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Homogeneous linewidth of the intraband transition at 1.55 μm in GaN/AlN quantum dots

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We present homogeneous linewidth measurements of the intraband transition at 1.55 μm in GaN/AlN quantum dots by means of non-linear spectral hole-burning experiments. The square-root dependence of the differential transmission signal with the incident pump power reveals the importance of electron-electron scattering in the population relaxation dynamics. We find on the contrary that this scattering process plays a minor role in the coherence relaxation dynamics since the homogeneous linewidth of 15 meV at 5K does not depend on the incident pump power. This suggests the predominance of other dephasing mechanisms such as spectral diffusion, and temperature-dependent measurements support this hypothesis.

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GaN/AlN nanostructures in the form of quantum wells or quantum dots (QDs) are of growing importance in the field of optoelectronics in the optical communication spectral range[1–4], in particular for high repetition-rate all-optical switches[5–7]. The applications rely on intraband transitions that reach exceptionally short wavelengths in the GaN/AlN system compared to arsenide semiconductor materials because of the huge conduction band offset of 1.75 eV between GaN and AlN. Intraband transitions in GaN/AlN QDs covering the spectral range from 2.5 μm to 1.4 μm have been experimentally demonstrated[3,8]. The large ratio between the intraband transition energy (800 meV) and the LO phonon energy in GaN (92 meV) together with the existence of intermediate confined states[9,10] opens many relaxation pathways for the unipolar excitations, and results in the ultrafast relaxation dynamics observed in GaN/AlN nanostructures[11,12]. However, a detailed insight into the microscopic processes governing the relaxation dynamics is still missing although of fundamental importance for the understanding and the optimization of the devices operation. In particular, there is no direct study of the homogeneous broadening which strongly determines the saturation properties of GaN/AlN QDs.

In this paper, we present homogeneous linewidth measurements of the intraband transition at 1.55 μm in GaN/AlN QDs by means of non-linear spectral hole-burning experiments. We observe a square-root dependence of the differential transmission signal with the incident pump power, therefore revealing the importance of electron-electron scattering in the population relaxation dynamics. However, the width of the spectral hole does not depend on the incident pump power. Moreover, the large homogeneous linewidth of 15 meV at 5K together with the significant thermally-assisted broadening suggests the predominance of other dephasing mechanisms such as spectral diffusion.

Our sample contains 200 layers of self-assembled GaN/AlN QDs grown in the Stranski-Krastanow mode by plasma-assisted molecular beam epitaxy on 1 μm thick AlN-on-sapphire (0001) template, as detailed in Ref. [14]. The QD areal density is of the order of 1.2x10^{12}cm^{-2}, and the QDs are doped in the GaN layer with Si at a concentration of 10^{20}cm^{-3}, which corresponds to an average population of 2.5 electrons per dot[12]. High-resolution transmission electron microscopy showed that the appropriate QD morphology for intraband absorption around 1.55 μm corresponds to QD diameter and height (including the wetting layer thickness) of 6±1 and 1.7±0.1 nm, respectively[13]. Linear absorption measurements were performed at room temperature using Fourier transform infrared spectroscopy on a sample polished to form a 45° multipass waveguide. The optical density displayed in dashed line in Fig. 1 is deduced from the transmission of TM-polarized light with four passes in the active region. As discussed in previous studies[8], the absorption line at 1.55 μm is ascribed to the $s - p_z$ intraband transition, and it is inhomogeneously broadened with a full width at half maximum (FWHM) of 128 meV.

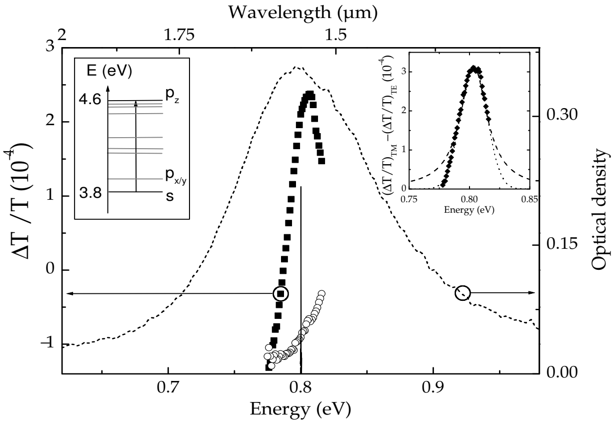

FIG. 1. Optical density (dashed line) of the GaN/AlN QD sample. Differential transmission measurements at 10K versus probe energy with TM- (squares) and TE-polarized (open circles) pump and probe, with a pump of 3.5 kW.cm^{-2} at 0.8 eV. System response function (solid line) of the pump-probe setup. Insets: (left) schematic level diagram with a reference energy at the highest confined valence state; (right) Gaussian (dotted line) and Lorentzian (dashed line) fits of the hole-burning signal.

