

## Observation of lattice thermal waves interference by photoluminescence blinking of InGaN quantum well devices

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(Received 14 September 2012; accepted 23 September 2013; published online 24 October 2013)

The photoluminescence of III-V wide band-gap semiconductors InGaN is characterized by local intensity fluctuations, known as “blinking points”, which despite decades of research are not yet completely understood. In this study, we report the experimental data and a theoretical interpretation that suggests that they are caused by the interference of thermal vibrations of the quantum well lattice. With far-field optical tests we could observe the lower frequency tail of these interference waves and study their dynamics as they propagate up to distances of several tens of microns. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4826088>]

The phenomenon called “photoluminescence (PL) blinking” has been observed in confined structures, for example, in semiconductors nanocrystals.<sup>1</sup> Because of the local nature of these systems, the theoretical treatment in literature is zero-dimensional (considering time-energy dependence in one point). The blinking process appears to be associated with the temporary quenching of photoluminescence due to highly efficient non-radiative recombination processes, such as, for example, the Auger effect or other non-radiative processes.<sup>2</sup> Nevertheless, in InGaN devices the band structure is an infinitely wide quantum well (QW), so a zero-dimensional description is insufficient and the understanding of the blinking phenomenon remains elusive.<sup>3–6</sup>

In this letter, we present experimental data and a wave model interpretation that suggests that the discussed optical fluctuations are a general phenomena caused by the interference of thermal vibrational waves that spread along the quantum well. We want to suggest that the spatial vibrations in the lattice alter the overlap integral of matrix elements responsible for photo-luminescence, modifying the recombination rate and promoting the PL blinking. This idea is difficult to grasp because thermally induced vibrations are supposed to be incoherent and of very high frequency, whereas PL blinking is observable as a slow phenomenon that shows some sort of regularity.

However, in confined regions, atomic composition and mechanical properties are homogeneous, giving rise to THz range vibrations with defined frequency and local coherence. In the presence of dislocations or impurities, the mechanical properties of adjacent domains differ, and confined vibrational interferences are generated. These interferences, hereafter called *beats*, have a broad frequency distribution centered around the frequency differential of the interacting oscillations. The lower tail of this interference pattern is what is observable as photoluminescence blinking.

We were able to confirm the thermal nature of the phenomenon with low temperature investigations. Moreover, using a simple far field correlation methodology we could

observe the propagation and spread of the PL *beats* along the quantum well area up to distances of tens of microns.

In a quantum well structured semiconductor as  $\text{In}_x\text{Ga}_{(1-x)}\text{N}$ , the photoluminescence intensity is regulated by the matrix element that mediate the electron-density and hole-density wave function in the vicinity of the well.<sup>7,8</sup> The spatial overlap of these two functions is proportional to the recombination rate, and the latter results in an increase or a reduction of photoluminescence. The difference of inter-atomic distances (lattice mismatch) between the doped and non-doped regions causes piezo-electric stresses that distort the energy bands. This results in diminished wave function overlap and lower recombination rates due to quantum confined stark effect (QCSE).<sup>9,10</sup>

Here, the Wurtzite InGaN quantum well is treated as a two-dimensional system subject to a stress along the direction  $xz$ ; because of spontaneous thermal vibration,<sup>11</sup> this stress is subject to very small oscillations that alter the energy band structure (see the sketch in Fig. 1).

The tension applied on the line of adjacent atoms is indicated by  $T_o$ ,  $x$  is a direction in the plane of the quantum well and  $z$  is the crystal growth vertical axis. We are interested only in displacements along the  $z$  axis, i.e., those that influence the matrix elements relative to the recombination rate. We derive the expression for the forces projected over the  $z$  axis

$$\rho dx \ddot{Z} = T_o \sin(\alpha) - T_o \sin(\beta), \quad (1)$$

where  $dx$  is a small displacement along the well,  $Z$  is the vertical displacement about the rest position, and  $\rho$  is a parameter proportional to the local linear mass density. This approach leads to the general differential expression of a wave oscillating along the crystal growth axis

$$\frac{d^2 Z(x, t)}{dt^2} = \frac{T_o}{\rho} \frac{d^2 Z(x, t)}{dx^2} - \frac{\phi}{\rho} \frac{dZ(x, t)}{dt} - \frac{\gamma}{\rho} Z(x, t). \quad (2)$$

