

Ultrafast hole burning in intersubband absorption lines of GaN/AlN superlattices

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The authors present evidence for a distinct optical phonon progression in the nonlinear intersubband absorption spectra of electrons in a GaN/AlN superlattice. Femtosecond two-color pump-probe experiments in the near infrared show spectral holes separated by the longitudinal optical (LO) phonon frequency and a homogeneous line broadening of approximately 50 meV. The nonlinear bleaching signal decays with a time constant of 160 fs due to intersubband scattering of delocalized electrons, followed by a weak picosecond component attributed to the relaxation of electrons from longer-lived localized states.

Optical intersubband (IS) excitations in semiconductor nanostructures provide direct insight into the ultrafast dynamics of coherent optical polarizations and nonequilibrium carriers and play a key role for devices such as quantum cascade lasers.^{1,2} Because of the very large conduction band discontinuity, nanostructures made of group-III nitrides have recently attracted considerable interest. In particular, intersubband absorption of quasi-two-dimensional electrons in GaN/AlGaIn multiple quantum wells (MQWs) has been observed at the communication wavelength $\lambda \approx 1.55 \mu\text{m}$ (photon energy of 800 meV).^{3,4} First time-resolved experiments on MQWs have obtained intersubband relaxation times between 150 and 300 fs.^{5,6} More recent femtosecond two-color pump-probe studies on similar structures have given IS relaxation times of 140 fs and a (sub)picosecond intraband redistribution of carriers by thermalization and energy transfer processes of the two-dimensional (2D) electron gas.⁷

So far, the line shape of IS absorption in GaN/AlGaIn nanostructures is not fully understood. In addition to inhomogeneous broadening due to structural imperfections, one expects a substantial homogeneous broadening caused by the ultrafast decoherence of the IS polarization. Moreover, the strong coupling between electrons and longitudinal optical (LO) phonons in group-III nitrides may give rise to spectrally distinct sidebands in electronic spectra.^{8,9} Very recently, a pronounced optical phonon progression was found in the nonlinear IS absorption of a GaN/AlGaIn heterostructure.¹⁰

In this letter we show that the IS transitions of electrons in GaN/AlN superlattices display a considerable homogeneous broadening of the order of 50 meV and couple strongly to LO phonons as observed through distinct sidebands in the nonlinear optical response. Transient IS spectra recorded in femtosecond two-color pump-probe experiments exhibit spectral holes on different vibronic transitions spec-

trally separated by the LO phonon frequency. More than 95% of the nonlinear decrease of IS absorption decay with $\tau = 160$ fs due to IS scattering of delocalized $n=2$ electrons, whereas a small residual bleaching decaying within several picoseconds may be attributed to excited electrons in localized states.

We investigated a GaN/AlN superlattice consisting of 20 periods of 2.2 nm thick GaN potential wells doped with Si and 2.2 nm thick AlN barriers (for details of the sample see Ref. 11). The coupling to the IS dipole moment is maximized by using p -polarized pulses and by the prism geometry of the sample [cf. Fig. 1(a) of Ref. 10]. The linear IS absorption at room temperature is plotted in Fig. 2 (solid line). This absorption band has a full width at half maximum of $\Delta E_{\text{IS}} = 140$ meV and is due to transitions of electrons from the $n=1$ to the $n=2$ subband. In our time-resolved experiments, electrons are excited from the $n=1$ to the $n=2$ subband by bandwidth-limited 100 fs pump pulses (pump intensity of about 100 MW/cm²), having different center frequencies within the IS absorption band. The nonlinear response is probed by independently tunable 50 fs pulses of much lower intensity. Pump and probe pulses at a 1 kHz repetition rate are derived from the output of two synchronized optical parametric amplifiers.¹² After interaction with the sample, the probe pulses are spectrally dispersed in a monochromator (3 meV resolution) and detected by a cooled InSb detector.

We performed an extensive series of time-resolved experiments with pump and probe pulses at different frequencies within the IS absorption band.

