Physica B 456 (2015) 66–71

Contents lists available at ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Macroscopic polarization and bowing constant of $Al_xGa_{1-x}N$

A. Pansari, V. Gedam, B.K. Sahoo *

Department of Physics, National Institute of Technology, Raipur-492010, Chhattisgarh, India

ARTICLE INFO

Article history:
Received 12 May 2014
Received in revised form
30 July 2014
Accepted 10 August 2014
Available online 19 August 2014

Keywords:
$Al_xGa_{1-x}N$ alloy
Piezoelectricity
Phonons
Bowing constants
Thermoelectric properties
Computer simulations

ABSTRACT

In this work, we have theoretically investigated the effect of macroscopic polarization (sum of spontaneous and piezoelectric polarization) on various thermal parameters of $Al_xGa_{1-x}N$ alloy. The macroscopic polarization contributes to the effective elastic constant of $Al_xGa_{1-x}N$ alloy. This contribution modifies the phonon group velocity, Debye temperature and Debye frequency of the alloy. The revised values of these thermal parameters have been estimated as a function of Al composition. Our calculation shows that these thermal parameters are enhanced and vary non-linearly with Al composition i.e., they show bowing. The cause of this bowing is the non linear dependence of spontaneous and piezoelectric polarization on Al composition. The bowing constant of above thermal parameters (with and without polarization) has been theoretically predicted by method of best fit. The results show that polarization mechanism not only enhances the parameters but also contribute significantly to bowing constant. The macroscopic polarization contributes around 48% to bowing constant of above thermal parameters. The obtained result will be useful for simulation of nitride optoelectronics devices to predict the effect of polarization mechanism on thermoelectric properties of $Al_xGa_{1-x}N$ alloy.

1. Introduction

Group III–V nitrides are currently the most promising materials for blue, green, and UV optoelectronics. Their applications are diverse and include biosensors, medical imaging, optical data storage, multimedia, etc. One of the important methods of controlling properties of these materials relies on alloying instead of employing pure binaries An, GaN etc [1]. By adjusting the composition, one can tune selected material parameters such as band gap, lattice constant or polarization to the desired optimal value. Recently, $Al_xGa_{1-x}N$ alloys, particularly Al-rich AlGaN alloy have attracted much interest due to their applications in solid-state UV light sources for bio agent detection as well as for general lighting. The AlGaN/GaN high electron mobility transistors (HEMTs), have emerged as attractive candidates for high voltage, high power operation at microwave frequencies [2]. The material possesses a number of superior properties such as larger peak electron velocity, larger saturation velocity, higher breakdown voltage and higher thermal stability, making this material very suitable for use as channel material in microwave power devices.

Macroscopic polarization is a unique property of nitrides. Its existence in III–V nitrides has been confirmed by both first-principle calculations and experiments [3–12]. This effect induces sheet charge density and electric field as high as 1 MV/cm and plays a significant role in influencing optical, thermal and electrical properties of nitrides and thus, is a controlling factor in increasing carrier concentration and surface recombination. These fields significantly influence the distribution and lifetime of excess carriers in AlGaN QWs and can have an important impact on the performance of AlGaN/GaN based optoelectronic devices [13]. Although these devices, have already reached outstanding performance further improvements of design and efficiency seems to be possible only on the basis of a more detailed understanding of polarization effect in group III nitride alloy nanostructures.

In our earlier works [14,15], we have investigated the effect of macroscopic polarization on various physical parameters and thermal properties of binary nitrides. On the basis of these results, one can expect equivalent enhancement in various physical parameters and thermal conductivity of nitride alloys due to polarization mechanism also. To the best of authors' knowledge, macroscopic polarization effect on various physical parameters of $Al_xGa_{1-x}N$ alloy, particularly on thermal parameters has not been investigated. The composition dependence nature of lattice constant and band gap energy of $Al_xGa_{1-x}N$ alloy have been studied by first principle calculations [16]. This calculation has predicted a finite amount of bowing (composition dependent nonlinear nature) in lattice constant and band gap energy. The bowing constant value of thermal parameters such as phonon velocity and Debye temperature are yet to be investigated. The significance is that once bowing constant is known, the physical parameter, in principle, can be predicted for

