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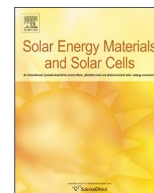

Refractive indexes and extinction coefficients of n- and p-type doped GaInP, AlInP and AlGaInP for multijunction solar cells

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ABSTRACT

The optical properties of p-type, n-type and nominally undoped $(Al_xGa_{1-x})_yIn_{1-y}P$ layers have been determined in a wide spectral range. The layers under study have been chosen with compositions and dopant concentrations which make them interesting for their use in III-V multijunction solar cells. The layers have been measured by variable angle spectroscopic ellipsometry and, irrespective of composition and doping, their optical response has been modelled using the same model dielectric function consisting of two asymmetric Tauc-Lorentz oscillators and a 3D-M_0 Adachi term. The results show that transition energy values change with layer composition, whilst for layers of the same material (i.e. GaInP or AlInP), the band-gap transition energy E_0 shows a strong dependence on the order parameter. The refractive indexes and extinction coefficients deduced from the ellipsometric data have been used to fit reflectance measurements for the same layers and an excellent agreement has been achieved, thus validating the model dielectric function proposed for this kind of materials.

1. Introduction

Ternary and quaternary III-V alloys such as $Ga_xIn_{1-x}P$, $Al_xIn_{1-x}P$ and $(Al_xGa_{1-x})_yIn_{1-y}P$ are widely used in many optoelectronic devices. In particular, $Ga_xIn_{1-x}P$ is a key material for the top cell in triple junction solar cells [1]. $Al_xIn_{1-x}P$ and $(Al_xGa_{1-x})_yIn_{1-y}P$ have emerged as potential candidates with bandgaps above 2.0 eV that can be used as top cells in multijunction solar cells with more than 3 junctions [2–5]. Moreover, $Al_xIn_{1-x}P$, $(Al_xGa_{1-x})_yIn_{1-y}P$ and $Ga_xIn_{1-x}P$ are usually employed as passivating and barrier layers in III-V solar cells, namely, front/back surface fields and buffer or nucleation layers with thicknesses ranging from tenths to hundreds of nanometers and up to the micrometer range (i.e. as in compositionally graded buffer layers in metamorphic structures). Even in cases where they act as photovoltaic inactive layers, their impact on light transmission to the underlying layers is not negligible. Therefore, the knowledge of the optical properties of $Ga_xIn_{1-x}P$, $Al_xIn_{1-x}P$ and $(Al_xGa_{1-x})_yIn_{1-y}P$ is of paramount importance for the development of III-V multijunction solar cells. In fact, analytical models [6] and drift-diffusion simulators typically used to predict solar cell performance require reliable optical constants (wavelength dependent refractive index and absorption coefficient) of the materials to improve their accuracy [7].

The optical behaviour of lattice matched $Ga_xIn_{1-x}P$ and the quaternary $(Al_xGa_{1-x})_yIn_{1-y}P$ films has been extensively studied using a variety of experimental techniques [8–16]. However, very few of these works have correlated the material optical properties with dopant type and level [14,15]. $Al_xIn_{1-x}P$ has attracted much less attention from the scientific community, although excellent reports have been published in the last years [17–19]. More recently, the study of the optical properties of lattice mismatched III-V alloys has been addressed, thus extending the compositional range for which data are available [20]. It is well known that the electrical and optical properties of solar cells are dependent on the growth conditions (temperature, V/III ratio, growth rate, etc.), doping level and dopant type. Growth conditions may induce different degrees of CuPt-type ordering on the group III-sublattice of the (Al)GaInP alloys [21]. In essence, CuPt-type ordering describes a superlattice of alternating Ga- and In-rich {111} planes (i.e., for the case of GaInP, $Ga_{0.5(1-\eta)}In_{0.5(1+\eta)}P/Ga_{0.5(1+\eta)}In_{0.5(1-\eta)}P$, where η is defined as the order parameter). For a completely disordered material $\eta = 0$, whereas for a completely ordered material $\eta = 1$, although, so far, only experimental values as high as $\eta = 0.6$ have been reported for GaInP grown using Metal Organic Vapour Phase Epitaxy (MOVPE) [22,23]. The degree of order modulates, among others, the energy bandgap (E_g) of the alloy [14,21,24,25]. In this respect, a decrease in the order parameter (low η) leads to a higher E_g.

