

# Inhomogeneous Model for the Investigation of the Optical Properties of the a-plane Oriented ZnO Epilayers Grown by Plasma-Assisted Molecular Beam Epitaxy

Alioune Aidara Diouf<sup>1, 2, \*</sup>, Bassirou Lo<sup>1</sup>, Balla Diop Ngom<sup>1</sup>, Abib Fall<sup>1</sup>, Aboubaker Chedikh Beye<sup>1</sup>

<sup>1</sup>Department of Physics, Faculty of Sciences and Techniques, Cheikh Anta Diop University, Dakar-Fann, Senegal

<sup>2</sup>Department of Science and Technology, Iqra Bilingual Academy, Point E, Senegal

## Abstract

The optical properties of the a-plane oriented ZnO described by an inhomogeneous model are investigated by using another form of Gaussian function. The reflectivity spectrum of the excitons A, B and C are represented by using the same physical parameters than the theory of the spatial resonance dispersion of Hopfield model. The data obtained are well discussed and are, as well, almost the same than the experimental data found by the authors.

## Keywords

Exciton A, B and C, Inhomogeneous Model, Reflectivity Spectrum, A-plane Oriented ZnO

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## 1. 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 thin films and heterostructures in nanoscale with high quality have attracted the attention of the investigators. With its wide band gap ( $E_g=3.37\text{eV}$ ) 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 transition metals such as V, Cr, Mn, Fe, Co, and Ni present a ferromagnetic behaviour as well as several other applications [8-18]. Beside the experimental

manipulation, the theoretical aspect played an important role for the investigation of all the excitonic parameters impossible to reach with the experimental manipulation. That is why J. J. Hopfield [19, 20], B. Lo [8] and A. A. Diouf [9] 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 ( $E \perp c$ ) 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 other researchers. In this article one presents another model unless the Gaussian distribution used by the authors [9] to investigate the experimental reflectivity

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\* Corresponding author

E-mail address: aliouneaidara.diouf@ucad.edu.sn (A. A. Diouf)

observed in the a-plane oriented ZnO in the both 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 ref. [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.

## 2. Computational Method

To model the optical properties, one has 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 oscillators:

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

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

similar to a plasma pulsation,  $\alpha_{0,j}$  polarizability of the exciton,  $\varepsilon_b$  the high-frequency dielectric constant of the material (outside the excitonic resonance),  $\Gamma$  the spectral width,  $h$  the Planck constant,  $\omega_0$  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 makes it possible to determine the exciton energy  $E_{0,j}$ , the oscillator strength  $A_j$  and the parameters for the widening of the excitonic ray  $(h/2\pi)\Gamma_j$ , it includes in general the influence of the temperature and the quality of the material (homogeneity, fluctuations in thickness etc.). Moreover, the modelling of the optical reflectivity, one can do an investigation of the thermal behaviour and turbulent of the system with the spectral widening which is a function of the temperature “ $T$ ” and the effective masse “ $m^*=0.59m_0$ ” 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.

## 3. 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 energies of the three excitons have been found experimentally by authors [8] with  $E_A=3.398$  eV ( $\omega_A=0.516 \times 10^{14}$  s<sup>-1</sup>),  $E_B=3.410$  eV ( $\omega_B=0.518 \times 10^{14}$ ) and  $E_C=3.438$  eV ( $\omega_C=0.522 \times 10^{14}$ ). Diouf and al. [9] have investigated theoretically and found almost the same values than the experimental values than B. Lo and al. by using the Gaussian distribution defined by:

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

Where the coefficient  $\alpha$  is related to the oscillator strength ( $4\pi N e^2 / m^*$ ),  $\omega_0$  is the resonance frequency,  $\varepsilon_0 = e/4\pi\varepsilon_0$  with “ $e$ ” designated as the elementary charge electron,  $R_0$  the reflection coefficient,  $A_i$  the spectral area and  $\Gamma_i$  the spectral width. In the present study, one has 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 (cf. Appendix) compared to the results found experimentally by the authors [8] (table 2, cf. Appendix). A comparison of the tables shows that the inhomogeneous model (c.f. Figure 1 and Figure 3) 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 made it possible, furthermore the excitons A, B (cf. Figure 1), to plot the free exciton C shown in the Figure 4, thing found by [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 ( $\omega$ ) are lower than the resonance frequency ( $\omega_0$ ) exactly like the simple Gaussian distribution used by authors [9]. The parameters used to plot the free exciton C are summarized in Table 1 (cf. Appendix). The inhomogeneous model can determine separately the both types of widening. The homogeneous widening which depends of the temperature noted  $\Gamma$ , 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 considered through the collection of harmonics oscillators, the energy which is distributed according to the Gaussian centred on the average energy  $E_0$  and the width halfway up  $2\sigma\sqrt{\ln 4}$ .

One has used also the spectral widening to investigate the thermal behaviour of a-plane oriented ZnO (Cf. Figure 7). One has noted that from 8K to 300K, the thermal profile of the sample of ZnO increases with the enhancement of the temperature. One can explain this phenomenon from the relation (4), by the fact that the temperature is an increasing function of the spectral widening. Reason why the authors [8] during their experiments used a cryostat to control the temperature. Unless the other models [28, 33], which are limited on the investigation of the c-plane oriented ZnO, the inhomogeneous model, moreover c-plane oriented ZnO, made it possible to investigate the structure of the a-plane oriented ZnO and one has plotted in the same time three excitons A, B and C something that one did not find in the literature with the other models unless the model presented recently by the authors [9].

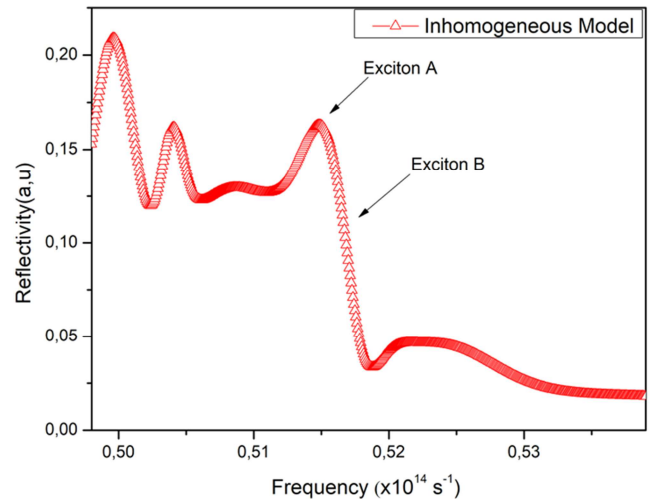


Figure 1. Theoretical reflectivity of exciton A and B.

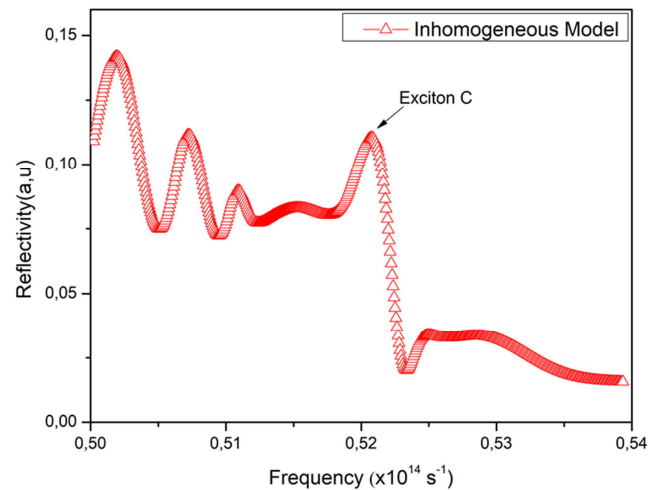


Figure 2. Theoretical reflectivity of exciton C.

