Terahertz study of ultrafast carrier dynamics in InGaN/GaN multiple quantum wells

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Abstract. Ultrafast carrier dynamics in InGaN/GaN multiple quantum wells is measured by time-resolved terahertz spectroscopy. The built-in piezoelectric field is initially screened by photoexcited polarized carriers, and is gradually restored as the carriers recombine. We observe a nonexponential decay of the carrier density. Time-integrated photoluminescence spectra have shown a complete screening of the built-in piezoelectric field at high excitation fluences. We also observe that the terahertz conductivity spectra differ from simple Drude conductivity, describing the response of free carriers, and are well fitted by the Drude-Smith model.

1. Introduction
The understanding of ultrafast dynamics in group-III nitride based quantum wells is important for the development of wide bandgap semiconductor devices, such as LEDs and lasers in the blue-green spectral region [1]. In such systems the large, strain-induced built-in piezoelectric field has significant influence on the optical and electronic properties of AlGaN/GaN and InGaN/GaN quantum wells, described by the quantum confined Stark effect (QCSE) [2]. The QCSE will manifest itself by a strong dependence of the transition energy, absorption coefficient and recombination rate on the carrier density in the quantum well [3]. Whereas in a previous terahertz investigated, the dynamics of the absorption coefficient has been studied [4], we will concentrate on the carrier density decay dynamics by using time-resolved terahertz spectroscopy.

2. Experiments and Results
Our sample consists of a 10-layer In_{0.2}Ga_{0.8}N/GaN multiple quantum well (MQW) with a well thickness of 1.8 nm and a barrier width of 7 nm. The quantum well structure is sandwiched between a 180 nm thick capping layer and a 2 μm buffer layer. The sample is grown on a sapphire substrate by metalorganic vapor-phase epitaxy. The lattice mismatch between In and Ga causes strain in the material, which results in large built-in piezoelectric fields of 3.1 MV/cm in the quantum wells and 0.5 MV/cm (with opposite polarity) in the barriers [4, 5].

The optical properties of a biased system are well described by the QCSE [2]. The tilt of the bandgap (Fig. 1(a)) results in a smaller transition energy and the reduction of the spatial overlap between the electron and hole wavefunctions leads to a smaller transition probability. During excitation with an ultrashort laser pulse, a dipole moment is created due to the shifted position of the electron and hole wavefunctions. This dipole moment has a polarity opposite to that of the built-in piezoelectric field and thus it will partly screen this field. With strong excitation fluences...
Figure 1. (a) Schematic of the InGaN/GaN QW bandstructure, at weak excitation fluence. (b) Schematic of the QW bandstructure, showing the screening of the piezoelectric field, at strong excitation fluence. $E_i$: initial transition energy, $E_f$: final transition energy after total screening of the piezoelectric field, $F_{\text{dipole}}$: dipole field induced by the electron-hole pairs, $F_{\text{bias}}$: build-in piezoelectric field, $F_{\text{Total}}$: net electric field of the combined dipole and piezoelectric field. (c) Time-integrated PL spectra with excitation fluences in the range 0.03-1.15 mJ/cm².

Complete screening of the built-in piezoelectric field can be achieved (Fig. 1(b)). The screening results in flattening of the bandstructure and hence in an increase of the optical transition energy and transition probability. Since every photoexcited carrier contributes to the screening, this process is highly dynamical. As the carriers recombine the reverse process will take place. Recombination of electron-hole pairs decreases the dipole field and thus causes the restoration of the built-in piezoelectric field. This may imply that during the recombination process the recombination rate will slow down due to increased spatial separation of the wavefunctions.

We use time-resolved THz spectroscopy to measure the decay of the photoexcited carriers. The setup is fed by a 3.5 mJ/pulse, 45 fs duration, Ti:sapphire amplifier with central wavelength around 800 nm. The THz pulses, used as a probe for the carrier density, are generated by optical rectification in a 1 mm thick ZnTe crystal. The temporal shape of the THz pulses are detected after normal incidence transmission through the InGaN/GaN MQWs by free-space electrooptic sampling in a 2 mm thick ZnTe crystal. Part of the Ti:sapphire amplifier output is used to generate 400 nm pump pulses by frequency doubling in a beta-barium borate (BBO) crystal. The 400 nm wavelength assures that with single photon absorption, electron-hole pairs can only be generated in the InGaN quantum wells and not in the GaN barriers. We observed no significant influence of two-photon absorption in GaN on our experimental results.

To show the screening of the built-in piezoelectric field, time-integrated photoluminescence (PL) measurements were performed with excitation fluences in the range of 0.03-1.15 mJ/cm² (Fig. 1(c)). A strong Stark shift is observed between low and high excitation fluences. The saturation of the blue shift for the highest excitation fluences indicates that complete screening of the built-in piezoelectric field takes place. The large shoulder at longer wavelength for the highest excitation fluences is a result of using a time-integrated technique. As carriers recombine the piezoelectric field is restored, which results in a decrease of the transition energy and thus in a red-shift of the PL spectra.

