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Mathematical approach for bandgap energy investigation of InGaNAs/GaAs structure using the Design of experiments

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Abstract—**In the present paper, we propose a simple mathematical model which describes analytically the composition dependence of the energy gap of the following alloys: $In_xGa_{1-x}N_yAs_{1-y}$ and GaN_yAs_{1-y}grown on GaAs substrates using the design of experiments (DoE).This approach does not require the BAC or any other parameters related to $In_xGa_{1-x}N_yAs_{1-y}$. The number of necessary parameters to predict the bandgap energy of $In_xGa_{1-x}N_yAs_{1-y}$ structure is restricted just to two concentrations*x*and*y*. The obtained bandgap predictions are in a good agreement with available experimental data. The influence of these two variables and their interactions on the bandgap energy is evaluated. From the findings of this study, the influence of nitrogen on reducing the band gap energy is larger than the indium effects. The interaction between these two factors In-N and the N-N interaction leads to the increase of the bandgap while the interaction In-In hasn't any influence. This simple method to predict the band gap energy for $In_xGa_{1-x}N_yAs_{1-y}$ and GaN_yAs_{1-y}can be applied to any other alloys.**

Keywords--Semiconductor, bandgap, design of experiments (DoE)

I. INTRODUCTION

The semiconductor III-V-Nalloys, in which electronegative nitrogen substitutes group V anions in standard group III-V compounds have in recent years emerged as a subject of considerable theoretical and experimental research interest [1-6]. However, $GaN_yAs_{1-y}$and $In_xGa_{1-x}N_yAs_{1-y}$are currently the most prominent members of this new class of semiconductor alloys due to theirproperties[7-8] and their wide application in electronic and optoelectronic devices [9-12].The introduced Nforms a resonant defect level above the conduction band edge in InGaAs due to the nitrogen's differing electronegativity and size. This isoelectronic N level resonant with the Γ conduction band causes a band anticrossing effect leading to the generation of $E_-$and E_+ non parabolic conduction subbands that varies with N content [13-14]. Other consequences will occur, as:a significant reduction of the bandgap [14], the strongly increased of the electron mass compared to the host material [15], the strong localization of electron wave functions and the strong reduction of the hydrostatic pressure coefficient of the band gap [16].The bandgap can be calculated from several theoretical methods such as the band anticrossing model [17],tight binding theory [18] or pseudopotential theory [19].These methods typically involve multiple parameters and relatively complex computations that are inconvenient or not sufficiently accurate for the device's design. Also, these methods are incapable to detect interactions between the variables and their effect in the final reply (because theyrequire separate comments for each variable). These interactions can lead to effects higher or lower than those that are observed if the variable acts separately.The design of experiments (DoE) is an important technique based on the principle of possible modeling of any real specific phenomenon using statistical tools [20]. By using this methodology, it is possible to determine which variables are most influential on the response,to estimate linear interaction effects of the factors and to make a prediction model for the response. The predetermined model is a valid prediction model inside the study domain, which must always be precisely established. So, (DoE) can help an investigator to extract maximum information from a dataset. Since its appearance, (DoE) has been widely used in science, engineering, technology…etc [21]. In recent times, DOE has found new and challenging applications in nanotechnology precisely in the field of semiconductors [22-23].

In the present work, a simple mathematical model has been proposed to parameterize the gap energy of the diluteternary GaN_yAs_{1-y} andquaternary$In_xGa_{1-x}N_yAs_{1-y}$alloys only as a

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function of concentration$x$ and$y$ using the design of experiments (DoE). The content (In) ranged from 0 to 40% and the nitrogen content (N) was varied between 0 and 5%. In the case of InGaNAs grown on GaAs substrate, these concentrations are at the limits of the good feasibility in the MBE and MOVPE growth [24].This mathematical model despite its simplicity, givesus the opportunity to analyze the effect of indium, nitrogen and their interaction on the bandgap. The model can describe the band gap energy in the study domain very well.

