Electroluminescence from GaN nanostructures

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Abstract

This work focuses on the study of electroluminescence (EL) intensity from GaN nanoclusters (ncs) as a function of different parameters, such as size of the ncs, applied voltage, wavelength, temperature, and time. To do this, we used the idea of quantum confinement model (QCM) and developed model equation that describes the *EL* intensity as a function of size of the nanoclusters and Fourier transform of the size dependent intensity into energy dependent intensity equations were derived; as well as experimentally fit equations were used and MATLAB codes were developed to generate data. From our result it is shown that as the number of ncs decreases, higher EL intensity is obtained. However, selecting the number of samples is based on the applied voltage and wavelength. The high efficient EL is obtained at low operating voltages about 6V (in our case). It is also observed that EL intensity decreases as the temperature increases and degrades with time.

Keywords: Gallium-nitride, nanocrystals, quantum confinement, recombination, electroluminescence.

Resumen

Este trabajo se centra en el estudio de la electroluminiscencia (EL) y de la intensidad de nanoclusters de GaN (NCS), como una función de diferentes parámetros tales como: el tamaño de las NCS, el voltaje aplicado, la longitud de onda, la temperatura y el tiempo. Para ello, se utilizó la idea de modelo cuántico confinamiento (QCM); y la ecuación de un modelo desarrollado que describe la intensidad EL en función de: el tamaño de los nanoclusters, de la transformada de Fourier, de la intensidad dependiente del tamaño en energía dependiente de la intensidad, las ecuaciones fueron derivadas. Experimentalmente se utilizaron ecuaciones de ajuste y se desarrollaron códigos de MATLAB para generar datos. De nuestro resultado se muestra que conforme el número de NCS disminuye, se obtiene mayor intensidad EL. Sin embargo, la selección del número de muestras se basa en la tensión aplicada y en la longitud de onda. La alta eficiencia EL se obtiene a voltajes de operación bajos, de aproximadamente 6V (en nuestro caso). También se observa que la intensidad EL disminuye a medida que aumenta la temperatura y se degrada con el tiempo.

Palabras clave: Galio-nitruro, nanocristales, confinamiento cuántico, recombinación, electroluminiscencia.

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I. INTRODUCCIÓN

Nanoscience is field of science that involves studying and working with matter on nanoscale (on the order of 10^{-9}m). It creates a plentiful supply of new materials which deeply impact our economy, environment, and society by nanotechnology [1, 2]. In nanoscale materials the surface-to-volume ratio is greatly increased and brings a better change in the intrinsic properties of the materials. When the device is small enough, the energy levels become discrete, which allows the output energy to be adjusted according to the size of the material [2]. In studying properties of semiconductor quantum confinement (QC) is so important. It is either or both the electron and hole experience **confinement** in semiconductors by a potential well in 1D (quantum well), 2D (quantum wire), or 3D (quantum dot) [3, 4]. It can significantly enhance the exciton binding energy which is related to EL intensity [5].

Quantum dots are semiconducting nanoparticles that are able to trap electrons in small spaces. QDs confine electrons, holes, or electron-hole pairs to zero dimensions on the order of electrons de Broglie wavelength. They contain a single unit of charge and give off different colors of light depending on size and specific energy levels [6].

The density of states in QDs is represented by the number of confined states divided by the energy interval. If the energy interval is approaching zero, then the density of states is simply a series of delta (δ) functions centered on the confined energy levels ($\varepsilon_1, \varepsilon_2, \varepsilon_3 \ldots$). The energy levels are entirely discrete and given by:

$$E_{nxnynz} = \frac{\pi^2 \hbar^2}{2m^*}\left(\frac{n_x^{\,2}}{L_x^{\,2}} + \frac{n_y^{\,2}}{L_y^{\,2}} + \frac{n_z^{\,2}}{L_z^{\,2}}\right). \quad (1)$$

The density of state is:

$$g(\varepsilon) = 2\theta(\varepsilon - \varepsilon_j) . \quad (2)$$

The factor 2 accounts for spin degeneracy semi-conductor nanoparticles or quantum dots (QDs) have rich optical properties that strongly depend on size, especially when the particle size is less than the exciton Bohr radius of the material [8, 9].

