Inhomogeneous Model for the Investigation of the Optical Properties of the A-Plane Oriented ZnO Epilayers Grown by Plasma-Assisted Molecular Beam Epitaxy

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Abstract

Experimentally the a-plane oriented ZnO showed three types of excitons A, B and C. The theoretical investigation became important to get access inside all excitonic parameters impossible with experimental. Authors used recently the Gaussian distribution by using new parameters to fit perfectly the free excitons. In the present work, one presents another model entitled an inhomogeneous model by taking account another form of Gaussian function, to plot and interpret the excitons A, B and C by using as well the same physical parameters than the theory of the spatial resonance dispersion of Hopfield model. The data found with the inhomogeneous model are almost the same than experimental data found the authors.

Keywords: Exciton A, B and C; Inhomogeneous model; Reflectivity spectrum; A plane oriented ZnO

Introduction

The zinc oxide (ZnO) is a semiconductor II-IV investigated since several years, that is why many physical properties of the ZnO in large number are already known [1-5]. Now a day, the possibility to increase the thins films and hetero-structures in nanoscale with high quality have attracted the attention of the researchers. With its wide band gap (E_g=3.37 eV) and its larger exciton binding energy (60 meV) several applications have been made such as the ultraviolet (UV)-stimulated emission induced by the exciton-exciton scattering at moderate pumping intensity [6], the diluted magnetic semiconductor with the authors [7] which demonstrated theoretically that at room temperature, a ZnO matrix doped with the transitions metals such as V, Cr, Mn, Fe, Co, and Ni present a ferromagnetic behavior as well as several others applications [8-18]. Beside the experimental manipulation, the theoretical aspect played an important role for the investigation of all the excitonic parameters impossible for access with the experimental manipulation. That is why, the authors [8,9,19,20] investigated theoretically and respectively the mixed states of excitons and photons by using the quantum theory of a classical dielectric. The temperature dependent PL of the A free exciton peak energy measured in the case of the perpendicular polarization (E ⊥ c) by using the Varshni empirical equation and the theoretical reflectivity of the excitons A, B and C in both polarization parallel and perpendicular by using the Gaussian distribution as well as several others researchers. In this article one presents another model unless the Gaussian distribution used by the authors [9] to investigate the experimental reflectivity observed in the a-plane oriented ZnO in the polarization, parallel and perpendicular, entitled the inhomogeneous model [21,22] in the framework of a numerical investigation [23-27], to plot the exciton A, B and C. One will compare our results with those obtained in Lo B, Diouf AA [8,9] using other theoretical treatments. In Section 2, Computational Method, one will present the inhomogeneous model which will allow us to determine the theoretical reflectivity of the a-plane oriented ZnO and the results obtained are well discussed in the section 3.

Computational Method

To model the optical properties, one used the inhomogeneous model defined by:

For one oscillator:

$$\varepsilon_r(E) = \varepsilon_b + \int \frac{A_j}{(x^2 - E^2 + i\Gamma E)} f(x, E_0)\, dx \quad (1)$$

With

$$f(x, E_0) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left\{\frac{-(x - E_0)^2}{2\sigma^2}\right\} \quad (2)$$

For N oscillator:

$$\varepsilon_r(E) = \varepsilon_b + \sum_{j=1}^{N} \int \frac{1}{\sqrt{2\pi}\sigma_j} \frac{A}{(x^2 - E^2 + \Gamma_j E)} \exp\left\{\frac{-(x - E_0)^2}{2\sigma_j^2}\right\} dx \quad (3)$$

With

$$A_j = \left(\frac{h}{2\pi}\omega_{pj}\right)^2 = \alpha_{0,j}\frac{h^2}{\pi}\omega_{0,j} = \frac{Ne^2h^2}{4\pi^2 m^*\varepsilon_0}, \omega_{pj}$$

Being similar to a plasma pulsation, $\alpha_{0,j}$ polarizability of the exciton, ε_b the high-frequency dielectric constant of the material (outside the excitonic resonance), Γ the spectral width, h the Planck constant, ω_0 is the resonance frequency, N the number of particles, e_0 the elementary charge electron, m^* is the effective mass and x is the resonance energy of excitons.