II. THEORY APPROACH

A. Template Presentation of the Design of experiments method

In order to conveniently describe the phenomenon under study, involving a responseY that depends on several independent variables x_i which we shall call factors, it is possible to link mathematically the response Y tothe factors, x_i, as follows:

$$\mathrm{Y} = f(x_1, x_2, \dots, x_k) + \varepsilon. \tag{1}$$

Where the form of the true response function f is unknown and perhaps very complicated, and εincludes the experimental error and the effect of eventual random factors that have not been considered in the model [25]. We assume that the manufacturing process can be represented by a polynomial regression model of maximum degree 2called the postulated model:

$$\mathrm{Y}=a_0 + \sum_{i=1}^{k} a_i x_i + \sum_{\substack{i,j=1 \\ i<j}}^{k} a_{ij} x_i x_j + \sum_{i=1}^{k} a_{ii} x_i^2 + \varepsilon. \tag{2}$$

where

- Yis the response of interest which is the bandgap energy in our study.
- x_i represents a level of variable$i$.
- x_j represents a level of variable j.

a_0, a_i, a_{ij}, a_{ii} are the unknown coefficients of the polynomial to be calculated from the experimental measurement values and they denote: the value of the response at the center of the study domain, the main factors effects, the order oftwo interactions and the coefficients of second degree terms, respectively.The main effect of a given factor on the methodis to provide information on both:Determining which variables (factors) are most influential on the response Y (the greater its absolute value is, themore influential it is) and on theenhancement oftheresponse (the responseand the factor vary in the same direction if it is positive, the oppositeif it is negative). Here, within (2), the information on thebehavior of the system is both quantitative and qualitative. Then,the experimenter can easily perform a hierarchy in the influenceof the factors on the response and is able to straightforwardly identifythe most influential one.

This system can be written in a simple wayusing matrix notation.

$$\mathrm{Y} = \mathrm{X\,a} + \mathrm{e}. \tag{3}$$

where

- Yis the response vector.
- X is the model matrix or the design matrix which depends on the experimentalpoints used in the design and on the postulated model.
- a is the coefficient matrix.
- $e$is the error matrix.

This system of equations cannot be, in general, solved simply because there are fewerequations than there are unknowns. To findthe solution, we must use special matrix methods generally based on the criterion of least-squares [25]. The results areestimations of the coefficients, denoted as matrix $\hat{a}$. The algebraic result of the least-squares calculations is

$$\hat{a} = (\mathrm{X'X})^{-1}\mathrm{X'Y}. \tag{4}$$

Where X' is the transpose of X.

Two graphical representations of a very great importance can be used in the design of experiments: the histogram of the model coefficients and a sector representation giving the contribution of each factor in terms of percentages.

B. Centered and scaled variable (CSV)

In this study,the first variable is the indium content (noted x in natural units) that we will name $x_1$in coded and scaled variables, the second one is the nitrogen content (noted y in natural units) and which will be noted x_2 in (CSV) as well.DOE theory is quite generalizable using the coded units.Two important changes occur: (i) change of origin, if we take the amount of indium as an example, the middle of the interval [-1, +1] is zero and corresponds to thevalue 20%. The numerical value of the new zero origin, therefore, differsfrom the origin when expressed in the original experimental units and (ii) change of units so that the low and high levels take the values -1 and +1 as mentioned in Table 1.

TABLE I. THE DOMAIN OF VARIATION FOR EACH VARIABLE

	Indium content	**Nitrogen content**
Level (-1)	0	0
Level (+1)	0.40	0.05

The change from the original variables A to the centered and scaled variable (CSV) and vice versais given by the following formula:

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$$\alpha = \frac{A - A_0}{Step}. \quad (5)$$

where

- α is the centered reduced variable measure in units of step.
- $A$is the variable in normal units (degrees Celsius, Kelvin, bar …).
- A_0 is the central value in normal units between the high and low levels, point chosen as the origin for the centered and scaled variable.

Let's calculate the step for the indium. It's equal to half the difference between the high and the low levels, so $Step_{(In)} = (A_{+1} - A_{-1})/2 = (0.40 - 0)/2 = 0.2$ and A_0 is half of the sum of the high and low levels $A_{0(\mathrm{In})} = (A_{+1} + A_{-1})/2 = (0.40 + 0)/2 = 0.2$.