The electronic and optical properties of group III nitride materials are of great interest for light-emitting diodes (LEDs), lasers, and other optoelectronic devices, because of their band structures and the large range of band gap (0.7-6.2 eV) that can be covered by these nitrides and their alloys. GaN with a band gap of 3.4 eV have received a great deal of attention for its blue and ultraviolet light emission and because of application in high-temperature, high-power devices [10].

When semiconductor gets an external perturbation with larger energy than its band gap, an electron in the valence band acquires suitable energy to reach to the conduction band. This induces formation of a hole in the valence band.

The generated electron and the hole tend to return to their ground state by giving up the excess energy through recombination. Before recombination, the generated electrons and holes experience various processes such as diffusion, drift, scattering, exciton formation, and so on. If the generated carriers give up their energy in the form of light, the recombination is called as radiative recombination.

Otherwise, if the generated carriers lose the excess energy in the form of heat, the process is called as non-radiative recombination [11].

EL has been reported for porous silicon during anodic oxidation, and also for porous silicon devices employing thin gold, indium tin oxide, silicon carbide, and polymer contacts.

EL is a light emission phenomenon caused by the electric current or strong electric field passing through a material.

When electrons and holes recombine in the material, the energy of the excited electrons will be released in the form of photons [12, 13]. EL lighting is commonly used in accent lighting applications, for automotive instrument panel back-lighting, aircraft panel lighting, back-lights for liquid crystal displays and cell phones, and in battery-operated devices like wristwatches, night-lights, computer monitors, and more [14].

II. DEPENDENCE OF EL INTENSITY ON DIFFERENT PARAMETERS

Semiconductors are transparent to photons whose energies lie below their bandgap and are strongly absorbing for photons whose energies exceed the bandgap energy. When light passes through a material, the light is partially scattered, absorbed, or transmitted. The transmitted beam intensity is exponentially decayed with the specimen thickness and absorption coefficient. The absorption coefficient, σ, is a property of a material which defines the amount of light absorbed by it. The light intensity as a function of the effective thickness of the sample (say z) is derived from the probability of light being absorbed or scattered out the beam in thickness dz and is given as [15]:

$$I(z) = I_o e^{-\alpha.Z} .$$

Where I_0 and I are the intensities of the incident light before and after it passes the sample and α (σc) is linear attenuation coefficient [11]. The absorption coefficient, $\alpha(E)$, for an photon energy (E) higher than the band gap energy is given by:

For a direct semiconductor

$$\alpha(E) = \alpha_o \sqrt{\frac{E - E_g}{E_g}} .$$

For an indirect semiconductor:

$$\alpha(E) = \alpha_o \frac{(E - E_g)^2}{E_g} .$$

Where E is photon energy, E_g is energy gap and α_o is absorption coefficient fitting parameter [11]. EL intensity depends on temperature. Its dependence on temperature was measured in the high temperature range [16]. EL intensity as a function of temperature with intensity at absolute zero temperature (I_0) can be fitted by a standard equation [17]:

$$I_{EL}(T) = \frac{I_o}{1 + C_1 \exp(\frac{-E_1}{K_B T}) + C_2 \exp(\frac{-E_2}{K_B T})} .$$

Where E_1 and E_2 are the two thermal quenching activation energies, T is the temperature and K_b is the Boltzmann constant, C_1 and C_2 are coupling coefficients. The EL thermal quenching activation energy of GaN is 190meV.

Interestingly, in the sample, the device current as a function of temperature depicts nearly exact anti-correlation with the EL intensity and exhibits activation energy of $\approx$ 200 meV [18].

The radiative and non-radiative processes occurring in the active layer containing GaN nanoclusters have been studied by means of time-resolved luminescence measurements. Time resolved measurements of the EL signals have been extracted by fitting the experimental curves with the equation given by [19, 20]:

$$I(t) = I_o \exp\left[-(t/\tau)^{\beta}\right].$$

Where I (t) and I_0 are the EL intensity as a function of time and at t = 0, respectively, τ is the EL decay time constant,

and β is a dispersion factor ≤ 1. In the case of single exponential decay the β equal to 1. If the decay consists of a combination of several exponentials, reflecting dispersion in life times for the same observation wavelength, then the value of β is less than 1 [21].