The inhomogeneous model allows to determine the exciton energy $E_{0,j}$, the oscillator strength A_j and the parameters for the widening of the excitonic ray (h/2π). Γ_j, it includes in general the influence of the

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temperature and the quality of the material (homogeneity, fluctuations in thickness etc.).

Moreover, the modeling of the optical reflectivity, one can do an investigation of the thermal behavior and turbulent of the system with the spectral widening which is a function of the temperature "T" and the effective masse "$m*=0.59m0$" of the exciton defined by:

$$\Gamma(T) = \sqrt{\frac{k_B \times T}{0.59 \times m_0}} = \beta.T^{1/2}$$

With

$$\beta = \sqrt{\frac{k_B}{0.59 \times m_0}} \quad (4)$$

T: Temperature

m_0: Mass of a free electron

k_B: Constant of Boltzmann.

Results and Discussion

Experimentally the optical characterization of the a-plane oriented ZnO shows three types of excitons A, B and C, according to the choice of the polarization, parallel or perpendicular. The energy of the three excitons have been found experimentally by authors [8] equal to $E_A=3.398$ eV ($\omega_A=0.516 \times 10^{14} s^{-1}$), $E_B=3.410$ eV ($\omega_B=0.518 \times 10^{14}$) and $E_c=3.438$ eV ($\omega_B=0.522 \times 10^{14}$). Diouf et al. [9] investigated theoretically and found almost the same values than the experimental values than Lo et al [8]. by using the Gaussian distribution defined by:

$$R(\omega) = R_0 + \alpha.\sum_{i=1}^{n}\left(\frac{A_i}{\Gamma\sqrt{\frac{\pi}{2}}}\right) \times \exp-\left(\frac{h^2(\omega - \omega_{0i})^2}{2\pi^2\Gamma_i^2}\right) \quad (5)$$

Where the coefficient α is related to the oscillator strength ($4\pi N e_0^2/m*$), ω_0 is the resonance frequency $e_0=e/4\pi\varepsilon_0$ with "e" designated as the elementary charge electron R_0 the reflection coefficient A_i the spectral area and Γ_i the spectral width. In the present study one built a program from the inhomogeneous model (1) to investigate the optical properties of the a-plane oriented ZnO. The results obtained by simulation are summarized in the Table 1 compared to the results found experimentally by the authors [8] (Table 2). A comparison of the tables shows that the inhomogeneous model Figure 1a and Figure 2a, is more accurate than the model used by the authors [8] to plot the reflectivity curve of the exciton A B. Otherwise one insists on the fact

that the parameters summarized in the (Table 1) are used to model the theoretical reflectivity of the exciton A B. In Table 2 one has the values used by the authors [8] to have their best fit of the free excitons. The method used with the inhomogeneous model allowed furthermore the excitons A B Figure 1b, to plot the free exciton C shown in the Figure 2b thing
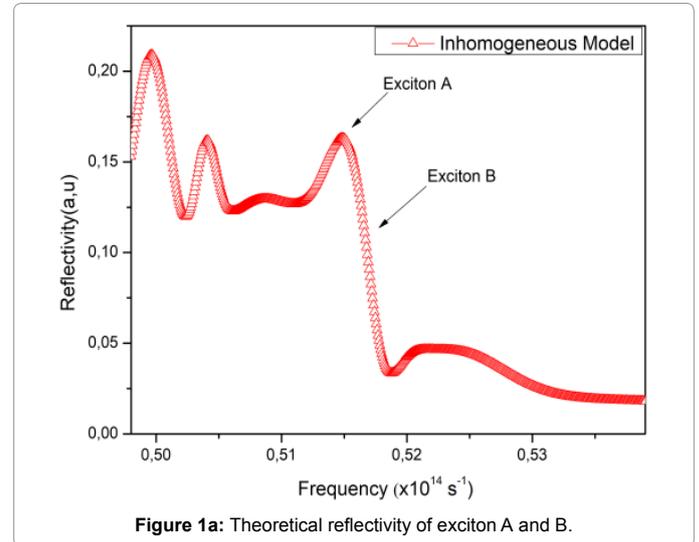