The same calculation is done for the nitrogen and we will have the $Step_{(N)} = 0.025$ and $A_{0(\mathrm{N})} = 0.025$. As an example, the corresponding coded units for the indium (x_1) and nitrogen (x_2) contents, equals to 0.35 and 0.015, respectively, becomes: $x_1 = (0.35 - 0.2)/0.2 = 0.75$ and $x_2 = (0.015 - 0.025)/0.025 = -0.4$, respectively.

Coded variables result from the ratio of two same sized physical units, so they have no dimension. The absence of natural units is due to the fact that all the variables have the same domain of variation (two coded units).

C. The coefficient of determination R^2

The analysis of variance allows the calculation of a very useful statistic: R^2, which is therefore a measure of the quality of the model. This statistic is the ratio of the sum of squares of the predicted responses (corrected for the mean) to the sum of squares of the observed responses (also corrected for the mean) [25].

III. RESULTS AND DISCUSSION

In this section, we predict a model for the bandgap energy of $In_xGa_{1-x}N_yAs_{1-y}$/GaAsas a simple function only of a composition and we answer the following question: which of the parameters and/or interactions that influencemost and significantly the energy gap (indium or nitrogen).

To develop the analytical model, we have used the centered reduced variables (CSV) to calculate the energy gap of $In_xGa_{1-x}N_yAs_{1-y}$as a function of coded variables x_1 and x_2.

Noted that our results are entered in this domain, so if we change the limits of variation of our variables, the model changes also. The experimental and (CSV) values for (x, y) experimental points are given in Table 2.

Table 2summarizes room temperature experimental values for the band gap of$In_xGa_{1-x}N_yAs_{1-y}$and GaN_yAs_{1-y}substrate on GaAs, noted Y_i gathered from several other works. Columns 2 and 3 of Table 2show the actual or natural unit values of x(In %) and y (N %), while columns 4 and 5 contain values of the corresponding coded variables x_1 and x_2 (obtained by the application of the equation (5) and falling between -1 and +1).

TABLE II. MATRIX OF EXPERIMENTS AND EXPERIMENTAL DATA

	NaturalVariables		**Coded variables**		**energy gap (eV)**	**References**
	x(In%)	*y (N %)*	x_1	x_2		
1	0	0	-1	-1	1.4240	
2	0	0.649	-1	-0.7404	1.2744	[26]
3	0	0.785	-1	-0.686	1.2653	"
4	0	0.86	-1	-0.656	1.2494	"
5	0	0.94	-1	-0.624	1.2400	"
6	0	1.17	-1	-0.532	1.2193	"
7	0	1.67	-1	-0.332	1.1546	"
8	0	1.85	-1	-0.26	1.1376	"
9	0	2.3	-1	-0.08	1.0973	"
10	0	4.2	-1	0.68	0.9787	"
11	7.3	3	-0.635	0.2	1.0065	"
12	11	3.4	-0.45	0.36	0.9274	"
13	8.8	4.2	-0.56	0.68	0.9185	"
14	0	4.5	-1	0.8	0.9560	[27]
15	0	0.08	-1	-0.968	1.4000	[28]
16	0	0.5	-1	-0.8	1.3000	[28]
17	34	0	0.7	-1	1.1210	[29]
18	34	2.4	0.7	-0.04	0.8980	"
19	35	3	0.75	0.2	0.9078	[30]
20	36	4.6	0.8	0.84	0.6070	[31]
21	36	5	0.8	1	0.8000	[32]
22	40	0.5	1	-0.8	0.9575	[33]
23	0	0.043	-1	-0.9828	1.400	[34]
24	38	2.2	0.9	-0.12	0.8406	[35]
25	38	3	0.9	0.2	0.8000	"
26	30	0.4	0.5	-0.84	1.0270	[36]
27	40	0	1	-1	1.0460	[33]

We will fit the modelusing the coded variables. In the case of two variables, the second-order model is:

$$Y = a_0 + a_1x_1 + a_2x_2 + a_{12}x_1x_2 + a_{11}x_1^2 + a_{22}x_{22}^2 \quad (6)$$

