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Investigation of the Reflectivity Spectrum of the a-Plane Oriented ZnO Epilayers Grown by Plasma-Assisted Molecular Beam Epitaxy from the Gaussian Distribution

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Abstract: The Photoluminescence spectra at low temperature of the a-plane oriented ZnO grown on r-plane (011-2) sapphire substrates by plasma-assisted molecular beam epitaxy, showed experimentally three types of excitons A, B and C. In the reflectivity spectra, authors used a program based on the theory of the spatial resonance dispersion Hopfield model to fit the free excitons. The A and B free excitons were fitted together and the C exciton with the band gap. But these fits were not perfect in the transparency zone at low energy. This is mainly due to the fact that the A and B free excitons are closer and the C exciton is closer to the band gap but another reason is the value of the oscillator strength. In the present work, we present a method taking account the Gaussian distribution, to fit perfectly the excitons A, B and C using almost the same physical parameters than the theory of the spatial resonance dispersion Hopfield model.

Keywords: Exciton A, B and C, Gaussian Distribution, Reflectivity Spectrum, a-Plane Oriented ZnO

1. Introduction

During the last decade, the zinc oxide ZnO as thin films, nanowires, nano-spheres as well as the correlative quantum structures, have attracted considerable attention both in theories and experiments. ZnO presents a particular interest for the researches for news applications because of its wide band gap (E_g=3.37eV) at room temperature and its larger exciton binding energy (60 meV). Several authors worked on the properties and applications of the semiconductor of ZnO. In solar energy, C. Y. Jiang and al [1] worked on the improvement of the dye-sensitized solar cells with a ZnO-nanoflower photoanode. Yun-Ju Lee and al [2] used ZnO nanostructures as efficient antireflection layers in solar and others authors used the nanostructures and thin films of ZnO to study the interest of zinc oxide [3-6]. In the optical properties, B. Lo and al [7] investigated the optical spectroscopy of a-plane oriented ZnO epilayers grown by plasma-assisted molecular beam epitaxy. In the same way M. Wraback and al [8] as B. Lo and al [7] grew on the r-plane sapphire substrates a ZnO films and investigated the high contrast, ultrafast optically addressed ultraviolet light modulator based upon optical anisotropy in ZnO films. E. V. Lavrov and al [9] worked on ZnO to understand the nature of hydrogen-related shallow donors in ZnO and J. R. Schneck and al [10] for their applications worked on the polar face dependence of the ultrafast U. V reflectivity of ZnO single crystal. Hisashi Yoshikawa and al. [11] gave a precious data to the researchers investigating exclusively the optical constants of ZnO. All these works show the important roles which play the zinc oxide in the new technology reason why the authors continued to study it for more information and applications [12-30]. Beside the experimental methods, certain authors used models to investigate the physical properties of the zinc oxide or others semiconductors. J. Lagois and al. [21] used the model of the harmonic oscillators and the model oscillators having spatial dispersion to describe the optical dielectrics functions. J. J. Hopfield [31, 32] investigated the mixed states of excitons and photons by using the quantum theory of a classical dielectric. B. Lo and al [7] used the Varshni empirical equation to fit the temperature dependent PL of the A free exciton peak energy measured in the case of the perpendicular polarization ($E \perp c$) and D. W. Hamby used the Manoogian and Wooley model [33] for the temperature dependent exciton PL of bulk ZnO.

Our aim in this article is to model the optical reflectivity of the a-plane oriented ZnO. We will use the Gaussian distribution in the framework of the *"bracketing"* method [34-40], to fit the exciton C, which presented a difficulty in certain range of energy because of the oscillator strength related in the ref. [7]. We will compare our results with those obtained in ref. [7, 41-43] using other theoretical treatments. In Section 2, Model, we explained the distribution use 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. Model

To model the optical properties we used the Gaussian distribution defined by:

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

Where

$\alpha = \dfrac{4\pi N e_0^2}{m^*}$ With $e_0 = e/4\pi\varepsilon_0$ and $m^* = 0.59m_0$

$$A_i = \int_{\omega_1}^{\omega_n} \frac{\delta r(\omega)}{\delta\omega} d\omega = \int_{\omega_1}^{\omega_2} \frac{\delta r_1(\omega)}{\delta\omega} d\omega + \int_{\omega_2}^{\omega_3} \frac{\delta r_2(\omega)}{\delta\omega} d\omega + \cdots + \int_{\omega_{n-1}}^{\omega_n} \frac{\delta r_n(\omega)}{\delta\omega} d\omega$$

and

$$r(\omega) = \left(\frac{1}{\Gamma \sqrt{\frac{\pi}{2}}} \right) \times \exp - \left(\frac{h^2 \left(\omega - \omega_0 \right)^2}{2\pi^2 \Gamma^2} \right)$$

R_0: Value of the reflectivity for $\omega=0$
A: Spectral area
Γ: Spectral Widening
ω_0: Resonance frequency
n: Number of intervals
α: Oscillator strength
N: Number of particles
e_0: Elementary charge electron
m*: Effective masse.