Homogeneous linewidth measurements are performed by implementing the non-linear spectral hole-burning technique at 1.55 μm. Our experimental scheme is based on differential transmission experiments in a spectrally resolved pump-probe setup in cw. The pump laser is a semiconductor distributed feed-back laser seeding an erbium-doped fiber amplifier. Its output is coupled to a monomode fiber providing up to 1W at 1.55 μm on a single transverse mode. The probe laser is an external-cavity semiconductor laser tunable from 1.52 μm to 1.6 μm. The pump-induced variation ΔT of the probe transmission T is detected by a lock-in amplifier at the frequency $f_1 + f_2$, where f_1 and f_2 are the modulation frequencies of the pump and probe laser intensity, respectively. The orthogonally-propagating laser beams are both incident at 45° on the sample mounted on the cold finger of a continuous-flow cryostat for measurements down to 5K.

In Fig. 1, we display the differential transmission measured at 10K for a pump power of 3.5 kW.cm^{-2}. For TE-polarized pump and probe (open circles), the non-linear signal varies smoothly with negative values corresponding to a background signal of photo-absorption. For TM-polarized pump and probe (squares), the differential transmission of the probe exhibits a pronounced bleaching with an amplitude of 3.10^{-4}. According to the selection rules of the $s-p_z$ intraband transition in GaN/AlN QDs[3], we attribute the photo-induced increase of the probe transmission to the partial saturation of the QD intraband absorption by the pump laser. Due to the limited tuning range of our probe laser, the spectral hole burned by the pump laser is not fully resolved on the high energy side. However we can estimate the FWHM of the bleaching line Γ_{HB} and we obtain a value of 27 meV which is much smaller than the 128 meV-inhomogeneous linewidth measured in linear absorption (dashed line in Fig. 1) and much larger than the width of the response function in the μeV range (solid line in Fig. 1) recorded for the coherent artefact on an InP substrate.

In the reference case of absorption saturation in an ensemble of two-level systems[15], the relationship between the homogeneous linewidth Γ and the hole-burning one Γ_{HB} is given by Γ_{HB}=Γ+Γ_s with Γ_s=$\Gamma\sqrt{1+I/I_s}$ where I and I_s are the pump and saturation intensities, respectively. In the weak saturation regime ($I \ll I_s$), $\Gamma_{HB} \sim 2\Gamma$, but we observe in our measurements two distinct features that differentiate our sample from the text-book example of an ensemble of two-level systems so that Γ may deviate from $\Gamma_{HB}/2$.

First of all, when fitting the hole-burning signal by substracting the data recorded with TM- and TE-polarizations (symbols, Fig. 1 inset), we observe a better agreement with a Gaussian profile (dotted line, Fig. 1 inset) than with a Lorentzian one (dashed line, Fig. 1 inset). Even if a definite shape-analysis remains difficult in our case because of the limited spectral range of investigation, the ultra-broad spectral hole is indeed consistent with a Gaussian absorption profile. Dephasing processes generally lead either to Gaussian or Lorentzian lineshapes depending on the product between the energy fluctuation amplitude Σ and the characteristic correlation time of fluctuation τ_c[16–18]. For spectral diffusion[19], phonon scattering[20] or Brownian oscillator scattering[21], the correlation time τ_c has typical values ranging from 0.1 to 10 ps in semiconductor nanostructures. Consequently, at the crossover from Lorentzian to Gaussian profiles, the homogeneous linewidth given by $\hbar/\tau_c$ is expected to take characteristic values between 50 μeV and 5 meV. In our case, the spectral hole of 27 meV at 10K is consistent with a Gaussian lineshape.

The second deviation from an ensemble of two-level systems consists in the sub-linear increase of the differential transmission signal with the incident fluence. As shown in Fig. 2(a), we observe that the non-linear signal scales like the square-root of the pump power whereas two-level systems give a linear dependence in the limit of weak excitation ($I \ll I_s$). This behavior stems from the existence of a population relaxation process with an efficiency proportional to the square of the electron population n^2 in the excited state, such as electron-electron scattering. If this non-linear relaxation process dominates the population relaxation dynamics, the stationary value of n scales like the square root of the pump power. Consequently, we attribute the sublinear dependence of our differential transmission signal to electron-electron scattering, and we conclude that the population relaxation from the p_z excited state is governed by Auger-type processes. As a matter of fact, our study reveals that optical phonon emission only plays a minor role, even if it may contribute to secondary processes during the carrier cascade to the fundamental s state via intermediate confined states[8–10] (Fig. 2(a), inset).

