Optical properties of multilayer (III–V semiconductors) thin films within the framework of DFT + characteristic matrix

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We present a new model of calculations based on DFT and Abeles matrix theory to understand the optical properties of semiconductor thin films which form the active layers in many optoelectronic applications. We show how optical quantities such as refractive index, extinction coefficient and band gap can change the optical representations of thin films. In this study two main types of calculations are carried out. The first is the calculation of the dielectric functions within the density functional theory framework (DFT). The second is the calculation of the optical transmittance, absorbance and reflection of monolayer and multilayer thin films within the Abeles matrix theory. The output of this combination (DFT + characteristic matrix) is very helpful and the effect of many parameters like thickness, incidence angle and light polarization on optical properties of multilayer systems can be easily investigated.

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1. Introduction

Wide band gap group-III nitrides, gallium nitride (GaN) and aluminum nitride (AlN), have attracted much attention for optoelectronic devices in the green to ultraviolet region [1–3]. The optical quantities of pure and alloy compositions (AlxGa1−xN) and the variations of these quantities with temperature or other parameters have been reported in different studies [3–11]. Since AlN and GaN are wide band-gap materials, it is expected that monolayer and double-layer of them (AlN/GaN) has high transparency and low absorption in the visible to infrared region. Such behavior is not expected for indium antimonide (InSb), because of its narrow band-gap. So, when a InSb thin film joins to AlN and GaN thin films in order to make multilayers such as AlN/InSb, GaN/InSb and AlN/GaN/InSb, considerable changes in optical representations are expected.

Multilayer thin films have numerous applications in various fields such as antireflection coatings, high reflection coatings, optical filters, solar cells technology, etc. But investigating the effect of refractive index, extinction coefficient and energy gap on optical representations of monolayer and multilayer systems was our main goal in this study.

The refractive index and energy gap of semiconductors represent two fundamental physical aspects that characterize their optical and electronic properties. So, as a first step the dielectric function of materials were calculated using the full potential linearized augmented plane wave (FP-LAPW) method with generalized gradient approximation (GGA) of Perdew–Burke–Ernzerhof [12], for the exchange correlation potential, within the density functional theory framework [13]. Calculations were developed within the framework of Abeles matrix theory in order to simulate the optical transmission, absorption and reflection of multilayer systems. Since the extraction of exact results within the super-cell method requires spending a lot of time, our model can be considered as a useful method. The framework of this study is very flexible and the effect of many parameters like thicknesses, arrangement of layers, incidence angle and light polarization on optical properties can be easily investigated.

The outline of the rest of the paper is organized as follows. In the next section the calculation method is discussed. A brief discussion about the results for optical properties such as transmission, absorption and reflection of monolayer and multilayer systems is given in Section 3. Section 4 presents a brief summary and conclusions of this study.

2. Calculation method

The calculations of the optical properties were performed in two steps. First, we calculated the imaginary part of the dielectric function of semiconductors using the scalar relativistic
full potential linearized augmented plane wave (FP-LAPW) method within the framework of density functional theory using WIEN2K package [14]. The generalized gradient approximation (GGA) of Perdew–Burke–Ernzerhof [12] was used for the exchange-correlation potential. Basis functions are expanded in combinations of spherical harmonic functions inside non-overlapping spheres at the atomic sites (muffin-tin spheres) and in plane waves in the interstitial regions. Inside the muffin-tin spheres of radius \(R_{\text{MT}}\), the \(l\)-expansion of the wave function were carried out up to \(l_{\text{max}} = 10\). The convergence parameter \(R_{\text{Kmax}}\), which controls the size of the basis sets in these calculation, was set to 8. The \(C_{\text{max}}\) parameter was taken to be 14.0 Bohr\(^{-1}\). The number of \(k\) points in the irreducible Brillouin zone was 165. The muffin-tin radii for Al, Ga, N\(_{\text{AIN}}\), N\(_{\text{GaN}}\), In and Sb were taken to be 1.78, 2, 1.78, 1.4, 2.1 and 2.2 a.u., respectively. Our materials in the study were in zincblende phase. In the second step we calculated the optical reflectance, transmittance and absorbance of multilayer systems within the Abee matrix theory [15–17]. In the following we present a brief summary of our calculations and mathematical relations.