To fit the excitons A, B and C, we started to compare two distributions Lorentzian and Gaussian (cf. Figure 1, Figure 3 Vs Figure 2, Figure 4). We noted that in the range of energy between 3.375 eV and 3.425 eV, which represent the reflectivity pic energy range, both distributions are accurate with the experimental reflectivity curves. But from 3.275 eV to 3.375 eV representing the range of transparency zone, we observed that the Gaussian distribution was more accurate than the Lorentzian distribution. We can explained this latter by the fact that, because of the frequencies, the Lorentzian distribution does not take account certain phenomenon when the frequencies (ω) are lower than the resonance frequency (ω_0). The comparison $\omega \ll \omega_0$ represents the range of transparence, a reason why in Figure 3 and Figure 4 we did not observe an accuracy between the experimental and theoretical reflectivity curves by the Lorentzian distribution for the excitons A, B and C in this range of frequency. Elsewhere, others authors already used [42] the Gaussian distribution to fit the emission spectra of the A-band ZnO, which related to the presence of acceptor impurities through different radiative mechanisms like donor-acceptor pair, a free bound transition or an exciton bound to a defect. All these facts and observations allowed us to work, in this paper, with a Gaussian instead of a Lorentzian distribution.

3. Results and Discussion

The reflectivity spectrum informs more about the free excitons peaks positions than the photoluminescence. Because of the pre-eminence of the bound excitons B. Lo and al. [7] observed in their experiments of reflectivity, in the case where $E \perp c$ polarization, two free excitons called A and B at energy positions E_A=3.398 eV and E_B=3.410 eV where their equivalences in frequencies are respectively $\omega_A = 0.516 \times 10^{14}$ s^{-1} and $\omega_B = 0.518 \times 10^{14}$ s^{-1}. Authors [7] used the theory of the spatial resonance dispersion Hopfield model [32] with the relative dielectric constant defined by:

$$\varepsilon_r = 1 + \frac{4\pi N e_0^2}{m^*} \left[\frac{\omega_0^2 - \omega^2}{\left(\omega_0^2 - \omega^2 \right)^2 - \Gamma^2 \omega^2} \right] \tag{2}$$

and

$$R = \frac{(n-1)^2 + k_0}{(n+1)^2 + k_0} \tag{3}$$

to fit the free excitons A, B (cf. Figure 1). Where the coefficient $4\pi N e_0^2 / m^*$ is related to the oscillator strength (α), "ω_0" is the resonance frequency, $e_0 = e/4\pi\varepsilon_0$ with "e" designated as the elementary charge electron, "R" the reflection coefficient, "n" the index of the medium and "k_0" the extinction coefficient. In the present study, the fact that we observe in the reflectivity a succession of curves which have the behavior of a Gaussian, allowed us to make a program using the Gaussian distribution. This latter, applied to the "bracketing" method [36] allowed us to model the reflectivity curves found experimentally by authors [7]. The results obtained by simulation are summarized in the table 1 (cf. Appendix) compared to the results found by the authors [7] (table 2, cf. Appendix). A comparison of the tables show that the Gaussian distribution (c. f. Figure 1 and Figure 2) is more accurate than the model used by the authors [7, 41] to fit the exciton A, B. We insist 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, these parameters have been used by the authors [7] to have their best fit of the free excitons. Our method, with the Gaussian distribution, allowed us, furthermore the excitons A, B (cf. Figure 1), to fit the free exciton C shown in the Figure 2. The parameters used to fit the free exciton C are summarized in Table 1 (cf. Appendix). Unless the others models [41, 45, 46, 47, 48], which are limited on the investigation of the c-plane oriented ZnO, our method with the Gaussian distribution, moreover c-plane oriented ZnO, allowed us to investigate the structure of the a-plane oriented ZnO and we fitted in the same time the excitons A, B and C something which we did not see in the literature with the others models.

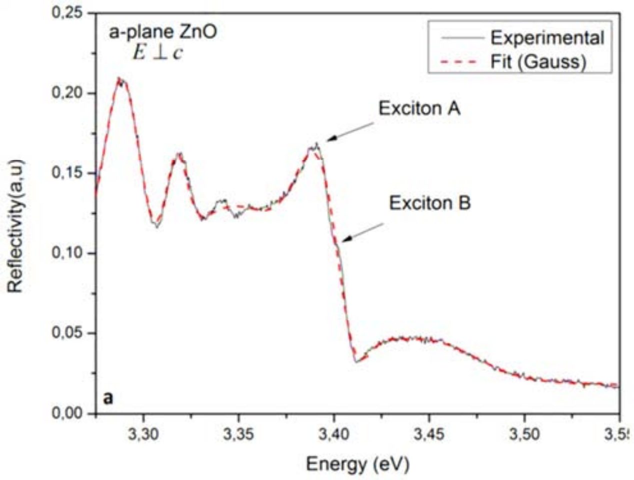

Figure 1. *Experimental and Theoretical (Gaussian distribution) reflectivity of exciton A and B.*