The experimental obtained values of the response (noted in Table 2) are given by:

$$Y = \begin{bmatrix} 1.4240 \\ 1.2744 \\ 1.2653 \\ 1.2494 \\ 1.2400 \\ 1.2193 \\ 1.1546 \\ 1.1376 \\ 1.0973 \\ 0.9787 \\ 1.0065 \\ 0.9274 \\ 0.9185 \\ 0.9560 \\ 1.4000 \\ 1.3000 \\ 1.1210 \\ 0.8980 \\ 0.9078 \\ 0.6070 \\ 0.8000 \\ 0.9575 \\ 1.4000 \\ 0.8406 \\ 0.8000 \\ 1.0270 \\ 1.0460 \end{bmatrix}. \quad (7)$$

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The X coefficient matrix can be easily obtained in a systematic way: the first columncorresponds to the a_0 values, the second and the third columns consist of the independent variables x_1 and x_2 CSV values which are the In and N concentration in natural units respectively, the entries in the fourth column are founded by multiplying each entry from x_1 by the corresponding entry from $x_2$and finally the two last columns are founded by squaring the entries in columns x_1 and x_2, respectively.

$$X = \begin{bmatrix} 1.0000 & -1.000 & -1.0000 & 1.0000 & 1.0000 & 1.0000 \\ 1.0000 & -1.000 & -0.7404 & 0.7404 & 1.0000 & 0.5482 \\ 1.0000 & -1.000 & -0.6860 & 0.6860 & 1.0000 & 0.4706 \\ 1.0000 & -1.000 & -0.6560 & 0.6560 & 1.0000 & 0.4303 \\ 1.0000 & -1.000 & -0.6240 & 0.6240 & 1.0000 & 0.3894 \\ 1.0000 & -1.000 & -0.5320 & 0.5320 & 1.0000 & 0.2830 \\ 1.0000 & -1.000 & -0.3320 & 0.3320 & 1.0000 & 0.1102 \\ 1.0000 & -1.000 & -0.2600 & 0.2600 & 1.0000 & 0.0676 \\ 1.0000 & -1.000 & -0.0800 & 0.0800 & 1.0000 & 0.0064 \\ 1.0000 & -1.000 & 0.6800 & -0.680 & 1.0000 & 0.4624 \\ 1.0000 & -0.635 & 0.2000 & -0.127 & 0.4032 & 0.0400 \\ 1.0000 & -0.450 & 0.3600 & -0.162 & 0.2025 & 0.1296 \\ 1.0000 & -0.560 & 0.6800 & -0.380 & 0.3136 & 0.4624 \\ 1.0000 & -1.000 & 0.8000 & -0.800 & 1.0000 & 0.6400 \\ 1.0000 & -1.000 & -0.968 & 0.9680 & 1.0000 & 0.9370 \\ 1.0000 & -1.000 & -0.800 & 0.8000 & 1.0000 & 0.6400 \\ 1.0000 & 0.7000 & -1.000 & -0.700 & 0.4900 & 1.0000 \\ 1.0000 & 0.7000 & -0.040 & -0.028 & 0.4900 & 0.0016 \\ 1.0000 & 0.7500 & 0.2000 & 0.1500 & 0.5625 & 0.0400 \\ 1.0000 & 0.8000 & 0.8400 & 0.6720 & 0.6400 & 0.7056 \\ 1.0000 & 0.8000 & 1.0000 & 0.8000 & 0.6400 & 1.0000 \\ 1.0000 & 1.0000 & -0.8000 & -0.800 & 1.0000 & 0.6400 \\ 1.0000 & -1.000 & -0.9828 & 0.9828 & 1.0000 & 0.9659 \\ 1.0000 & 0.9000 & -0.1200 & -0.108 & 0.8100 & 0.0144 \\ 1.0000 & 0.9000 & 0.2000 & 0.1800 & 0.8100 & 0.0400 \\ 1.0000 & 0.5000 & -0.840 & -0.420 & 0.2500 & 0.7056 \\ 1.0000 & 1.0000 & -1.000 & -1.000 & 1.0000 & 1.0000 \end{bmatrix} \quad (8)$$

The application of (4) gives us the vector, $\hat{a}$:

$$\hat{a} = \begin{bmatrix} 0.9475 \\ -0.1421 \\ -0.2034 \\ 0.0450 \\ -0.0067 \\ 0.0895 \end{bmatrix}. \quad (9)$$