The relative differential transmission of the THz probe between an excited and unexcited sample can be related to the THz conductivity within the measured 0.5-2.5 THz spectral bandwidth. An easier method to monitor changes in THz conductivity than measuring the full THz pulse is to measure only the peak of the THz pulse, for different pump-probe delay
Fig. 2. (a) Photoinduced conductivity dynamics in a 1.8 nm thick InGaN/GaN MQW with pump fluences in the range 0.03-1.15 mJ/cm². (b) Real and imaginary parts of the terahertz conductivity, 5 ps after optical excitation with a fluence of 0.14 mJ/cm². The blue are fits of the data to the Drude-Smith model.

Fig. 2(a) shows the transient photoconductivity in InGaN/GaN MQWs for the same pump fluences as in Fig. 1(c). A nonexponential decay of the photoconductivity is observed, more enhanced for large pump fluences. At early pump-probe delay times and high pump fluences, the complete screening of the built-in piezoelectric field results in a large overlap of the electron and hole wavefunction and hence a large recombination rate. At later pump-probe delay times, the carrier density decreases due to electron-hole recombination, resulting in the restoration of the built-in piezoelectric field, which will decrease the recombination rate, causing a nonexponential carrier density decay. The same nonexponential decay dynamics are observed by time-resolved PL experiments in AlGaN/GaN quantum wells, where the QCSE is also present [6]. The dependence of the decay rate on the carrier density can also be observed by the faster initial decay at higher fluences (Fig. 2(a)).

Relating the induced photoconductivity dynamics directly to the carrier density dynamics implies a constant carrier mobility. Information about the carrier mobility can be obtained by time-resolving the THz pulses for various times after optical excitation and compare them with a reference pulse, transmitted through an unexcited sample. The Fourier transforms of the THz pulses can be related to the photoinduced complex THz conductivity, $\sigma(\omega)$ [8, 9, 10, 11]. The real, $\sigma_1$, and imaginary, $\sigma_2$, parts of the complex conductivity, 5 ps after optical excitation are shown in Fig. 2(b). The simple Drude-model, describing a conductivity response caused by free carriers, observed in bulk n-GaN [10] and n-GaN films [11], fails to explain the observed features here. Deviations from the Drude model in n-GaN films have also been reported [12]. The smaller conductivity for low frequencies and the zero-crossing of the imaginary part of the conductivity at non-zero frequency indicate a non-Drude conductivity response. The observed deviations from Drude conductivity can be described by a classical generalization of the Drude model, known as the Drude-Smith model [13]. THz conductivity spectra of various nanostructures have been successfully described by the Drude-Smith model [8, 9]. This model reads:

$$\sigma(\omega) = \frac{\epsilon_0\omega_p^2\tau}{1 - i\omega\tau} \left[1 + \sum_j \frac{c_j}{(1 - i\omega\tau)^j}\right].$$

(1)
The first term is the Drude term, where $\epsilon_0$ is the permittivity of free space, $\omega_p$ is the plasma frequency and $\tau$ is the scattering time. The second term describes the persistence of the carriers initial velocity after a certain scattering event, where $c_j$ is the persistence of velocity coefficient for scattering event $j$. The blue curves in Fig. 2(b) are fits to the conductivity spectrum with the Drude-Smith model, where only the first scattering event is taken into account ($j=1$). The real and imaginary part of the conductivity are fitted to the same set of parameters simultaneously. From these fits we obtain, $\omega_p/2\pi = 493 \pm 5$ THz, $\tau = 63 \pm 1.4$ fs and $c = -0.595 \pm 0.009$. The conductivity spectra at later times after optical excitation have shown similar values for $\tau$ and $c$ (not shown here). The change in plasma frequency, $\omega_p$, for later times is caused by a change in carrier density. The time and carrier density independent scattering time, $\tau$, and the persistence of velocity coefficient, $c$, implies that the mobility is also independent of the carrier density. This means that the observed nonexponential decay of the photoconductivity (Fig. 2(a)), is completely caused by a nonexponential carrier density decay.

3. Conclusion
A nonexponential carrier density decay is observed in InGaN/GaN MQWs as a result of the QCSE, caused by the restoration of the built-in piezoelectric field as carriers recombine. Time-integrated PL spectra have shown the initial screening of the built-in piezoelectric field. The THz pulses are time-resolved to obtain THz conductivity spectra at various times after optical excitation. Fits to the Drude-Smith model with a non-zero value of $c$ show that the conductivity response differs from that of free carriers.

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