For generality friction ( $\phi$ ) and restoring ( $\gamma$ ) parameters are also included. In this classical approximation, the influence of far away atoms is dumped by friction, hence the local solution is a wave of the form  $\sin(\omega t - kx)$ , where  $\omega$  is related to  $T_o$  and  $\rho$  by  $\frac{T_o}{\rho} \approx (\frac{\omega}{k})$ .

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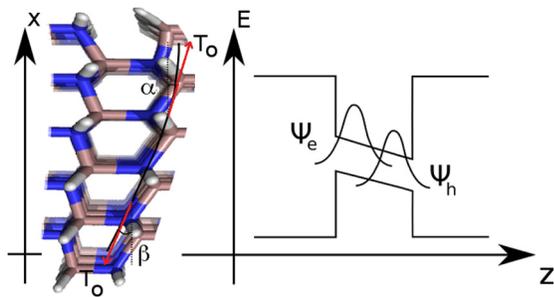


FIG. 1. An exaggerated scheme in which the Wurtzite structure is stretched along the crystal growth direction  $Z$  and the corresponding energy density diagram. The mechanical stress along adjacent atoms is represented in two arbitrary points by the tension vector  $T_0$ , shown in red. The points are tangent to the deformation with angles  $\alpha$  and  $\beta$ . The deformation modifies the overlap of electron-hole wave functions  $\psi_e$ ,  $\psi_h$  and this results in observable photoluminescence variations. The  $x$  represents one of the spatial coordinates.

Elastic mechanical oscillations can interact quasi-coherently in the confined scales; with the term *quasi-coherent* we mean that the waves have phase relation in confined ranges, but this relation is lost over longer distances.

Observed optical intensities are fluctuating of about 10%–20% around a mean value. Experiments on  $\text{In}_x\text{Ga}_{(1-x)}\text{N}$  devices show that the quantum well thickness dependence on the intrinsic radiative lifetime is about 1 order of magnitude per nanometer.<sup>12,13</sup> Supposing that the PL fluctuations are solely due to this phenomenon, then the greatest spatial displacement along the growth direction is in the range of 0.1 nm, a value compatible with the GaN crystal structure.

Since the phenomenon is driven by spontaneous forces, wavelengths are in the order of  $1000\text{ cm}^{-1}$  (about 10THz) as shown in Raman studies and related literature.<sup>14–17</sup> It may not seem possible that the observed slow optical oscillations are induced by frequencies of such high value. Nevertheless, we have to consider that vibrations are localized in a real, non-ideal, crystal system with different Indium concentrations, dislocations, and impurities that alter the overall vibrational modes. In a classic interpretation, this implies that the frequency of oscillation  $\omega$  is in actual fact a spectrum of distributed values that depends on coordinates on the crystal plane. The mechanical interference of different distributed frequencies results in a wide band of lower frequencies beatings centred around  $\delta\omega = \omega_1 - \omega_2$ . In this,  $\omega_1$  and  $\omega_2$  are center frequencies of the vibration spectrum relative to two adjacent domains. Since mass distribution changes are subtle,  $\delta\omega$  have extremely small tails when compared with the original pulsation  $\omega$ . Higher frequency oscillations are cut off by the sampling rate, that is in the order of 30 ms in our case. Previous Fourier measurements<sup>18</sup> demonstrated blinking rates centred on low values of about 5 Hz in nano-scaled blinking domains.

If the observed optical intensity fluctuations are caused by elastic oscillation due to thermal energy, we should observe the characteristic Boltzmann temperature profile of an elastic lattice in spontaneous vibration.<sup>19,20</sup> To test this, we setup an experiment in which the blinking is observed and characterized in controlled temperature conditions.