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In spite of the extensive literature in the field, most of the existing works have studied $Ga_xIn_{1-x}P$, $Al_xIn_{1-x}P$ and $(Al_xGa_{1-x})_yIn_{1-y}P$ layers with an intrinsic doping which does not correspond to the typical doping values (and thus degree of disorder) used in conventional III-V solar cells. Therefore, the determination of the optical constants of III-V phosphide-based materials with the typical doping levels and growth conditions applied in the fabrication of multijunction solar cells is essential to improve the accuracy of the models used to predict the solar cell performance.

Accordingly, the purpose of this paper is to determine the optical constants (refractive index and absorption coefficient) by variable angle Spectroscopic Ellipsometry (SE) of $Ga_{0.51}In_{0.49}P$, $Al_{0.53}In_{0.47}P$ and $(Al_xGa_{1-x})_{0.51}In_{0.49}P$ lattice matched to GaAs (hereafter referred to as GaInP, AlInP and AlGaInP, respectively) for different doping levels and dopant types – and subsequent order parameter – as the ones typically used for III-V solar cells. It is known that CuPt-type ordering induces a symmetry lowering and therefore a small birefringence in the layer dielectric function in the near-bandgap spectral region [26]. However, such a fine analysis is beyond the scope of this article and anisotropy was not taken into account in the data treatment to calculate the optical constants as previous works proved the suitability of the isotropic approximation for similar III-V layers [9,14,15,17,18].

Complementary characterization techniques such as X-ray diffraction (XRD), Raman spectroscopy, Scanning Electron Microscopy (SEM), Photoluminescence (PL), are used to determine thicknesses, order parameter and composition of the layers which are required to carry out the analysis. The comparison of the reflectance obtained from the deduced optical constants with experimental data along a wide spectral range, is used to validate the model describing the optical behaviour of the layers. The results of the paper are also useful for devices other than solar cells, such as LEDs, lasers, etc and will contribute to extend knowledge on the optical properties of the aforementioned materials.

2. Experimental

For this study, a batch of GaInP and AlInP structures, plus a AlGaInP one, were epitaxially grown on (100) GaAs wafers misoriented 2° towards the nearest (111)A plane in a low pressure horizontal Aixtron MOVPE reactor (AIX-200/4). Trimethyl-gallium (TMGa), trimethyl-indium (TMIn), trimethyl-aluminium (TMAl), phosphine (PH_3) and arsine (AsH_3) were the sources used for the GaAs, GaInP and AlInP layers. Dimetylzinc (DMZn) and ditertiarybutylsilane (DTBSi) were used as dopant sources. Growing conditions were chosen in order to get layers with the typical doping levels and compositions actually used in III-V multijunction solar cells. Two GaAs layers were grown in such a way that they sandwiched the GaInP, AlInP or AlGaInP layer that will be the object of this study. A buffer GaAs layer, 500–1000 nm thick, is grown on the substrate before the phosphide layers while a thin GaAs cap layer finishes the stack to protect the phosphide layers that will be analysed in this paper. Prior to the optical characterization, the cap layer was chemically etched using NH_4OH:H_2O_2:H_2O (2:1:10) (see Fig. 1).