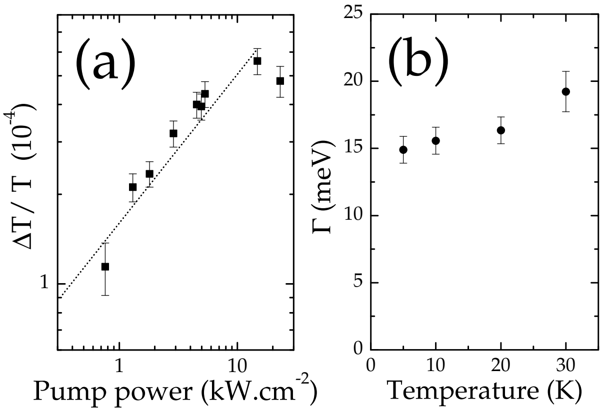

FIG. 2. (a) Amplitude of the differential transmission signal for a probe energy of 0.8 eV versus incident pump power, at 5K. The dotted line is a guide for the eye corresponding to a $P^{1/2}$ function. (b) Homogeneous linewidth of the GaN/AlN QD intraband transition versus temperature.

By taking into account the Gaussian absorption profile and the sublinear dependence of the non-linear signal with the pump power, we are able to revisit the simple relation $\Gamma_{HB} \sim 2\Gamma$

found for two-level systems. Following the approach from Ref. [15] based on a rate equations analysis of the pump-induced population changes in an inhomogeneously broadened line, we find that the spectral hole has a linewidth given by $\Gamma_{HB}^2 = 2\tilde{\Gamma}_s^2 + \Gamma^2$ where $\tilde{\Gamma}_s$ takes into account the modification of the homogeneous profile by the pump. The sum of the squared linewidths comes from the convolution of Gaussian absorption lines instead of Lorentzian ones[15], and the factor 2 for $\tilde{\Gamma}_s^2$ from the square-root dependence of the population variation as a function of pump power. Since the differential transmission signal is of the order of 10^{-4}, we assume that $\tilde{\Gamma}_s \sim \Gamma$ so that the homogeneous linewidth is finally given by $\Gamma \sim \Gamma_{HB}/\sqrt{3}$, corresponding to a value of 16 meV at 10K. The comparison with time-resolved pump-probe experiments at room temperature on the same system[12] suggests that the coherence and population relaxations are not governed by the same processes since Γ is much larger than $\hbar/T_1$, which was estimated to 4 meV. We also conclude that the estimation of Γ from emission measurements in a QD ensemble[13] is incorrect, probably because of the Raman-type configuration that makes any estimation speculative.

We have further studied the dependence of the homogeneous linewidth with various parameters. We do not observe any dependence of Γ with the pump power. In agreement with our interpretation above, we conclude that the coherence relaxation is not dominated by electron-electron scattering in opposition to the population relaxation. As far as the temperature is concerned, our measurements (Fig. 2(b)) show a thermally-assisted broadening. Previous studies on intraband transitions in InAs QDs attribute this effect to acoustic phonon dephasing[22]. However, in the present case, the thermal-broadening of 4 meV between 5 and 30K is by two orders of magnitude larger than in InAs QDs, and thus poorly consistent with this mechanism. Moreover, the large value of 15 meV measured at 5K appears close to the zero-temperature limit, so that the understanding of the temperature-dependent measurements certainly requires a distinct dephasing mechanism. Indeed, spectral diffusion may bring a major contribution to the coherence relaxation and also explain the thermally-activated broadening[23]. Further studies as a function of doping concentration should help to further understand and control the homogeneous broadening. This point appears as an important issue in the prospect of optimizing the saturation properties of GaN/AlN QD-based devices.

In summary, we have reported on homogeneous linewidth measurements of the intraband transition at 1.55 μm in GaN/AlN QDs by means of non-linear spectral hole-burning experiments. The square-root dependence of the differential transmission signal with the incident pump power reveals the importance of electron-electron scattering in the population relaxation dynamics. The homogeneous linewidth of 15 meV at 5K does not depend on the incident pump power, suggesting the predominance of other dephasing mechanisms such as spectral diffusion.

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