III. METHODS AND FORMULATION OF MODELS FOR EL

A. Methods

We designed computational method which is based on theoretical study and numerical analysis of *EL* intensity from GaN-NCS as a function of different parameters (such as wavelength, number of nanoclusters, applied voltage, time and temperature). The theory is supposed to be developed model equation, solving the problem analytically and numerical method for determining the properties of EL intensity as a function of different parameter. That is, we developed theoretical model equation that describes the EL intensity as a function of size of the nanoclusters using quantum confinement model and used Fourier transform of the size dependent intensity into energy dependent intensity equations. After we have derived model equation as a function of energy, we developed suitable MATLAB code for our master equation and theoretically given general equation to generate data and plotted their graphs in different legends.

B. The Quantum Confinement Model (QCM)

Quantum confinement is a very successful model for understanding the electronic structure of nanometer size semiconductor structures such as quantum wells, nanocrystals, and clusters. It predicts that with decreasing particle sizes the band gap increases due to shifting of the band edges [22]. In this model, the luminescence process is attributed to an energy shift of carriers and is proportional to L^{-2}, where 'L' is diameter of the GaN-ncs.

Assuming that a Gaussian size distribution about the mean diameter L_0 for the nanocrystallites [23]:

$$I(d) = \frac{1}{\sqrt{2\pi\sigma}} \exp\left(-\frac{(L-L_o)^2}{2\sigma^2}\right). \quad (1)$$

Where L is the cluster diameter and σ is the standard deviation.

The number of carriers (N_c) in a column diameter 'L' participating in the EL process is proportional to L^2.

Therefore it is given as:

$$N_c \approx L^2 \Rightarrow N_c = aL^2 \ . \quad (2)$$

Where, "a" is suitable normalization constant.

For a GaN sample consisting of varying column diameters the probability distribution of electrons participating in the *EL* process is:

$$I(d) = \frac{1}{\sqrt{2\pi\sigma}} aL^2 \exp\left(-\frac{(L-L_o)^2}{2\sigma^2}\right). \quad (3)$$

The luminescence intensity can be determined by the Fourier transform of Equation 3 to the energy axis as:

$$I(\Delta E) = \int_0^\infty \frac{1}{\sqrt{2\pi\sigma}} aL^2 \delta\left(\Delta E - \frac{C_1}{d^2}\right) \exp\left(-\frac{(L-L_o)^2}{2\sigma^2}\right) dL \ . \quad (4)$$

Where, C_1 is arbitrary constant.

The Dirac delta function facilitates a straight forward integration. Simplifying and applying the properties of Dirac delta function, the above integral gives the intensity as a function of ΔE.

$$I(\Delta E) = \frac{aC_1^{\frac{3}{2}}}{\sqrt{8\pi\sigma}} \Delta E^{\frac{-5}{2}} \exp\left(-\frac{L_o^2}{2\sigma^2}\left[\left(\frac{\Delta E_o}{\Delta E}\right)^{\frac{1}{2}} - 1\right]^2\right). \quad (5)$$

Where ΔE is the energy shift due to the confinement given by:

$$\Delta E = h\nu - (E_g - E_b). \quad (6)$$

Where h is Planck's constant E_g is the bulk band gap of GaN in this sample and E_b is the exciton binding energy.

Depending on temperature the values of E_g ranges from 3.39 eV to 3.52 eV; however, we took 3.4 eV which is actually reported in most standard experiments at room temperature, while the value of E_b varies with nanocrystal radius of the sample. For GaN-ncs it is reported as in the range 20.4meV to 28meV [17, 24, 25].

The EL spectrum is caused by injection of strong electric field **E** or current, the energy associated to this field is:

$$\varepsilon = NZeEL = NZVe \ . \quad (7)$$

Where N is the number of nanocrystallites GaN taken in the sample, Z is the number of electrons in each crystal, e is the charge of electron, E is the magnitude of external applied electric field which is responsible for EL and in the direction of diameter, 'L' is the diameter of the single nanocrystal and V is the voltage applied to the nanocrystal sample; since EL and light emitted due to photon energy (photoluminescence (PL)) occurs at the same energy, using the analogy of the PL spectrum model we can write the energy shift as:

$$\Delta E = NZVe - (E_g - E_b). \quad (8)$$

According to the QC model, the emission wavelength and intensity depends on nanocrystal diameter, size distribution and concentration [26, 27].