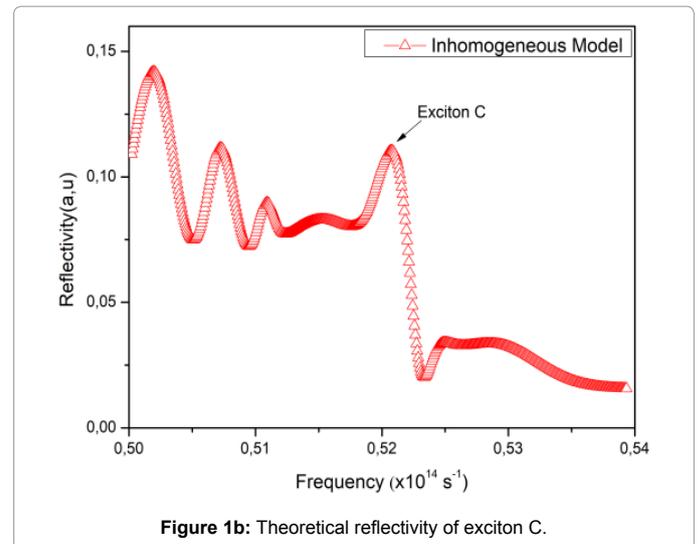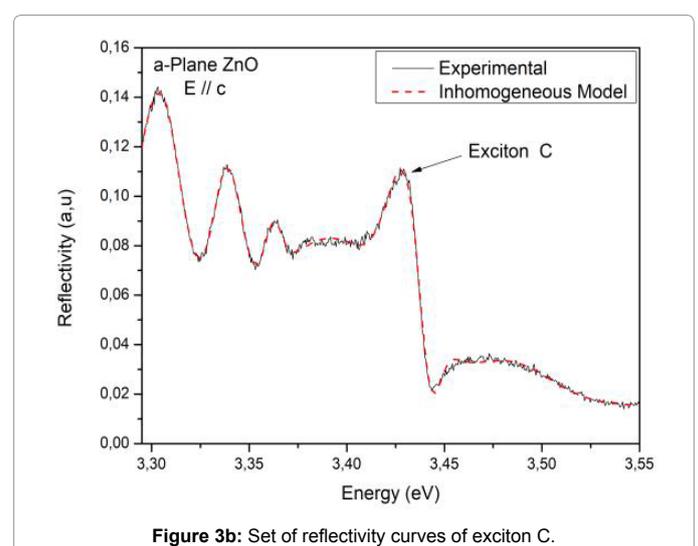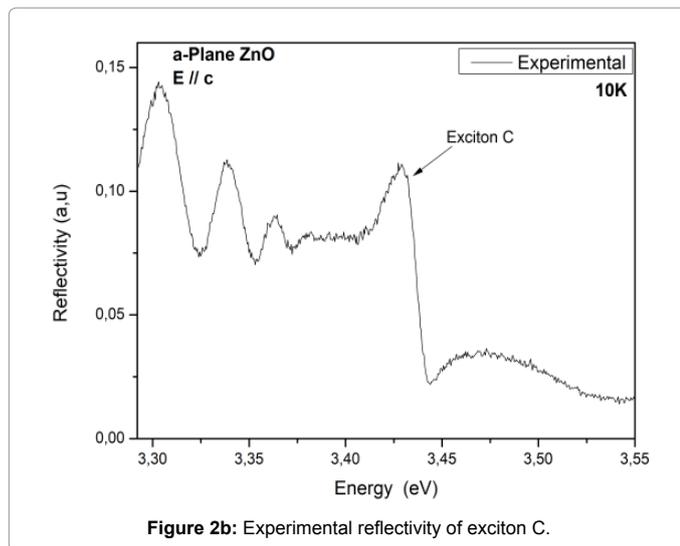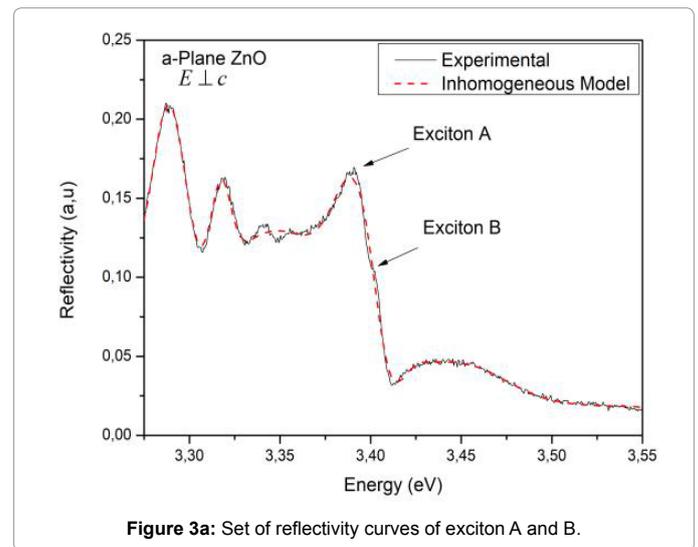**Figure 1a:** Theoretical reflectivity of exciton A and B.