The optical properties of matter is described by the transverse dielectric function \(\varepsilon(\omega)\). The imaginary part of dielectric function is represented by \(\varepsilon_2(\omega)\) and the real part of dielectric function is represented by \(\varepsilon_1(\omega)\). The imaginary part of dielectric function is calculated as follows:

\[
\varepsilon_2(\omega) = \frac{V e^2}{2 \pi \hbar n_m \omega} \int d^3k \sum_{n,n'} |\langle kn | p | kn' \rangle|^2 f(kn) \times (1 - f(kn')) \delta(E_{kn} - E_{kn'} - \hbar \omega)
\]

(1)

where the sum is over all possible transitions from occupied to the unoccupied states, \(\hbar \omega\) is the energy of the incident photon, \(p\) is the momentum operator \(i \hbar \partial/\partial x\), \(|kn\rangle\) is the eigenfunction with eigenvalue \(E_{kn}\), and \(f(kn)\) is the Fermi distribution function. The evaluation of the matrix elements of the momentum operator in Eq. (1) is done over the muffin tin and the interstitial regions separately [18,19]. Generally, the elements of imaginary dielectric tensor are not equal and we used the average value of \(\varepsilon_2(\omega)\):

\[
\varepsilon_2(\omega) = \varepsilon_{2xx}(\omega) + \varepsilon_{2yy}(\omega) + \varepsilon_{2zz}(\omega)
\]

(2)

When the imaginary tensor calculated using DFT theory, the real part of dielectric function \(|\varepsilon(\omega)|\) can be determined from the imaginary part by using the Kramers–Kronig relation [20,21]. Since the real part is also a tensor, the mean value of it was used again.

The refractive index \((n)\) and extinction coefficient \((k)\) of materials were calculated by the following relations:

\[
n = \left(\frac{\varepsilon_1^2 + \varepsilon_2^2}{2} + \varepsilon_1\right)^{1/2}
\]

(3)

\[
k = \left(\frac{\varepsilon_1^2 + \varepsilon_2^2}{2} - \varepsilon_1\right)^{1/2}
\]

(4)

The complex refractive index of \(j\)th layer is given by

\[
\tilde{n}_j = n_j + ik_j
\]

(5)

The incident angle of light in \(j\)th layer \((\theta_j)\) can be determined by the Snell’s law:

\[
\tilde{n}_j \sin \theta_j = \tilde{n}_0 \sin \theta_0
\]

(6)

In the above relation, \(\tilde{n}_0\) and \(\theta_0\) are the refractive index and incident angle in vacuum, respectively. Phase shift due to \(j\)th layer \((\delta_j)\) is then given by

\[
\delta_j = \frac{2\pi \tilde{n}_j d_j \cos \theta_j}{\lambda}
\]

(7)

In the relation (7), \(d_j\) is the thickness of \(j\)th layer and \(\lambda\) is the light beam wavelength.

The impedance of \(j\)th layer for \(s\)-polarization and \(p\)-polarization is defined by

\[
\eta_j = \sqrt{\frac{\varepsilon_0}{\mu_0}} \tilde{n}_j \cos \theta_j \quad \text{(for } s\text{-polarization)},
\]

\[
\eta_j = \sqrt{\frac{\varepsilon_0}{\mu_0}} \tilde{n}_j \cos \theta_j \quad \text{(for } p\text{-polarization)}
\]

(8)

In the relation (8), \(\varepsilon_0\) is the permittivity of free space and \(\mu_0\) is the permeability of free space.