Hence the model equation is written as:

$$Y = 0.9475 - 0.1421x_1 - 0.2034x_2 + 0.045x_1x_2 - 0.0067x_1^2 + 0.0895x_2^2. \quad (10)$$

This can be converted into an equation using the naturalvariables x and y by substituting the relationshipsbetween x_1 and x and x_2 and $y$using (5). So, the new simple formula that we proposed for the gap energy of the quaternary $In_xGa_{1-x}N_yAs_{1-y}$/GaAs is given by the following simple expression:

$$E_g = 1.4208 - 0.8685x - 17.096y + 9xy - 0.1675x^2 + 143.20y^2 \quad (11)$$

The number of necessary parameters to predict the band gap energy for $In_xGa_{1-x}N_yAs_{1-y}$ alloys can be reduced just to 2 where it was 7 in reference [37]; also this model does not require the BAC or any other parameters related to $In_xGa_{1-x}N_yAs_{1-y}$.

Table 3 shows the experimental values Y_i, the corresponding fitted values $\hat{Y}_i$, the corresponding values of BAC model Y_{BAC} and the residuals from this modelcompared with those obtained from the BAC model [16].

The BAC values are calculated with parameters recommended by Vurgaftman and Meyer [38]. However, it must be noted that the results are within limits of experimental error of about 18%, whereas the BAC model is within limits of error of about 36%. Also, the R^2 of this model is equal to 0.9561, value that is close to 1 which means that this mathematical model tends to represent well the experimental results. In the contour plot in figure. 1, we show a best fit of (11) to a set of data of the band gap of InGaNAs compiled in Table 3. The fit is excellent.

TABLE III. EXPERIMENTAL VALUES, FITTED VALUES AND ERRORS COMPARED WITH BAC MODEL

Observations	Y_i	$\hat{Y}_i$	Y_{BAC}	**E(%)**	**BAC error (%)**
1	1.4240	1.4208	1.4240	0.23	0
2	1.2744	1.3159	1.2919	-3.15	-1.35
3	1.2653	1.2954	1.2724	-2.33	-0.56
4	1.2494	1.2844	1.2623	-2.72	-1.02
5	1.2400	1.2728	1.2519	-2.57	-0.95
6	1.2193	1.2404	1.2239	-1.70	-0.37
7	1.1546	1.1752	1.1702	-1.76	-1.34
8	1.1376	1.1535	1.1528	-1.38	-1.32
9	1.0973	1.1033	1.1122	-0.55	-1.34
10	0.9787	0.9554	0.9722	2.44	0.66
11	1.0065	0.9922	0.9849	1.44	2.15
12	0.9274	0.9412	0.9202	-1.46	0.78
13	0.9185	0.9109	0.8893	0.83	3.29
14	0.9560	0.9415	0.9532	1.54	0.29
15	1.4000	1.4072	1.4006	-0.51	-0.043
16	1.3000	1.3389	1.3151	-2.91	-1.15
17	1.1210	1.1061	0.9538	1.34	17.52
18	0.8980	0.8518	0.7578	5.43	18.51
19	0.9078	0.8068	0.7092	12.52	28
20	0.6070	0.7521	0.6078	-19.29	-0.14
21	0.8000	0.7516	0.5868	6.44	36.34
22	0.9575	0.9827	0.8375	-2.56	14.33
23	1.4000	1.4135	1.4109	-0.95	-0.77
24	0.8406	0.8350	0.7312	0.67	14.96
25	0.8000	0.7852	0.6803	1.89	17.6
26	1.0270	1.0899	0.9611	-5.77	6.86
27	1.0460	1.0466	0.8823	0	18.55

Figure. 2 represents the histogram of the coefficients of our model, from this representation we can easily and quickly see the relative importance of each factor and therefore to compare the influence of each one. The absolute values of the coefficients point out the magnitude of factors influence.