IV. RESULTS AND DISCUSSIONS

Figure 1 shows the EL intensity as a function of wavelength for GaN nanostructures. When we compare our result and with the experimental result in [28], nearly they have the same nature Gaussian shaped curve.

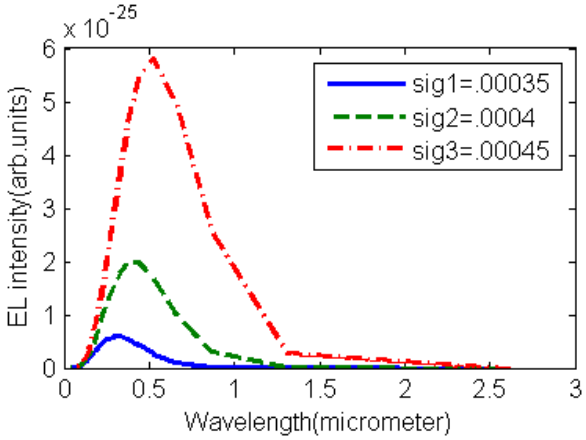

FIGURE 1. EL intensity vs wavelength.

In both cases the EL intensity has sharper peak when near the center of the Gaussian curve. The figure shows that as wavelength increases the intensity spectra shifts to lower energy (red-shifted) and as we go further intensity becomes out of visible range.

Figure 2a shows EL intensity as a function of number of sample GaN nanoclusters for different fixed values of applied voltage. Decreasing the number of nanoclusters in the sample results in decreasing the diameter of the nanoclusters, this results increase in the luminescence intensity since it is related by the equation of our model.

When the size of nanoclusters decreases, the surface-to-volume ratio increases leading to quantum confinement effect that enhance radiative recombination rate of excitons for emission of light.

It is shown that in case of smaller particles, higher EL intensity is obtained. However in selecting the number of samples in order to get EL spectrum in the visible range, it should be based on the applied voltage. As shown in the figure for 20V at the lower number there is no EL intensity.

This implies that when we choose too large or too small sample for a fixed voltage, the EL spectrum will lie in the ultraviolet or infrared region respectively, which has no physical utility for applications since both are out of visible light ranges. Figure 2b shows EL intensity as a function of voltage for different fixed number of nanoclusters of GaN.

As shown in the figure the EL intensity increases as the value of applied voltage for each nanoclusters increase and then reaching certain maximum peak it decrease infinitely with increasing value of the applied voltage.

The causes for this effect may be carrier linkage at high forward currents, Auger recombination, or junction heating carrier delocalization. Yet to have a spectrum in the visible range for a given size of nanoclusters we need to select the appropriate range of values of voltage. In our sample case we get maximum average intensity when the applied voltage is about 6v.

Figure 3 shows that the dependence of EL intensity on the temperature of the nanoclusters, which relates with in Ref [19]. As shown in the figure, as the temperature increases the EL spectrum shifted decreasing the intensity of EL.

This is due to high absorption and the shrinkage of band gap with increasing temperature. The EL quenching at increased temperature is a combination of a reduced confinement and thermally activated non-radiative recombination processes within "active" crystallites.