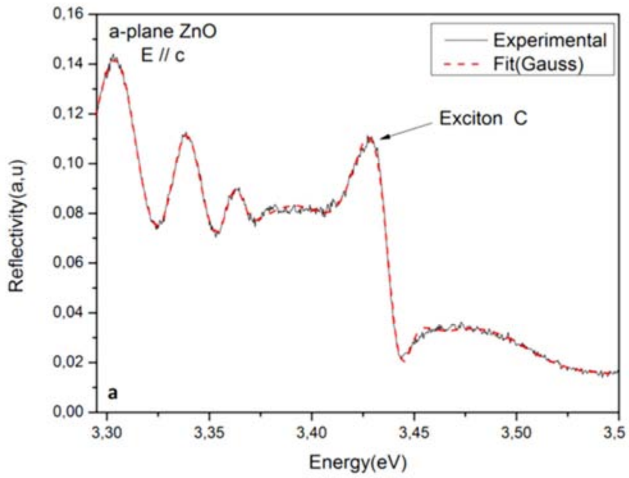

Figure 2. *Experimental and Theoretical (Gaussian distribution) reflectivity of exciton C.*

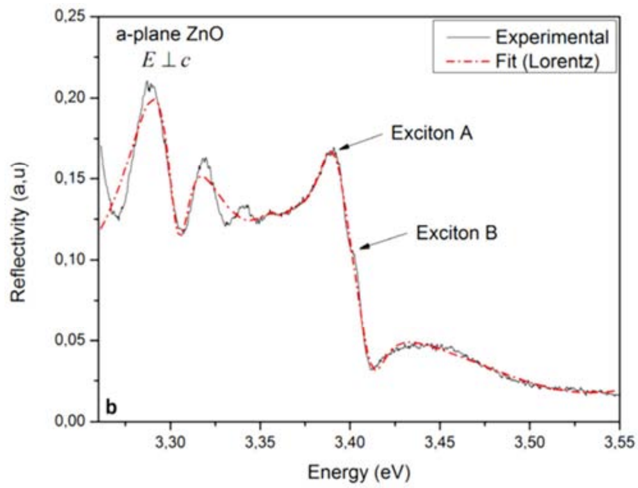

Figure 3. *Experimental and Theoretical (Lorentzian distribution) reflectivity of exciton A and B.*

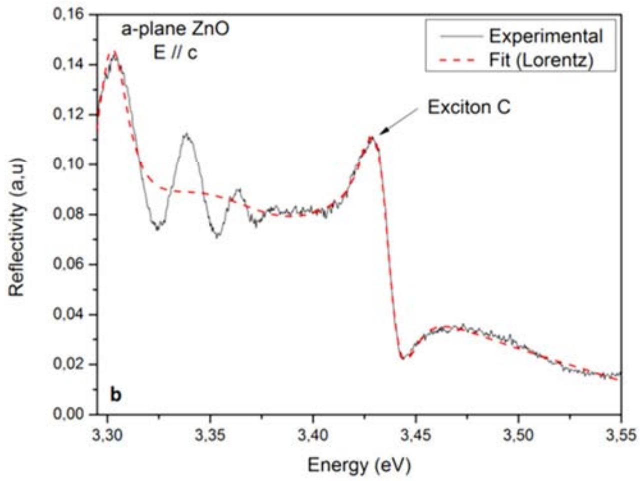

Figure 4. *Experimental and Theoretical (Lorentzian distribution) reflectivity of exciton C.*

4. Conclusion

In summary, we investigated the reflectivity of a-plane oriented ZnO thin film. Our model based on the Gaussian distribution allowed us to fit the free excitons A, B and C in the range of reflectivity pic energy and even in the transparency zone. We compared our model with those used by the authors [41, 45, 46, 47, 48] and we validated our model with the experimental results in the ref [7]. The interest of the obtained results is that they can be applied to semi-conductors which have the same structural properties such as GaN, CdS, α-SiC etc. to model and investigate the optical properties and even other physical properties [49]. Our next perspective is to investigate the diluted magnetic system type ZnO for the understanding of their magnetics [50] properties.

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Appendix

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	Frequency (ω_0) (s^{-1})	Spectral widening (Γ) (m. s^{-1})	Effective mass (m*)	Area
A	0.515E14 (E=3.389eV)	3.251E12 (Γ=2.14meV)	$0.59m_0$	2.2E-4
B	0.516E14 (E=3.401eV)	3.996E14 (Γ=26.3meV)	$0.59m_0$	1.12E-4
C	0.521E14 (E=3.430eV)	2.53E10 (Γ=1.66meV)	$0.59m_0$	1.3E-4

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 [7].*

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

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