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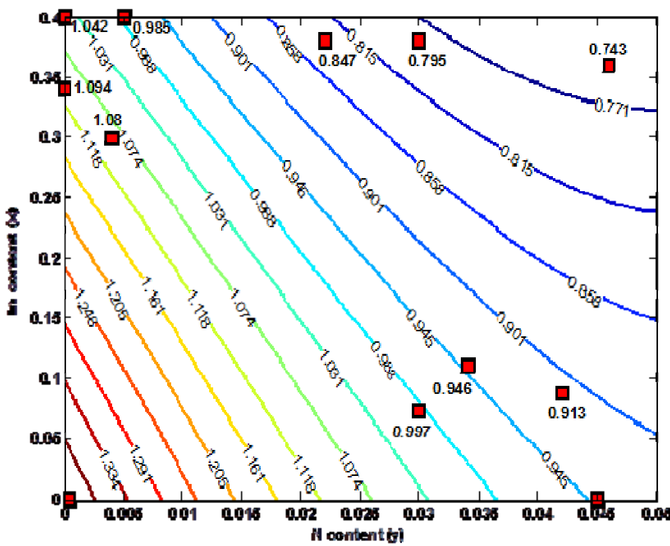

Figure 1. Effectof the factors on the InGaNAs/GaAs gap energy.

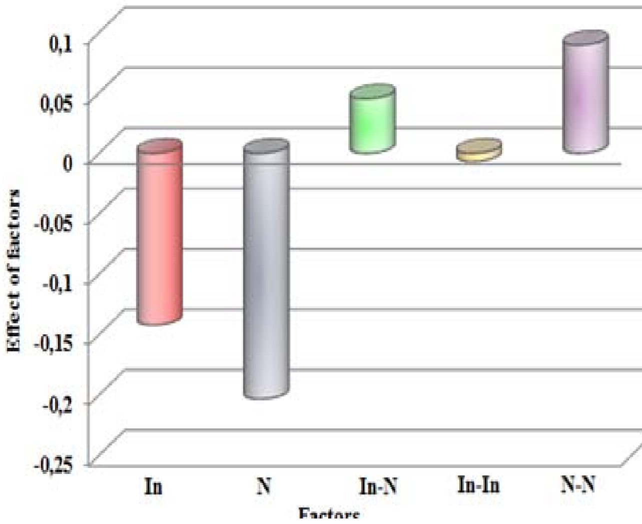

Figure 2. Contour plot of band gap for InGaNAs/GaAs.

- Indium concentration (factor 1) and nitrogen concentration (factor 2) are the most influential one, a_1= -0.1421and a_2= -0.2034,both coefficients are negative, which means that the band gap decreases as the corresponding factor increases (when they change from their lowest level to their highest level). Therefore the nitrogen influence on the reduction of the band gap is larger than that of indium.
- The interaction term a_{12}=0.045 is positive which means that the interaction between indium and nitrogen In-N leads to increase the energy band
- The (In-In) interaction represented by the coefficient a_{11}=-0.0067 hasn't any influence on the band gap while the(N-N) interaction is evaluated through the coefficients a_{22}=0.0895. This interaction leads to the increase of the bandgap.

Now our model gives us that the interaction between the nitrogen and himself raise the gap energy and we suggest that it might be the answer for the blue shift of the band gap in the thermal annealing cases, the roles will be reversed and wesuggest that this term will increase and will have the large effect which leads to the blue shift of the band gap while the singular effect of nitrogen will decrease. However, this suggestion should be verified in the next work.

In this section we have used the model proposed in this work to calculate theenergy gap of GaN_yAs_{1-y}.An indium content of 0% is therefore equal to −1 in coded units. We can use this value in (11) and it will give us the new formula that we proposed for the band gap of GaN_yAs_{1-y}/GaAs as a function only of increasing nitrogen concentration, y, which will be represented bythe following simple expression:

$$E_g = 1.4208 - 17.096y + 143.20y^2. \quad (12)$$

Figure. 3 shows that the effect of the nitrogen is negative, which means that the increase of nitrogen leads to a decrease of the gap energy while the interaction N-N has a positive effect and leads to an increase of the bandgap.