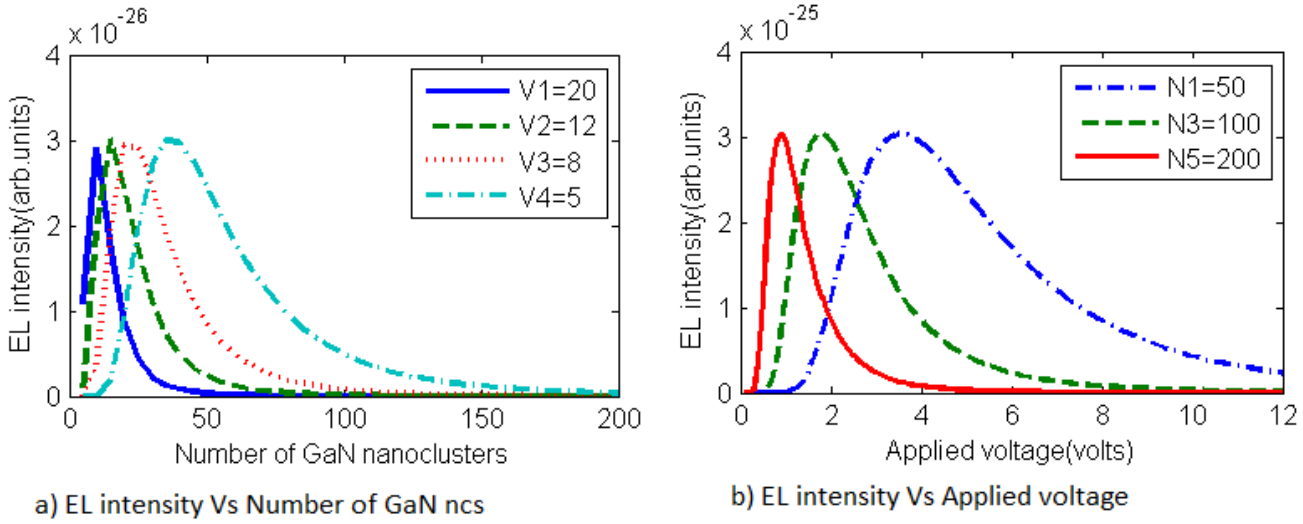

FIGURE 2. EL intensity as a function of: a) number of sample GaN nanoclusters, b) applied voltage.

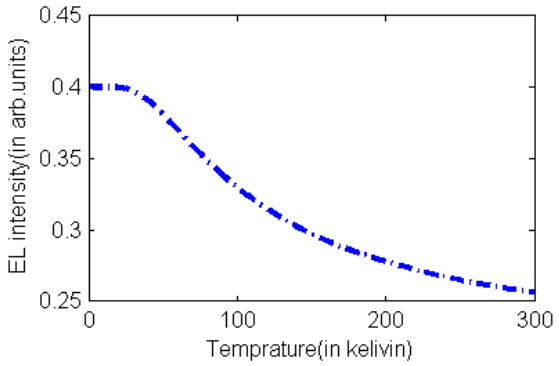

FIGURE 3. EL intensity as a function of temperature.

The figure 4 shows the time dependence of EL intensity of our result which is much with experimental value in [20], both have similar pattern. A shorter decay time constant allows a higher modulation frequency, but reduces the efficiency. This is due to the decrease in oscillation period and resulting non radiative recombination of the particles in the nanocrystal.

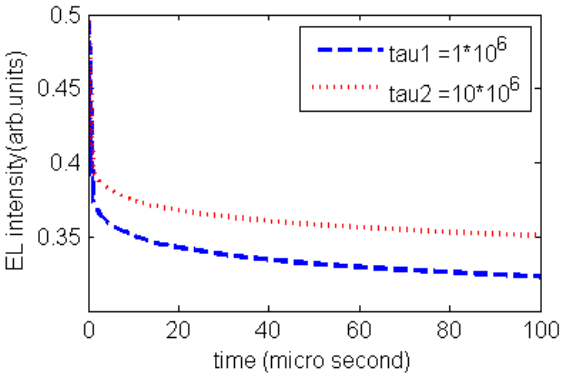

FIGURE 4. EL intensity as a function of time.

IV. SUMMARY AND CONCLUSION

This work was inspired by the recently discovered phenomenon that GaN nanostructures in the nanometer size range are able to emit visible light. The aim was to investigate the EL intensity properties from GaN nanostructures. QC effect is more prominent in nanostructures as an efficient luminescence source because it enhances radiative recombination rate of excitons and carrier localization effect with decreasing the cluster size.

Band gap increases with decreasing size which allow light to emit and useful to observe the intensity of emitted light. EL spectrum occurs by electrical excitations (current and high injection field), has Gaussian sub peaks and more red shifted with increasing wavelength, nanocrystals size and applied voltage. A blue shift of EL is observed with decreasing these parameters.

Generally, the intensity of the spectrum depends on the HOMO-LUMO gap, the size of the nanocluster and the energy shifted due to confinement, applied voltage, temperature, transient time and size and nature of the nanocluster sample taken. For smaller particles, higher EL intensity is obtained and light emission starts at lower threshold. EL shows either reversible or permanent degradation with time. It usually increases at lower temperature due to inhibition of electron-phonon interactions and thereby increases the excited electronic state lifetime.

However further treatment improves the stability of the EL intensity.

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