* Corresponding author. Tel.: +91 9589186713; fax: +91 771 2254600.
E-mail address: bksahoo.phy@nitrr.ac.in (B.K. Sahoo).

http://dx.doi.org/10.1016/j.physb.2014.08.008
0921-4526/

whole Al composition. For accurate prediction of optical and thermal properties of $Al_xGa_{1-x}N$ alloy, bowing constant of elastic constant, phonon velocity and Debye temperature with and without polarization mechanism are required [17]. As thermal conduction controls the design, performance and long life of the device, a study of polarization effect on thermal parameters and on their bowing nature deserves attention. The obtained result will be useful to study the effect of macroscopic polarization mechanism on thermoelectric properties of $Al_xGa_{1-x}N$ alloy [18].

2. The model

The macroscopic polarization P is composed of two parts: (i) spontaneous polarization,P^{sp} and (ii) piezoelectric polarization P^{pz}. Spontaneous polarization is an intrinsic property of the material originates due to crystal structure asymmetry. During the growth of nitride film on a substrate, strain is generated due to lattice mismatch between film and substrate. This strain produces piezoelectric polarization in the film. The spontaneous and piezoelectric polarization fields are composition dependant properties [6,7]. The elastic constants are also composition dependant parameters of the nitride alloys [19]. The composition dependence nature of elastic constants of $Al_xGa_{1-x}N$ alloy has been studied by first principle calculations [16]. The study has predicted finite degree of bowing (composition dependent nonlinear nature) in these parameters. This first principle calculation prediction has been validated by measuring surface acoustic wave velocity in $In_xGa_{1-x}N$ alloy by means of high resolution Brillouin spectroscopy [20]. Due to macroscopic polarization, the coupling between the elastic strain and electric field induces additional electric polarization [21]. This contributes the effective elastic constant of the alloy. The effective elastic constant of $Al_xGa_{1-x}N$ alloy can be written as [14,15]

$$C_{44,p}^{AlGaN} = C_{44}^{AlGaN} + [(e_{15}^2 + e_{31}^2 + e_{33}^2 + p_{sp}^2)/\varepsilon_0\varepsilon]^{AlGaN} \tag{1}$$

where $C_{44,\,p}^{AlGaN}$ is elastic constant of $Al_xGa_{1-x}N$ where polarization effect is taken into account and C_{44}^{AlGaN} is elastic constant without polarization. e_{15}, e_{31} and e_{33} are piezoelectric constants and p^{sp} is spontaneous polarization constant of $Al_xGa_{1-x}N$ alloy. ε_0 and ε are respectively, vacuum permittivity and dielectric constant of $Al_xGa_{1-x}N$ alloy. The elastic constant $C_{44,\,p}^{AlGaN}$ can be estimated once C_{44}^{AlGaN} and polarization constants of $Al_xGa_{1-x}N$ alloy are known. The elastic constant without polarization C_{44}^{AlGaN} can be calculated by Refs. [19,20]

$$C_{44}^{AlGaN} = xC_{44}^{AlN} + (1-x)C_{44}^{GaN} + b_{44}x(1-x) \tag{2}$$

where $b_{44} = -0.25$ GPa is bowing constant of C_{44}^{AlGaN}. The piezoelectric polarization constants of $Al_xGa_{1-x}N$ can be calculated by Vegard's formula [5]

$$e_{ij}^{AlGaN} = xe_{ij}^{AlN} + (1-x)e_{ij}^{GaN} \tag{3}$$

The constants e_{15}, e_{31} and e_{33} show linear variation with Al composition [5,11] whereas $p^{sp\ AlGaN}$ varies non-linearly with composition as [6]

$$p^{sp\ AlGaN}(x) = -0.090x - 0.034(1-x) + 0.021x(1-x) \tag{4}$$

The dielectric constant also show linear variation with composition which can be written as [5,6]