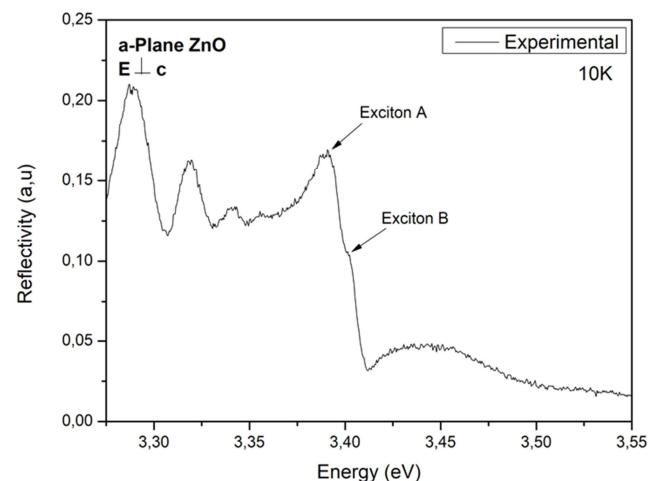


Figure 3. Experimental reflectivity of exciton A and B.

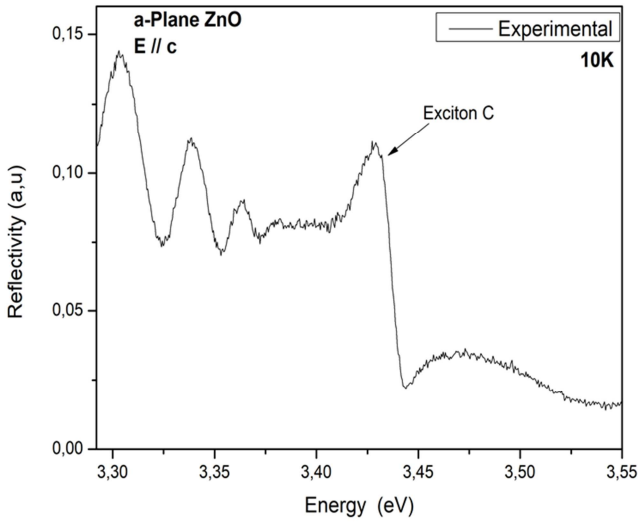


Figure 4. Experimental reflectivity of exciton C.

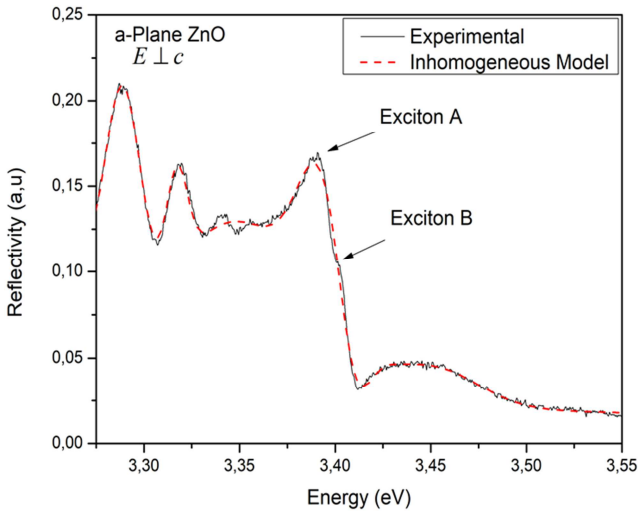


Figure 5. Set of reflectivity curves of exciton A and B.