First of all, we derive the appropriate Boltzmann distribution profile using InGaN elastic potential, then we plot its temperature dependence and test it with our experimental photo-luminescence data.

TABLE I. Table of the predicted thermal behaviour of the lattice's vibrational states at different temperatures according to Boltzmann statistic. The higher and lower probability values  $n_{mx}$ ,  $n_{mn}$  represent a band of  $\pm 2\%$  in temperature,  $\delta$  is the spread. This profile is compared with photoluminescence tests where the increase in intensity and the spread of luminosity at higher temperatures are confirmed. The elastic potential is calculated using GaN Young modulus  $Y = 350\text{ GPa}$ , for simplicity  $S = 1\text{ nm}^2$ ,  $L_0 = 1\text{ nm}$ , the displacement from the rest position is set to  $x = 0.01\text{ nm}$ .

T (K)	$n_{mn}$ (a.u.)	$n_{mx}$ (a.u.)	$\delta$ (a.u.)
200	1.99	1.54	0.44
230	4.49	3.6	0.88
250	6.92	5.65	1.27
270	10.0	8.29	1.71
280	11.7	9.83	1.95
290	13.7	11.5	2.20

In a solid, we use as potential energy the expression  $P.E. = -\frac{1}{2}\frac{YS}{L_0}x^2$ , where  $Y$  is the Young modulus,  $S$  the surface involved in the oscillation,  $L_0$  its rest position, and  $x$  the displacement from it. Assuming  $n = n_0e^{-P.E./kT}$ , where  $n$  is the probability density per unity of area, fixing a displacement one tenth of the maximum discussed above,  $x = 0.01\text{ nm}$ , and using InGaN Young modulus of 350 GPa,<sup>21</sup> we can derive the theoretical density dependence in the range of temperatures used in cryostat experimental tests (Table I).

The theoretical behaviour shown in Table I was confirmed with very straightforward tests. A fluorescence microscope (BX51WX, Olympus) was coupled with a vacuum cryostat chamber (RC102-CFM, CIA, Inc.) to observe the blinking at different temperatures ranging from 290 down to 200 K. At lower temperatures not only the average luminosity slightly reduces but also the variations become less prominent (less blinking) accordingly to the thermal model. Figure 2 compare raw intensity values for a blinking and not blinking point at room temperature, Figure 3 shows data for a single point at several temperatures and in Figure 4 the corresponding luminosity distribution histograms are plotted. The predicted spread and reduced luminosity of the blinking are evident.

We were able to directly detect the horizontal propagation of these interference waves alongside the quantum well. A CCD camera was mounted on an Olympus BX51-W1 microscope to record the photoluminescence on a wide area of an InGaN crystal. The sample is excited with 365 nm light

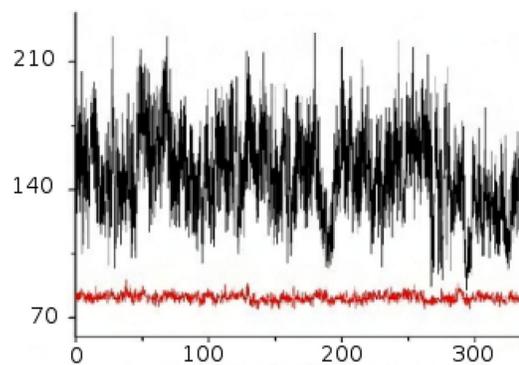


FIG. 2. An example time intensity profile for a blinking point (upper) and a not blinking one, at room temperature. Not blinking points do not show thermal dependence. The luminosity is integrated over an area of about  $0.25\text{ }\mu\text{m}^2$ , time resolution about 30 ms, about 10 seconds of data are shown.

