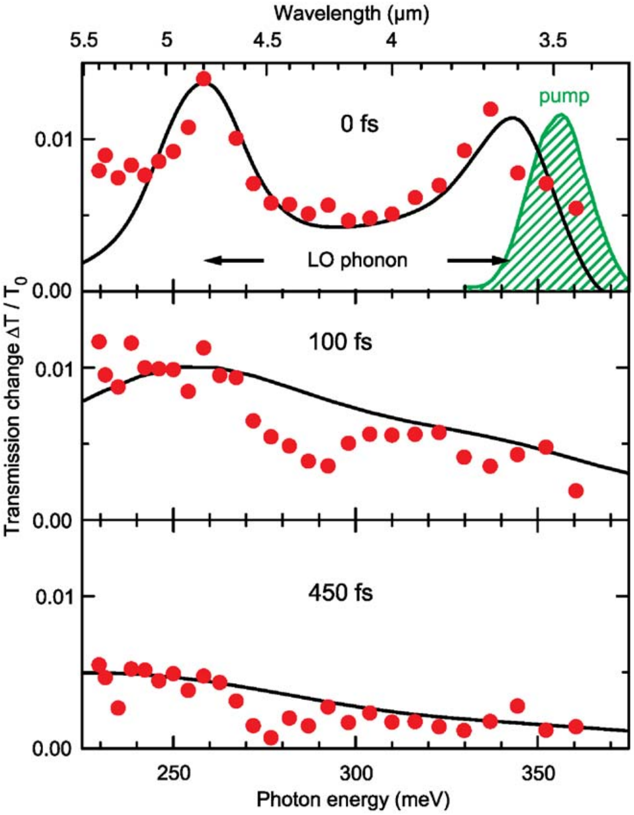

Fig. 3. Measured (symbols) [11] and calculated (solid line) transient spectra from two-color pump-probe experiments for different delays between pump and probe. In the upper panel the spectrum of the 100-fs pump pulse is shown for comparison.

constant shape with an amplitude decaying with a decay time of 380 fs [11].

4. Discussion

From the instantaneous transmission increase for all spectral positions (see Fig. 2) we conclude that homogeneous broadening is an important contribution to the total IS linewidth. If there would be only homogeneous broadening, the transient spectra would have the same spectral shape as the absorption. Since this is obviously not the case (see Fig. 3), especially not at short delays, inhomogeneous broadening is also present.

For the theoretical description of our results, it does not make sense to consider electrons and phonons separately because of the strong coupling of electrons to LO phonons in these materials (the Frohlich coupling constant $\alpha = 0.5$ [12] is equivalent to an average electron–phonon scattering time of 5 fs). Already in the excitation process one does not excite only electrons, but rather quasiparticles (IS polarons), consisting of strongly coupled electrons and LO phonons. These new quasiparticles result in the phonon sidebands observed around time delay zero (Fig. 3). Without any coupling to other elementary excitations, the spectra at later times would be identical to the one at 0 fs. The coupling to other elementary excitations is the cause of the decay of the signal at later times.

An appropriate model for such phenomena is the independent boson model (IBM), in most cases up to now applied to molecular systems [13–18]. Within this phenomenological model both linear and nonlinear optical properties of systems with strong electron–phonon coupling can be calculated. The IBM describes the relevant elementary excitations as a thermal bath of harmonic oscillators coupling linearly but with arbitrary strength to the electronic system under consideration. In our case, we use an electronic two-level system coupled strongly to the LO phonon, causing the experimentally observed sidebands, and to a continuum of oscillators, representing all other scattering mechanisms, e.g., carrier–carrier scattering and disorder scattering. This continuum of oscillators is equivalent to one strongly overdamped oscillator, the so-called Brownian oscillator [17].

This model accounts for both the linear absorption spectrum and the full set of nonlinear pump-probe data. The calculated spectra are in good agreement with all our experimental results (see, e.g., the solid lines in Figs. 2 and 3). In particular, the position, width and amplitude of the spectral holes around time delay zero are well reproduced by the model.

The fast decay constant observed (80 fs) corresponds in the IBM to ultrafast spectral diffusion, which quickly washes out the spectral features around time delay zero. The slow decay constant (380 fs) is determined by the electron lifetime in the excited state.

5. Conclusions

In conclusion, we studied the ultrafast IS dynamics of electrons in a GaN/AlGaN heterostructure by mid-infrared two-color pump-probe measurements. We observe phonon sidebands of the IS transition as spectral holes separated by the LO phonon frequency. These sidebands decay after excitation with a decay time of 80 fs. Our results show that a large part of the broad linewidth of intersubband transitions in this material system is caused by intrinsic effects.

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