In the multilayer system, the idea of matching the electric \(E\) and magnetic \(H\) fields of the incident light at the interfaces of layers yield the matrix relation:

\[
[\begin{array}{c}
B \\
C
\end{array}] = \prod_{j=1}^{N} \left[\begin{array}{c}
\cos \delta_j - i \sin \delta_j \\
\eta_j \sin \delta_j \cos \delta_j
\end{array}\right] \left[\begin{array}{c}1 \\
\eta_{0j}\end{array}\right]
\]

(9)

The \(2 \times 2\) matrix on the right hand side of this equation is known as the characteristic matrix of \(j\) thin film and the \(2 \times 1\) matrix on the left hand side of this equation is known as the characteristic matrix of assembly. \(B\) and \(C\) are total electric and magnetic field amplitudes of the propagating light in the medium. \(\eta_{0j}\) is the impedance of the substrate and \(\eta_j\) is the impedance of vacuum.

Optical transmittance \((T)\), absorbance \((A)\) and reflectance \((R)\) can be derived by the following relations:

\[
T = \frac{4\tilde{n}_0 \operatorname{real} (\eta_{0j})}{(\eta_0B + C)(\eta_0B + C)}
\]

(10)

\[
A = \frac{4\tilde{n}_0 \operatorname{real} (BC^* - \eta_{0j})}{(\eta_0B + C)(\eta_0B + C)}
\]

(11)

\[
R = \left(\frac{\eta_0B - C}{\eta_0B + C}\right)^2 = 1 - T - A
\]

(12)

3. Result and discussion

Calculated imaginary and real parts of dielectric function of our materials are shown in Figs. 1 and 2, respectively. Unlike to other cases, the curve of InSb begins to grow at low energies and three main peaks that centered around 1.7 eV, 3.5 eV and 4.7 eV are seen.
The AlN and GaN imaginary curves have negligible values at energies under 5 eV and 3 eV, respectively. So, it is expected that AlN and GaN both have a negligible absorption within the visible and infrared wavelength ranges while InSb has a considerable absorption in these regions. Fig. 2 shows that AlN and GaN real curves behave in a similar way at energies under 8 eV but the InSb curve has considerable variations and reaches negative values at energies above 3.5 eV.

The values of refractive index ($n$) and extinction coefficient ($k$) of AlN, GaN and InSb are represented in Figs. 3 and 4, respectively. Fig. 3 shows that AlN and GaN refractive index curves have many similarities and their values are situated between 2 and 2.5 but the InSb refractive index curve has considerable variations and several small peaks that centered around 165 nm, 250 nm, 370 nm and 830 nm are seen. As indicated in Fig. 4 AlN and GaN extinction coefficient curves are similar at wavelengths above 150 nm but their values decrease with wavelength. The extinction coefficient values for AlN and GaN are negligible at wavelengths above 250 nm and 376 nm, respectively. The InSb extinction coefficient values increases with wavelength within the range of 150–332 nm and several small peaks around 162 nm, 250 nm, 332 nm and 650 nm can be seen.

The transmittance spectra of AlN, GaN and InSb monolayer thin films are represented in Figs. 5–7, respectively. As can be seen from these figures, since AlN and GaN are wide band gap materials, they have high transparency in the visible and infrared regions and there is a sharp fall in transmission near the fundamental absorption, but InSb is a narrow band gap material and has low transparency, even at small thicknesses. Fig. 5 shows that thin films of AlN exhibit high transmittance at higher wavelengths ($\geq 300$ nm). This figure also shows that in the visible to infrared region the curves are at high levels of transparency but in the ultraviolet region reduce to
Fig. 8. Transmittance of double layer AlN(900 nm)/GaN thin films at different thicknesses of GaN.

Fig. 9. Transmittance for double layers of AlN(900 nm)/InSb thin films at different thicknesses of InSb.

Fig. 10. Transmittance for double layer of AlN(400 nm)/GaN(40 nm) thin film at different degrees.

Fig. 11. Transmittance of double layer AlN(400 nm)/GaN(50 nm) for S and P polarizations when the incidence angle is 30°.