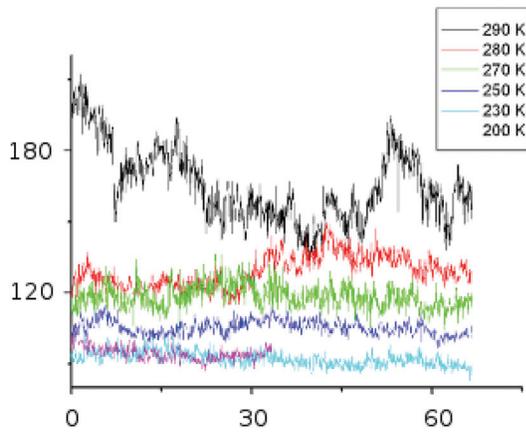


FIG. 3. The behaviour of a single blinking point at the six different temperatures indicated. The blinking becomes less prominent at lower temperatures, with a trend compatible with the thermal vibration beating wave hypothesis. The optical data are acquired from the same blinking point at an interval of  $\tau = 30$  ms for a total of about 70 seconds. Variation of luminosity over 50% is observed at higher temperatures, against less than 10% at lower values. Non-blinking regions of the sample do not show such a typical thermal dependence. This is excluding that the behaviour is a generalized thermal noise phenomena.

from a filtered Xenon light integrated in the microscope. The sample was grown on a sapphire substrate (0002) of about half a centimetre area composed by a  $1.5 \mu\text{m}$  layer of undoped GaN, over which a 3 nm thick single quantum well (SQW) active layer of Indium doped GaN and a final 5 nm capping layer of GaN is deposited. The precise Indium

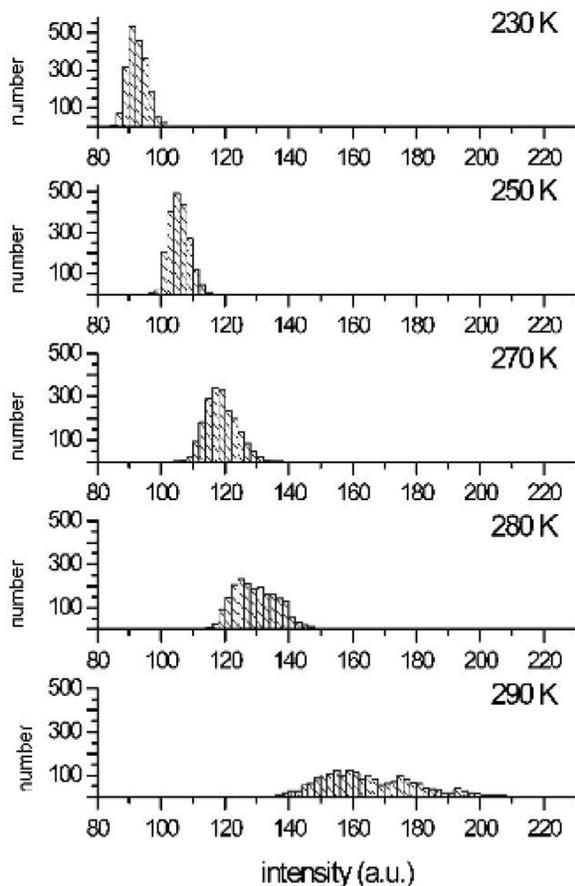


FIG. 4. Using each temperature of the data in Fig. 3, five histograms are plotted. Increased intensity and the spread of luminosity at higher temperatures are confirmed in accordance with a vibrational Boltzmann statistic.

concentration of the QW cannot be measured directly<sup>16</sup> but accordingly to the formula  $\text{In}_x\text{Ga}_{(1-x)}\text{N}$ , we estimate  $x \approx 0.2$ . First, we chose a dominant blinking *target* point in the recorded area, then we analysed the time-correlation of every other pixel to this point that we call  $x_o$ . The analysis was done in a time span of 120 s. In Figure 5, we show the correlation coefficient map referred to a blinking point located at  $x_o = (10,30)$ . Correlation  $r$  is calculated as

$$r = \frac{\text{cov}(x_o(t) \times x_i(t + \text{lag}))}{\sigma(x_o(t)) \times \sigma(x_i(t + \text{lag}))},$$

where  $x_o$  and  $x_i$  are the intensities at central *target* point  $o$  and generic point  $i$ ,  $\text{cov}$  represents the covariance function, and  $\sigma$  the standard deviation one. The map in panel (a) shows that regions adjacent to the target have an optical dynamics that is correlated to that point, whereas far away points have a more independent dynamics. This behaviour is expected, however, at increased time lag values, we observe that the correlation diminishes in the vicinity of the target, and

