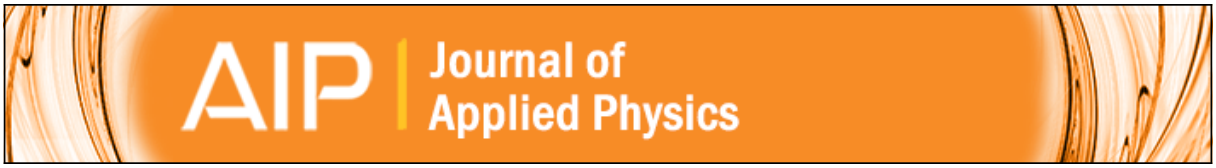

Temperature dependence of optical transitions in AlGaAs

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Temperature dependence of optical transitions in AlGaAs

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AlGaAs structures with different aluminum concentration ($x=0.0$, 0.17, 0.30, and 0.40) were characterized by photoluminescence and photoreflectance techniques. The temperature dependence of optical transitions in the temperature ranging from 2 to 300 K were investigated. Y. P. Varshni [Physica (Utrecht) **34**, 194 (1967)], L. Viña *et al.* [Phys. Rev. B **30**, 1979 (1984)], and R. Pässler [Phys. Status Solidi B **200**, 155 (1997)] models were used to fit the experimental points. The Pässler model gave the best adjustment to the experimental points. The tree models showed that the empirical parameters obtained through the adjustment of the experimental data in the three different models are aluminum composition dependent in the ternary alloy. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1367875]

I. INTRODUCTION

AlGaAs, at low aluminum concentration, $0<x<0.45$, has attracted much attention due to properties which make it useful for many types of heterojunction structures used in microelectric and optoelectronic devices.[1–3] The temperature variation of the band gap energy or of the excitonic transition associated to it has been widely investigated both theoretically and experimentally during the last decades for many different semiconductor materials.[4–9] Two mechanisms are responsible for the temperature dependence of the excitonic transitions in semiconductor materials: the electron–phonon interaction and the thermal expansion of the lattice. To many semiconductor crystals, the principal mechanism is the electron–phonon interaction. To GaAs, the mechanism of thermal expansion gives a contribution of about 44% of the total gap reduction.[10] This contribution is higher than that found in other semiconductors.

The temperature dependence of the forbidden gap or of the excitonic energy of semiconductor materials is generally described by the empirical relation proposed by Varshni,[11] or the semiempirical relations proposed by Viña *et al.*,[12] or Pässler.[13] According to many authors, the Varshni model provides a good approximation to high temperatures; however, for the GaAs the parameters obtained by performing numerical fittings on the basis of the Varshni formula differed significantly from paper to paper.[13] For the AlGaAs, it has been argued that the fitting by the Varshni model does not present correct values and the parameters of Varshni formula depend on samples quality.[14] For ternary alloys such as AlGaAs, some works have proposed either a linear[15] or a quadratic dependence[16] for the Varshni and Viña parameters on the aluminum composition in the alloy system AlGaAs. A systematic study of the aluminum composition dependence of the equations parameters proposed by the different models has not been carried out yet. Moreover, the Pässler model has not yet been applied to AlGaAs. Using photoluminescence (PL) and photoreflectance (PR) techniques, we describe in the present work the temperature dependence of excitonic energies in AlGaAs alloys. Four samples with concentration in the range $0<x<0.40$ are investigated. A comparison between the three analytical models proposed by Varshni, Viña, and Pässler, employed to fit the temperature dependence of excitonic transition for the AlGaAs alloy is presented din this work. A composition dependence of the fitting parameters used in the three models is proposed.