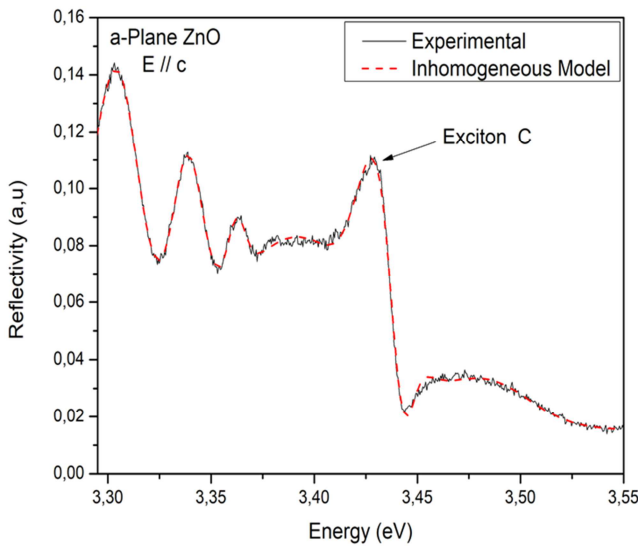


Figure 6. Set of reflectivity curves of exciton C.

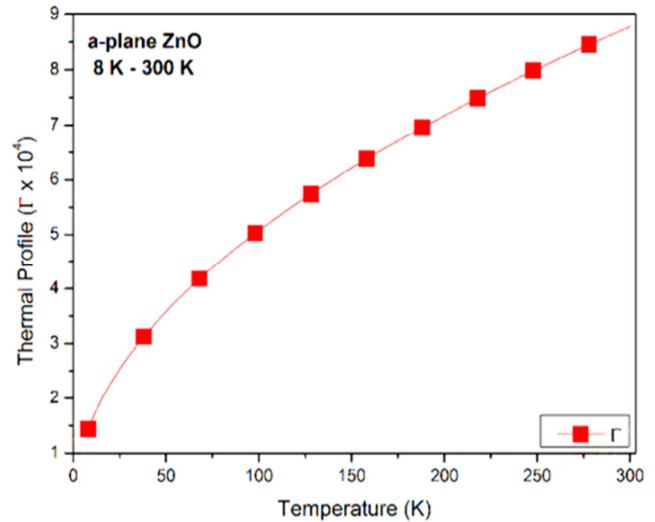


Figure 7. Thermal profile of the a-plane oriented ZnO from 8K - 300K.

## 4. Conclusion

In this paper one has presented another optical model which can be used for the optical characterization. One has investigated theoretically the reflectivity dependent-temperature and -polarization of undoped *a*-plane ZnO. One has found theoretically almost the same reflectivity curves of the free excitons A, B and C, in comparison with the experimental results from B. Lo and al. by using the same parameters than them [8]. The results found by these last, indicated that the *a*-plane-oriented ZnO layer is of high optical quality, thing demonstrated theoretically by the results displayed with the model used. One has identified as well with the inhomogeneous model the A, B and C free exciton transitions found experimentally by authors [8]. Otherwise, one has 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 introduced in the Gaussian distribution in [9]. One has investigated as well, the thermal profile of the *a*-plane oriented ZnO using the spectral widening of the inhomogeneous model. The results have displayed that the temperature increase exponentially, that is why during the experimental investigations authors [8] used a cryostat to control the experimental temperature. All these results made it possible us to present these 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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## Appendix

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

Free Exciton	Frequency ( $\omega_0$ ) ( $s^{-1}$ ) Energy (eV)	Oscillator Strength ( $\alpha$ )	Spectral widening ( $\Gamma$ ) ( $m.s^{-1}$ )	Effective mass ( $m^*$ )
A	0.515E14 (E=3.390eV)	1.705	3.251E12 ( $\Gamma=2.14meV$ )	0.59 $m_0$
B	0.516E14 (E=3.401eV)	0.759	3.996E14 ( $\Gamma=26.3meV$ )	0.59 $m_0$
C	0.521E14 (E=3.430eV)	-2.044	2.53E10 ( $\Gamma=1.66meV$ )	0.59 $m_0$

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

Free Exciton	Energy (E) (eV)	Oscillator Strength ( $\alpha$ )	Spectral widening ( $\Gamma$ ) (meV)	Effective mass ( $m^*$ )
A	3.393	1.708	10.38	0.59 $m_0$
B	3.403	0.77	11.479	0.59 $m_0$

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