$$\varepsilon^{AlGaN}(x) = x\varepsilon^{AlN} + (1-x)\varepsilon^{GaN} \tag{5}$$

Now using the equations from (2) to (5) one can easily calculate $C_{44,\,p}^{AlGaN}$ as a function of In composition. In nitride crystals, the phonon velocity v depends on the direction and magnitude of the phonon wave vector. The phonon velocities along transverse directions are degenerate and have the same velocity given by $v_t^{AlGaN} = (C_{44}^{AlGaN}/\rho^{AlGaN})^{1/2}$. According to Callaway's formulation the average velocity v along a specified direction is [22]

$$(v^{AlGaN})^{-1} = \frac{1}{3}[(v_t^{-1} + v_t^{-1} + v_l^{-1})], \tag{6}$$

where $v_l = \sqrt{C_{33}^{AlGaN}/\rho^{AlGaN}}$ is the phonon velocity along longitudinal direction. The elastic constant C_{33}^{AlGaN} can be calculated by Ref. [19]

$$C_{33}^{AlGaN} = xC_{33}^{AlN} + (1-x)C_{33}^{GaN} + b_{33}x(1-x), \tag{7}$$

where $b_{33} = -1.14$ GPa is bowing constant of C_{33}^{AlGaN}. The material density ρ^{AlGaN} can be calculated by $\rho^{AlGaN}(x) = x\,\rho^{AlN} + (1-x)\,\rho^{GaN}$ [19,21,23]. It is required to calculate phonon velocity. Let us know the polarization contribution to phonon velocity. Due to the inclusion of macroscopic polarization, the transverse velocity becomes $v_{t,p}^{AlGaN} = (C_{44,p}^{AlGaN}/\rho^{AlGaN})^{1/2}$. It should be noted that longitudinal velocity does not change due to polarization effect. The average velocity v_p including polarization effect along a specified direction can be written as [22]

$$(v_p^{AlGaN})^{-1} = \frac{1}{3}[(v_{t,p}^{-1} + v_{t,p}^{-1} + v_l^{-1})] \tag{8}$$

The other two thermal parameters which are very important for the calculation of thermal conductivity and thermoelectric properties of AlGaN are Debye frequency and Debye temperature. The Debye frequency can be calculated by the relation $\omega_D = v(6\pi^2/V_o)^{1/3}$. Here, N is the number of atoms present in volume V_0 of the crystal and v is average phonon velocity. The Debye temperature θ_D which is defined by $\theta_D = \hbar\omega_D/k_\beta$, can be calculated from the knowledge of Debye frequency. Here h is Planck's constant and k_β is Boltzmann's constant. The Debye frequency and Debye temperature depend directly on phonon velocity so these parameters are revised due to macroscopic polarization effect.

3. Results and discussions

3.1. Applicability of Vegard's rule

We first discuss the applicability and deviation of Vegard's rule. The physical properties of the ternary compounds are usually investigated using Vegard's law, i.e., assuming that the lattice constants of the ternary compound can be expressed as a linear combination of the lattice constants of the two forming binary compounds. The validity of this approach is recognized among the researchers [24]. For $Al_xGa_{1-x}N$ alloy, the Vegard's law can be written as

$$P^{AlGaN}(x) = xP^{AlN} + (1-x)P^{GaN} \tag{9}$$

here, P stands for a physical parameter of $Al_xGa_{1-x}N$. The application of the Vegard's law to lattice and elastic constants of binaries has proved to be a useful tool for understanding the elastic behaviour and compositional nature of ternary and quaternary III-N semiconductor alloys [17]. In Ref. [24], this theoretical conception is supported by a solid experimental data set. However, there are serious indications that deviations from this simple rule should be expected for lattice constants, band gap energy and elastic properties [16,17,19,25], similar to nonlinear dependence on composition predicted for spontaneous and piezoelectric properties of iii–v nitrides alloys [7]. This prediction has been confirmed by experiments [6]. This non linearity is attributed essentially to sources like (i) chemical effects due to the different cation electro negativity of the components; (ii) internal strain