In Fig. 1, the time evolution of the spectrally integrated transmission changes $\Delta T/T_0 = (T - T_0)/T_0$ (closed symbols) is presented for pump photon energies of $E_{\text{ex}} = 660$ meV and $E_{\text{ex}} = 755$ meV (T, T_0 : sample transmission with and without excitation). In both cases the overall bleaching displays a fast decay with a time constant of 160 fs (solid lines). In contrast to the simple shape of the spectrally integrated transients, transients taken for specific probe energies (open symbols) show a more complicated shape around time delay zero. For

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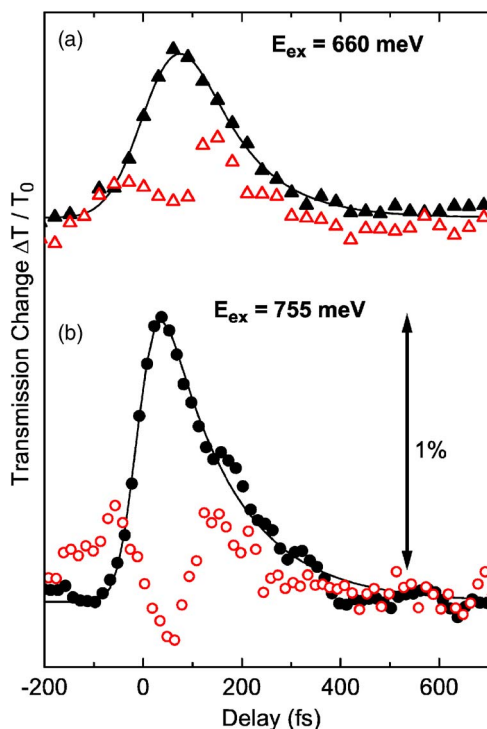


FIG. 1. (Color online) Time dependent change of transmission measured with pump pulses centered at (a) $E_{\text{ex}}=660$ meV and at (b) $E_{\text{ex}}=755$ meV. The closed symbols show the transmission change spectrally integrated over all probe photon energies, while the open symbols for a probe photon energy of (a) 756 meV and (b) 747 meV. The solid lines are single-exponential fits to the spectrally integrated data.

delays larger than about 150 fs they again follow the time-integrated data.

Transient spectra are presented in Fig. 2 for the same pump photon energies as in Fig. 1; the transmission change is plotted (symbols) as a function of the probe frequency for different pump-probe delays. For both pump photon energies, we observe in the nonlinear transmission for early delays two pronounced peaks, i.e., spectral holes. Such holes occur at the spectral position of the pump pulse and at a second position separated by the energy of the GaN longitudinal optical phonon of 92 meV. These spectral features wash out on a time scale of about 300 fs. A weak spectral hole around 650 meV remains for longer delays [Fig. 2(a)], decaying on a time scale of a few picoseconds. The spectral holes observed at early delay times are substantially broader than the spectrum of the pump pulses [compare the dots and the open triangles with the dashed line in Fig. 2(a)].

In our experiments, the pump pulses promote electrons from the $n=1$ to the $n=2$ subband, resulting in a bleaching of IS absorption. The spectrally integrated transmission changes (Fig. 1) yield a lifetime of electrons in the $n=2$ subband of 160 fs. This value is in good agreement both with previous experimental results^{5,7} and with theoretical calculations of the IS scattering rate of delocalized 2D electrons via the emission of LO phonons.

Our experimental situation is very similar to that of spectral hole burning experiments in large molecules,¹³ which have been theoretically analyzed in Ref. 14. Even in the case of a structureless, predominantly homogeneously broadened, linear absorption band, the theory predicts during the overlap of pump and probe sharp spectral holes on various vibronic transitions of the system. The holes are caused