II. THEORETICAL MODELS

Many analytical models to describe the temperature dependence of excitonic transitions have been developed in the last decades. The more conventional is the Varshni[11] (1967) empirical model described by the following equation:

$$E_g(T)=E_g(T=0)-\alpha_{\mathrm{Var}}\frac{T^2}{\beta+T}, \tag{1}$$

where $E_g(T)$ is the energy gap at the temperature T and α_{Var} and β are parameters to fit the experimental data. Another recent model containing the Bose–Einstein occupation factor for photons, although still semiempirical, is proposed by Viña *et al.*[12] (1984) and is described as

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$$E_g(T)=E_B-a_B\left[1+\frac{2}{\exp(\Theta_B/T)-1}\right], \tag{2}$$

where a_B represents the strength of the electron–phonon interaction, $\Theta_B \equiv \hbar w/k_B$ is the characteristic temperature representing the effective phonon energy on the temperature scale[12,13,15,16] and $E_g(T=0)=E_B-a_B$. A more recent analytical model is proposed by Pässler[17] (1997) and can be represented by the following equation:

$$E_g(T)=E_g(T=0)-\frac{\alpha\Theta}{2}\left[\sqrt[p]{1+\left(\frac{2T}{\Theta}\right)^p}-1\right], \tag{3}$$

where $E_g(T=0)$ is the energy gap at 0 K, $\alpha \equiv S(\infty) \equiv -(dE(T)/dT)_{T\to\infty}$ is the high-temperature limiting value of the forbidden gap entropy,[18,19] Θ is a characteristic temperature parameter of material specific representing the effective phonon energy $\hbar w = k_B\Theta$ in units of the absolute temperature, and p is a empirical parameter related to the shape of the underlying electron–phonon spectral functions.[20,17] According to the aforementioned expression, the Varshni and Viña models present three parameters while the Pässler model presents four adjustable parameters to fit $E_g(T)$ experimental data. Pässler demonstrated the inevitability of adding a fourth parameter in the expressions used to fit the temperature dependence of forbidden band gap or excitonic transition.[17] Experimental data observed in the literature present typical curves for $E_g(T)$ with nonlinear dependencies at low temperatures ($T<30$ K) and linear dependencies at high-temperatures ($T>100$ K). The main difference between these three alternative expressions used to fit the temperature dependence of $E_g(T)$ in GaAs is in the low temperature range (2 K$<T<$30 K).[17] In this range of temperature, the Varshni model leads to an overestimation of the measured temperature dependence and the Viña model leads to an underestimation.[17] To our knowledge, a comparison among the three models in the AlGaAs has not been done yet. In this article, we make a comparison among these three analytical models for the temperature dependence of excitonic transitions in AlGaAs alloy in the range $T=2$ K to $T=300$ K. From the experimental data on the temperature dependence of excitonic transitions, we can obtain the compositional dependence of the parameters used in the (1), (2), and (3) relations on the basis of the Varshni, Viña, and Pässler models, respectively. Considering the relative contribution of the thermal expansion mechanism to the gap energy variation on the GaAs, this article is concerned with the overall behavior of the different adjustment curves obtained by the different models. This relative contribution should not be a problem once the contributions of the thermal expansion mechanism present a temperature dependence roughly similar to that of the electron–phonon interaction mechanism, especially for high temperatures.[17] The standard approach to defining the best fit is to choose the parameters so that the sum of the deviation squares of the theoretical curve from the experimental points is minimum. For this, we used the expression:[21]

$$S^2=\left(\frac{1}{n-m}\right)\sum_{i=1}^{n}(E_{g_i}^{\exp}-E_{g_i}^{\text{ajust}})^2, \tag{4}$$

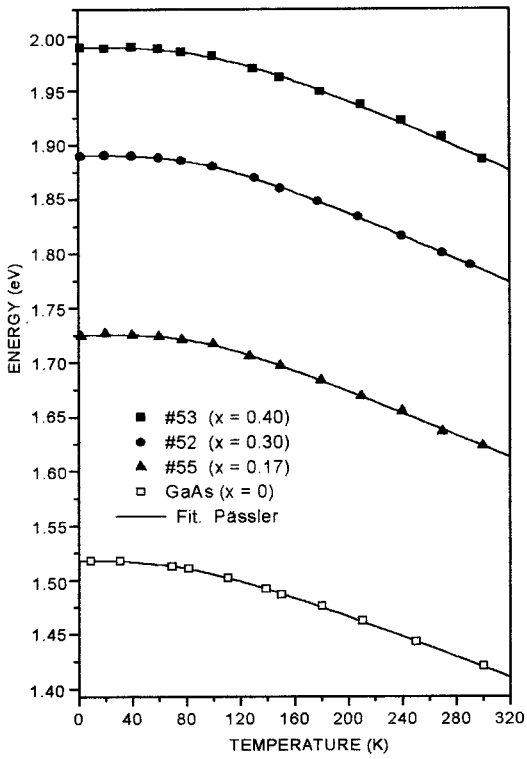

FIG. 1. The temperature dependence of excitonic transition in the range 2–300 K of AlGaAs alloy for $x=0$ (the data for GaAs reported by Logothetidis *et al.*[22]), 0.17, 0.30, and 0.40.