Fig. 12. Absorbance of InSb(1000 nm) monolayer thin film, AlN(400 nm)/GaN(100 nm) double layer thin film and AlN(400 nm)/GaN(100 nm)/InSb(1000 nm) threefold thin film.

extremely low levels and the thicknesses of layer enhances this reduction. Similar behavior is reported in Fig. 4(a) of reference [23]. Fig. 6 shows that GaN thin films can represent high transmittance within the wavelengths range of 450–1000 nm but the transmission in the ultraviolet region decreases with thickness of layers. Figs. 5 and 6 have similar patterns but there is a sharper incline in AlN curves, because of its wider band gap. Fig. 7 shows that for thicknesses above 50 nm, the transmission of InSb layers in the visible region is negligible but for thickness under 50 nm, the transmission property can display its role even in the ultraviolet region. Generally, at low wavelengths, the transmittance reduces with thickness of thin films. This result is in agreement with our physical intuition and those reported about II–VI semiconductors [23,24]. On the other hand, these figures show that the optical energy gap decreases with thickness of layers. Such patterns of optical transmittance curves are reported experimentally for materials such as InSb, ZnO and GaAs [25–27]. It should be noted in order to check the reliability of our results due to the lack of empirical reports, we have used the similar reports of other materials.

We also have reported the transmittance spectra for AlN/GaN and AlN/InSb double layer thin films in Figs. 8 and 9, respectively. Fig. 8 shows that AlN/GaN double layers can represent high levels of transparency for wavelengths within the range of 400–1000 nm but transmittance in the ultraviolet region decreases with thickness of layers. Fig. 9 shows that when an AlN thin film joins to a InSb thin film, the transmittance decreases considerably. For example the transmittance in the visible region decreases from 90% in AlN monolayer to 20% in AlN/InSb double layer (here, the thickness of InSb layer is only 20 nm). The transmittance of AlN/InSb double layer in the ultraviolet region is also negligible when the thickness of InSb layer is above 40 nm (here, the thickness of AlN layer is 900 nm).

The effect of incidence angle and light polarization on the transmittance of double layers of AlN/GaN is reported in Figs. 10 and 11, respectively. Fig. 10 shows that as the angle of incidence increases, the transmittance decreases. As indicated in Fig. 11 the optical transmittance for p polarization is higher than s polarization. Such patterns can be seen in Refs. [17,28,29].

In the following, we report the absorption spectra for three different kinds of thin films in Fig. 12. This figure clearly shows that InSb monolayer has about 50% absorption in ultraviolet, visible and infrared regions but the absorption of twofold AlN/GaN thin film in the visible to infrared region is negligible. This figure also shows, when a narrow band gap thin film like InSb joins to a wide band gap twofold thin film like AlN/GaN, the absorption increases not only in the ultraviolet but also in the visible and infrared regions.
(here, over 70%). This effect can be considered in the field of solar cells designing. Such behavior can be seen for other materials in Ref. [17].

4. Conclusions

This study has a training aspect that investigate the effect of band gap, refractive index and extinction coefficient on the optical properties of multilayer thin films. This study is based on combination of first principles calculations and Abeles matrix theory. We studied the optical properties of three different materials of III-V semiconductors using FP-LAPW method. We have shown that AlN and GaN thin films have high transmission and low absorption even at large thicknesses because of their wide band gap and negligible extinction coefficients at wavelengths above 400 nm. But InSb have low transmission and high absorption even at small thicknesses because of its narrow band gap and nonzero extinction coefficient in the ultraviolet, visible and infrared regions. We also examined the joint states of these materials. We have shown that when a low band gap material such as InSb is joint to a large band gap thin film such as AlN/GaN, the absorption increases and the transmission decreases considerably. This effect can be considered in the field of solar cell designing. The effect of light polarization and incident angle was investigated, too. We have shown that the light transmittance for p-polarization is greater than s-polarization. We also, have shown that the transmittance decreases with incident angle. Framework of our approach is very flexible and the influence of many parameters can be easily investigated.

References