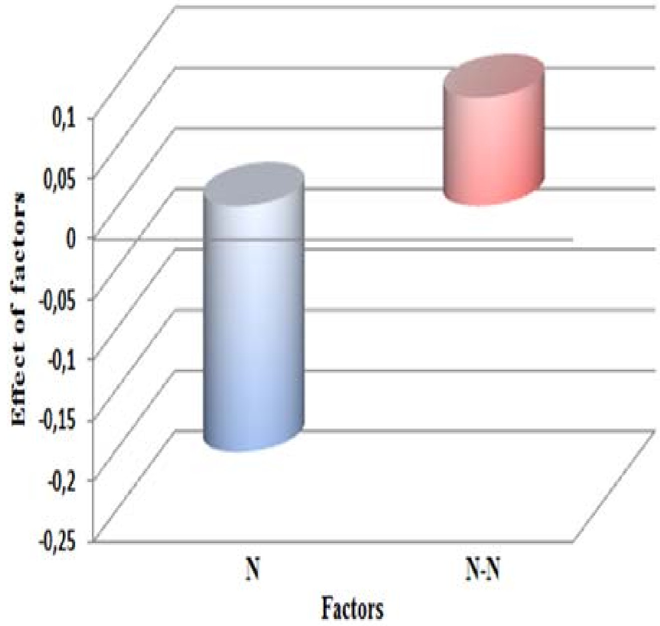

Figure 3. Effect of the factors on the GaNAs/GaAs gap energy.

Figure. 4 shows the variation of the room temperature bandgap energy as a function of N content. The figure also shows experimental values reported by various groups over the years [26,27,39,40].Figure. 4reported an excellent agreement between the calculated nitrogen composition dependence of E_g using the model proposed in this work (solid line)and experimental data.

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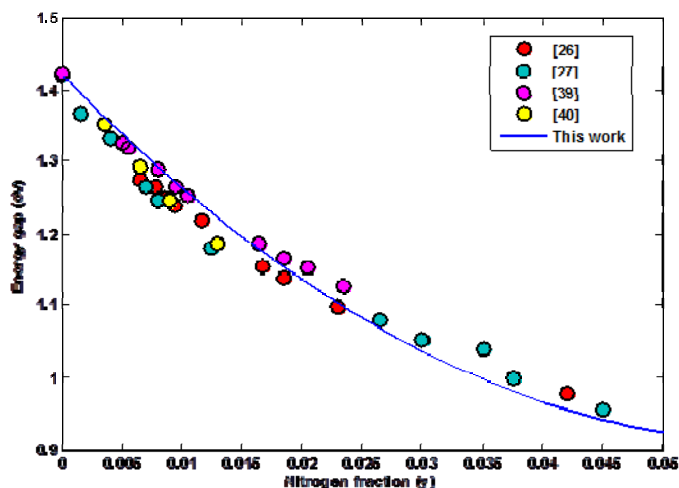

Figure 4. Variation of the room temperature band gap energy of GaN_yAs_{1-y} /GaAs asa function of the N content.

IV. CONCLUSION

This work presentsa methodology that could be applied to the determination of the bandgap energy model. It is, to our knowledge, the first time this method is being put into use for develop and understand the behavior of the bandgap of $In_xGa_{1-x}N_yAs_{1-y}$alloys.The number of parameters is restricted just to two parameters. Also, we haven't needed any BAC or other parameters. This model has R^2=0.9561which means that the model represents well the experimental results. It's worthy to note that our model is valid only inside the study domain,but the method is very easy and can be applied for each range of factors variation. The influence of the two variables and their interactions on the gap energy for $In_xGa_{1-x}N_yAs_{1-y}$/GaAs was evaluated. Therefore, when the indium and nitrogen increase, the nitrogen influence on the reduction of the band gap is larger than that of indium. Both interactions In-N and N-N lead to increase the gap energy while In-In interaction has no influence. Whereas for GaN_yAs_{1-y}/GaAs, reduction of bandgap energy is dominated by the introduction of an increasing amount of nitrogen and the interaction N-N leads to an increase of the bandgap. So, the gap energy of$In_xGa_{1-x}N_yAs_{1-y}$ and GaN_yAs_{1-y}alloys can be accurately described in terms of our simple analytical expressions. Another area of future work is the extension of the method to multiple response processes for dilute nitride alloys.

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