where the superscript exp indicates experimental values *cal* calculated values of E_g using the relations (1), (2), and (3), for the ith data point. $(n-m)$ is the number of degrees of freedom left after fitting n data points with m variable parameters in the fitting function.

III. EXPERIMENTAL DETAILS

A set of four nominally undoped $Al_xGa_{1-x}As$ samples used in the experiments was grown by molecular beam epitaxy on (100) oriented, undoped semi-insulating GaAs substrates. The aluminum composition was 0.17, 0.30, and 0.40. The experimental data for $x=0$ were redigitalized from the work of Logothetidis *et al.*[22] The PL measurements were performed in the temperature range of $T=2$ to 150 K, using a 441.6 line of a He–Cd laser as excitation source. The emission spectra were analyzed with a 0.5 m monochromator and detected by a liquid N_2 cooled Ge photodetector using standard lock-in techniques. PR measurements were performed in the temperature range of $T=150$ to 300 K. The probe beam was produced using a tungsten halogen lamp and a 0.5 m monochromator. The PR spectra were analyzed with a Si photodiode using the standard lock-in technique. The composition of the layer was determined by double crystal x-ray diffraction with an accuracy of about $\pm$ 1% as described elsewhere by Bassignana *et al.*[23]

IV. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of excitonic transitions in the 2 to 300 K range for the AlGaAs alloy at

$x=0$,0.17,0.30,0.40, and the numerical fit on basis of Pässler model—Eq. (3). With the considerable reduction of the PL signal in temperatures higher than 150 K, the PR technique was used in the 150–300 K interval. Due to the different physical processes involved in the different techniques, a difference in the energy denominated ''Stokes shift'' is expected. Schubert *et al.*[24] verified that for high enough intensity, the PL spectrum energy peak and the energy obtained from the optical absorption spectrum for the InGaAs alloy nearly coincide. Recently, Shan *et al.*[25] observed through the statistical distribution model (this model was proposed by Schubert *et al.*[26] for the random distribution of Al and Ga atoms in the sites from the group-III on the AlGaAs alloy) that for highly sufficient temperatures, the Stokes shift between the excitonic transitions for the InGaN alloy obtained through the PL and PR techniques is nearly annulled. In our work, the PR spectra were obtained for temperatures over 150 K and for the alloy with random distribution (homogeneous samples).[26] Therefore, in agreement with the aforementioned authors, a small Stokes shift is expected between the spectra of PL and PR in the alloy analyzed by us. A 1 to 2 meV difference between the PL and PR lines at 130 K was observed. This difference was taken into account for experimental data comparatibility reasons in order to obtain the temperature excitonic transition curve for the temperature interval considered (2–300 K). The values obtained for the difference between the transition energy values at 2 and at 300 K in this article and in that of El Allali *et al.*[14] $[E_g(2\,\mathrm{K})-E_g(300\,\mathrm{K})]$, which used the PL technique alone at the same temperature interval discussed here, are very similar. This indicates that the Stokes shift is small and varies insignificantly with the temperature for the AlGaAs. A comparison between the three semiempirical models used to fit the temperature dependence of excitonic transitions for sample with $x=0.30$ is shown in Fig. 2. As for the GaAs, the main difference between these models for the AlGaAs alloy is in the range of low temperatures ($2\,\mathrm{K}<T<30\,\mathrm{K}$). In this range of temperature, the Varshni model leads to an overestimation of the measured temperature dependence and the Viña model leads to an underestimation. The same results are observed in the numerical fits for all samples discussed here. Then, the behavior of numerical fits using the models of Varshni, Viña, and Pässler observed in the GaAs is also found in the AlGaAs. The fitted curve obtained by the Pässler expression gives a better adjustment to the experimental points.