The thickness of the layers of interest was measured by in situ reflectometry during the epitaxial growth and confirmed by SEM once the layer was grown. The reflectometry tool is an EpiRAS 2000 instrument from Laytec. SEM equipment is a FEI Helios Nanolab 650 Dual Beam microscope. The dopant concentration in the layers was measured by electrochemical capacitance–voltage (CV) profiling using a WEP Control CVP21 tool. The mismatch between the layer of interest and the substrate was analysed by a QC2a X-Ray Diffractometer Bede Scientific XRD.

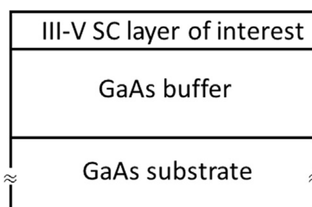

Fig. 1. Sketch of the semiconductor structure after chemical etching of the cap layer, prior to the optical characterization.

PL measurements were performed at 20 K excited with a 532 nm laser at 26 mW. Raman spectroscopy measurements were conducted to determine differences in the ordering parameter among layers with the same composition. The equipment is a Raman Confocal Microscope Invia Reflex from RENISHAW, using Ar 514.5 nm (12 mW) as excitation line. Spectral resolution achieved was 1 cm^{-1} by selecting a 1800 l/mm grating and a slit opening of 50 µm. Spectra were acquired in backscattering geometry at room temperature. The dielectric response of the layers has been characterized in a Semilab GES-5E variable angle spectroscopic ellipsometer in the region from 233 to 984 nm (approximately 5.3–1.26 eV) at room temperature. The layers were analysed with a microspot configuration with a spot size of approximately 400 µm and angular aperture of 4.25°, the angle of incidence was varied between 60° and 75°. Specular reflectance was performed on a Perkin Elmer Lambda 1050 UV/VIS/NIR WB InGaAs Spectrometer with defined angle of incidence and reflection of 6°. Reflectivity simulations have been performed by using the generalized matrix method [27]. The structure used in the simulations is exactly the one depicted in Fig. 1. An incoherent optical behaviour was assumed within the substrate due to its large thickness (350 ± 25 µm) while the other layers were treated coherently. No roughness was assumed in any layer due to the specular behaviour of the samples.

3. Results

The basic characterization results of the nine layers grown for this study have been gathered in Table 1. For each type of sample, this table includes the doping type, doping concentration and layer thickness as measured in situ with EpiRAS, and *ex-situ* through SEM. Some other structural characteristics of the layers have also been included. Fig. 2 shows an example of a SEM cross-section image. As can be seen, the GaAs buffer layer is slightly recognizable, while an abrupt interface with the layer under study is clearly visible for all the structures.

3.1. Structural characterization

XRD measurements were performed in order to obtain the lattice mismatch between the semiconductor layers (GaInP, AlInP) and the GaAs substrate. The determination of their lattice constants can be calculated by:

$$a[Ga_xIn_{(1-x)}P] = xa[GaP] + (1-x)a[InP] \tag{1}$$

where aGaP and aInP are the lattice constants of GaP and InP. For x = 0.516 = X_{LM} (lattice matched) at 25 °C, the GaInP layer has a lattice constant of 0.56532 nm and it will be lattice matched to the GaAs substrate [28]. The lattice mismatch is calculated by:

$$a\frac{[GaInP] - a[GaAs]}{a[GaAs]}\% \tag{2}$$

As can be seen in Table 1, most of the layers are lattice matched to GaAs, just the undoped GaInP layer shows a slight mismatch in agreement with [28]. For the AlGaInP layer, the peak shift to that of the GaAs substrate (Δθ = 80″) shows again a good lattice matching, but due to complexity of lattice mismatch calculations for quaternaries elements, only a rough estimate of the Al quantity around 10–20% (i.e. $(Al_{0.15}Ga_{0.85})_{0.51}In_{0.49}P$) can be made.

The estimation of the order parameter for the GaInP layers was performed, as in previous works, from PL measurements at 20 K [29,30]. The relation between the order parameter, η, and the energy bandgap (i.e. peak energy from low temperature PL measurements) is described by:

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