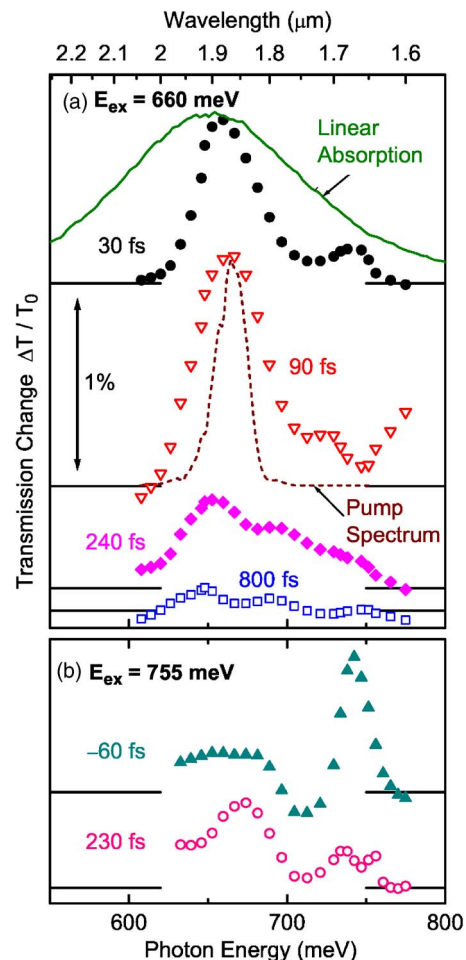


FIG. 2. (Color online) Transient spectra measured in two-color pump-probe experiments (symbols) with pump pulses centered at (a) $E_{\text{ex}}=660$ meV and at (b) $E_{\text{ex}}=755$ meV. The transmission change $\Delta T/T_0=(T-T_0)/T_0$ is plotted as a function of probe frequency for different pump-probe delays (T , T_0 : transmission with and without excitation). Solid line, linear IS absorption spectrum; dashed line, pump spectrum.

by coherent coupling between pump and probe. Their spectral width is determined essentially by the pump pulse. In particular, the spectrally resolved transients (open symbols in Fig. 1) around zero delay are strongly influenced by such phenomena,¹⁵⁻¹⁷ even leading at certain delays to negative signals. Only for strictly sequential pump-probe interaction, i.e., for delays larger than about 150 fs, is it possible to use the classical interpretation in terms of population holes.

The transient spectra of Fig. 2 display spectral holes separated by the LO phonon energy. This behavior is a direct consequence of the strong electron-LO phonon coupling (Fröhlich coupling constant $\alpha=0.5$) in GaN, resulting in a sequence of vibronic transitions between coupled electron-phonon states in the $n=1$ and $n=2$ subbands, and an optical phonon progression in the IS absorption. IS excitation results in a bleaching on the progression line overlapping with the pump spectrum and on neighboring lines sharing the initial or final state with the excited transition. This mechanism represents the origin of the different spectral holes in the nonlinear spectra of Fig. 2. A similar behavior has been observed in a recent study of nonlinear IS absorption in a GaN/AlGaIn heterostructure.¹⁰ The already mentioned effect of pump-probe coupling is the reason that the phonon sideband in Fig. 2(a) can be clearly observed for a delay of 30 fs,

but is not very pronounced for a delay of 90 fs.

In Fig. 2, the spectral hole measured for early delays [30 fs (dots) and 90 fs (open triangles)] has a width of 55 meV (full width at half maximum), substantially larger than that of the pulse spectrum (dashed line). Such behavior occurs for a homogeneous broadening of IS absorption exceeding the spectral pulse width. The width of the spectral hole corresponds to the homogeneous broadening contribution to the total linewidth of $\Delta E_{IS} = 140$ meV, the latter containing additional inhomogeneous broadening contributions, e.g., by structural disorder in the superlattice. A homogeneous width of 55 meV corresponds to a dephasing time of approximately 25 fs. Both electron-electron and intraband electron-LO phonon scattering contribute to this ultrafast dephasing on the IS transition. It should be noted that the dephasing times found here are substantially shorter than those measured for IS transitions in the GaAs/AlGaAs material system.¹⁸

Our measurements give evidence for a weak bleaching component after IS relaxation of electrons. The squares in Fig. 2(a) show the time-averaged nonlinear spectrum of this signal for delays from 800 to 900 fs. The remaining spectral hole is centered around the maximum of the linear IS absorption, i.e., redshifted by ≈ 20 meV with respect to the pump pulse, and decays within several picoseconds, substantially slower than intraband carrier cooling.^{7,10} We attribute this weak long-lived signal to electrons scattered into localized states with long lifetimes.

In summary, the ultrafast IS dynamics of electrons in a GaN/AlN superlattice was studied in two-color pump-probe measurements in the near infrared. IS transitions of electrons in GaN/AlN superlattices experience a considerable homogeneous broadening of the order of 50 meV and couple strongly to LO phonons as observed through distinct sidebands in the nonlinear optical response. The IS scattering time of $n=2$ electrons has a value of 160 fs. Both the strong homogeneous broadening of the IS transition and the short IS

relaxation times are properties that have to be addressed in the design of optoelectronic devices, e.g., light emitting quantum cascade structures, made from highly polar materials.

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