Figure 1b: Theoretical reflectivity of exciton C.

Free Exciton	Frequency (ω_0) (s^{-1}) Energy(eV)	Oscillator Strength (α,	Spectral widening (Γ, (m.s^{-1})	Effective mass (m*)
A	5.15E+13 (E=3.390 eV)	1.705	3.25E+12 (Γ=2.14 meV)	0.59 m_0
B	5.16E+13 (E=3.401 eV)	0.759	4.00E+14 (Γ=26.3 meV)	0.59 m_0
C	5.21E+13 (E=3.430 eV)	-2.044	2.53E+10 (Γ=1.66 meV)	0.59 m_0

Table 1: Values of A, B and C free exciton frequency (ω, (s^{-1}), the spatial widening (Γ), the effective mass (m*) and the Area (A_i) of the reflective curves obtained by using the Gaussian distribution.

Free Exciton	Energy (E) (eV)	Oscillator Strength (α)	Spectral widening (Γ) (meV)	Effective mass (m*)
A	3.393	1.708	10.38	0.59 m_0
B	3.403	0.77	11.479	0.59 m_0

Table 2: Values of A and B free exciton Energy E(eV), Oscillator Strength (α), the spatial widening (Γ) and the effective mass (m*) of the exciton obtained by using the Hopfield model [8].

Figure 2a: Experimental reflectivity of exciton A and B.

Figure 3a: Set of reflectivity curves of exciton A and B.

Figure 2b: Experimental reflectivity of exciton C.

Figure 3b: Set of reflectivity curves of exciton C.
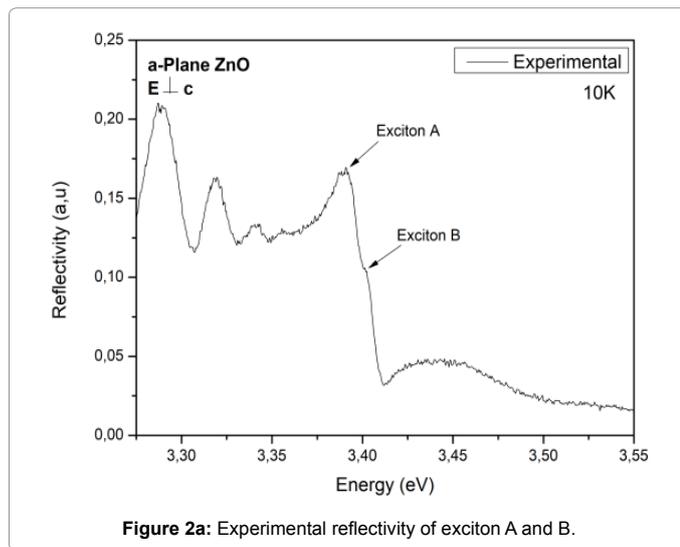
found by Diouf [9]. One can explain the phenomenon but the fact that the reflectivity curves are composed by several Gaussian shapes thus with the inhomogeneous model according to the Gaussian distribution it was easy to find the reflectivity curves by playing in the value of the oscillator strength for more accurate. As well a comparison between the authors [8] with the Hopfield model and the inhomogeneous model show important difference summarized in the tables unless the simple Gaussian model (5, used by the authors [9]. One noted that in the range of energy between 3.375 eV and 3.425 eV which represents the reflectivity pic energy range all the models are accurate with the experimental reflectivity curves. But from 3.275 eV to 3.375 eV representing the range of transparency zone one observed that the inhomogeneous model used plots the whole reflectivity curve obtained experimentally by the authors [8]. One can explain this result by the fact that the inhomogeneous model takes accounts the optical properties when the frequencies (ω, are lower than the resonance frequency (ω_0, exactly like the simple Gaussian distribution used by authors [9]. The parameters used to plot the free exciton C are summarized in Table 1. The inhomogeneous model can determine separately the both types of widening (Figure 3). The homogeneous widening which depends of the temperature noted Γ represents the interaction exciton-photon it