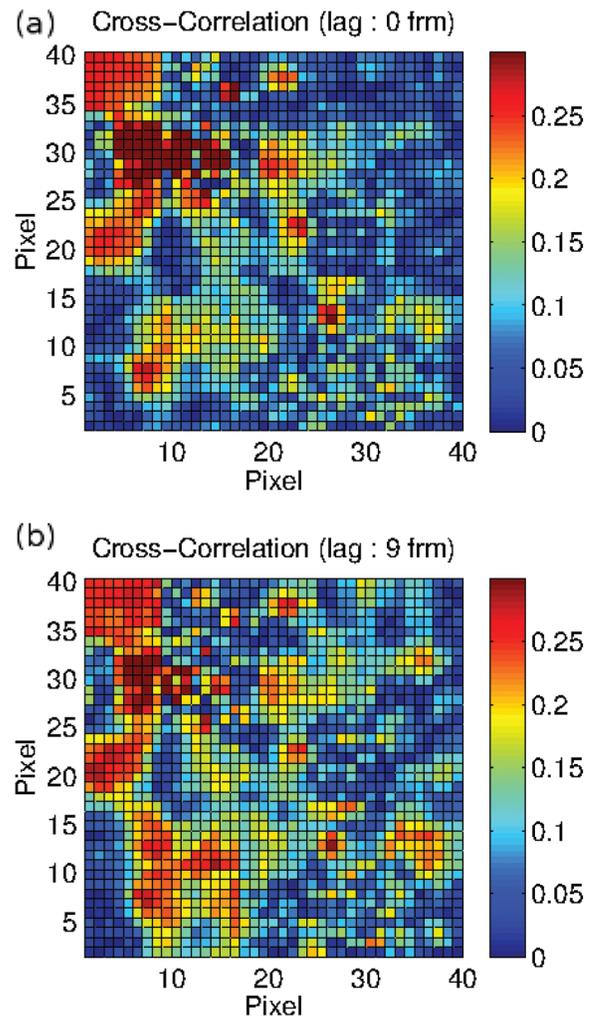


FIG. 5. The cross-correlation coefficient map to the point located in (10,30) for two different time lags. The blinking behaviour of this single point is correlated with the rest of the map at different times (panel (a), simultaneously, panel (b) with a delay equal to 9 frames, about 270 ms). The correlation coefficient decreases in the source and increases and expands at greater distance with time, suggesting that the *beating* wave is propagating. Correlation values higher than 0.3 have the same colour for ease of identification, one pixel corresponds to about  $1 \mu\text{m}$ .

regions relatively distant from the center of blinking appear to increase in correlation (see area around  $x_i = (12,10)$  in Fig. 5, panel (b)). This behaviour reveals the expansion of mechanical beats located on the target and diffusing away from it. The structure and granularity of the correlation coefficient map indicates changing compositional and mechanical properties along the lattices.

We have developed a simple model and experimental tests that suggest that optical instabilities in InGaN quantum wells are caused by mechanical beats of thermal vibrations of the quantum well lattice. Optical effects induced by these beats have been observed experimentally through monitoring with CCD camera the photoluminescence and making a time resolved correlation analysis. The thermal dependence of the phenomena is compared with a Boltzmann distribution of a harmonic oscillating lattice, resulting in good match and agreement with the original hypothesis.

We believe that the understanding of the thermal nature of the optical blinking in these devices is important not only for the research of the fundamental phenomena involved in the emission of InGaN materials, but it can also constitute the basis for a new general methodology of analysis of InGaN subtle local mechanical properties, revealing with a very simple and pure optical method, extremely small variation of doping concentration and possibly other fine compositional or structural anomalies of the crystal.

This study was supported by the Japanese grant KAKEN (project number 24560014), we also want to thank AAA Translations UK for help in improving the manuscript.

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