increases with the temperature otherwise the inhomogeneous widening depends on the quality of the material through the inhomogeneity of the thickness composition or stress. As well with the inhomogeneous widening it is taken into account through the collection of harmonics oscillators the energy which is distributed according to the Gaussian centered on the average energy E_0 and the width halfway up $2\sigma\sqrt{\ln 4}$.

One used also the spectral widening to investigate the thermal behavior of a-plane oriented ZnO (Figure 4). One noted that from 8 K to 300 K the thermal profile of the sample of ZnO augments with the enhancement of the temperature. One can explain this phenomenon from the relation (4), by the fact that the temperature is an increase function of the spectral widening. Reason why the authors [8] during their experiments used a cryostat to control the temperature. Unless the others models [28-31] which are limited on the investigation of the c-plane oriented ZnO. But the inhomogeneous model moreover c-plane oriented ZnO allowed to investigate the structure of the a-plane oriented ZnO and one plotted in the same time the excitons A B and C something that one did not find in the literature with the others models unless the model presented recently by the authors [32-34].
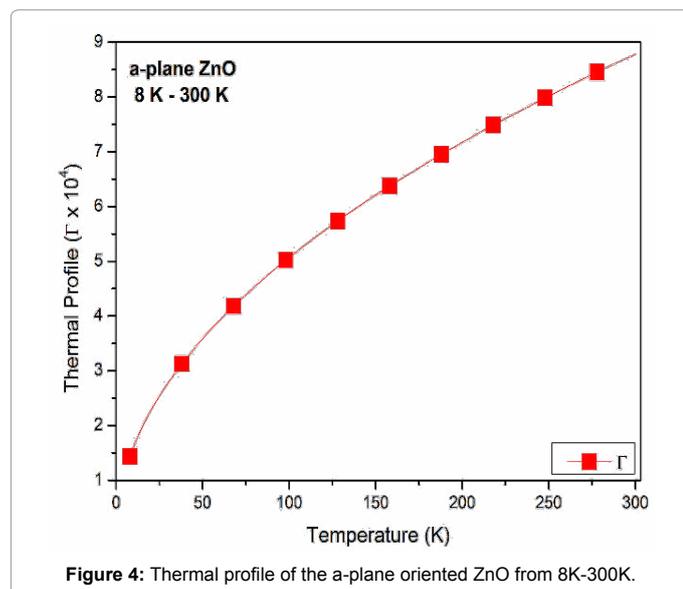
Figure 4: Thermal profile of the a-plane oriented ZnO from 8K-300K.

Conclusion

In this paper one presented another model entitled inhomogeneous model which can be used for the optical characterization. One found the same theoretical reflectivity curves of the free excitons A, B and C in comparison with the experimental results found by Lo et al. by using the same parameters than the authors [8]. Otherwise, one compared the inhomogeneous model with the simple Gaussian distribution used by authors [9], one has the same results the only difference is the spectral area used in the Gaussian distribution [9]. One investigated as well, the thermal profile of the a-plane oriented ZnO using the spectral widening of the inhomogeneous model. All these results allowed us to present two mathematics models which can be used for the characterization of the nonlinear optical properties with a specific simulation code before getting the experimental results.

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