Figure 3 shows the temperature dependence of the excitonic transitions for sample with $x=0.30$ fitted by the Varshni, Viña, and Pässler models in the temperature range of 2 to 125 K and extrapolated to 300 K. The fitted curves by the Varshni, Viña, and Pässler expressions give us a good description of the experimental points as can be seen by the S^2 values in Table I. When extrapolated to a high-temperature region, however, the curves of the three analytical models diverged significantly. This divergence showed a difference of 15 meV in the Varshni model, 8.5 meV in the Viña model, and 2 meV in the Pässler model between the fitted curves and the experimental point at room temperature. Then, the Pässler estimates are found to be in better agreement with the experimental points in the extrapolated curves in the high-temperature region. Thus, the Pässler model showed to be more adequate for the extrapolation at high temperatures from results obtained through optimized techniques for low temperatures as in the case of the PL technique. Table I gives the Varshni, Viña, and Pässler parameters values and S^2 calculated from Eq. (4) for the three models from the four samples studied.

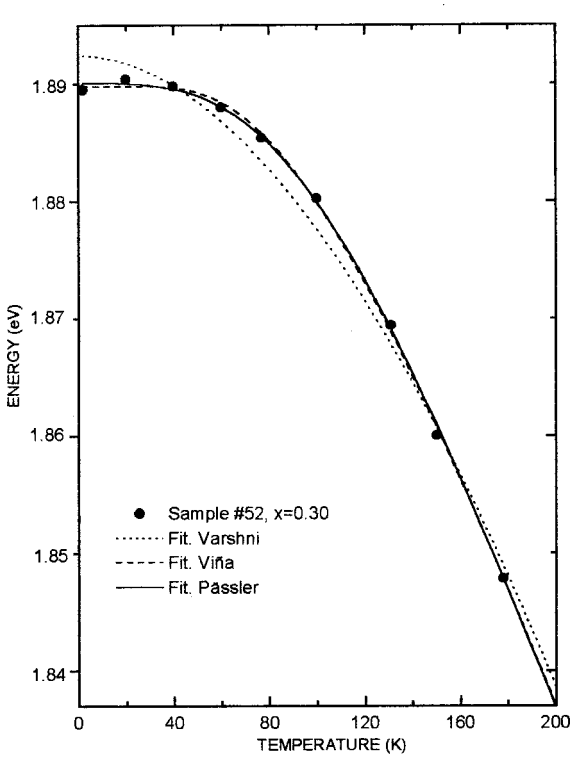

FIG. 2. Comparison between the three analytical models of Varshni (dotted line), Viña (dashed line), and Pässler (solid line) and experimental data for sample with $x=0.30$.

In Fig. 4, we present the compositional dependence of the empirical parameters for the three models analyzed. In Fig. 4, we see that the optimized parameters increase as the alloy composition increases. The dotted lines represent the linear or quadratic fit of the dependence of optimized parameters on the aluminum concentration. The results these fits can be presented by

$$\text{Varshni}\begin{cases} E_{\mathrm{Var}}(T=0,x)=1.523+1.23x \quad (\mathrm{eV}), & (5a)\\ \alpha_{\mathrm{Var}}(x)=5.5+3.35x+50x^2 \quad (10^{-4}\,\mathrm{eV/K}), & (5b)\\ \beta(x)=198+88x+4300x^2 \quad (\mathrm{K}), & (5c)\end{cases}$$

$$\text{Viña}\begin{cases} E_B(x)=1.576+1.33x \quad (\mathrm{eV}), & (5d)\\ a_B(x)=60+81x \quad (\mathrm{meV}), & (5e)\\ \Theta_B(x)=240+180x \quad (\mathrm{K}), & (5f)\end{cases}$$

$$\text{Pässler}\begin{cases} E(T=0,x)=1.517+1.23x \quad (\mathrm{eV}), & (5g)\\ \alpha(x)=4.9+0.7x+3.7x^2 \quad (10^{-4}\,\mathrm{eV/K}), & (5h)\\ \Theta(x)=202+5x+260x^2 \quad (\mathrm{K}). & (5i)\end{cases}$$

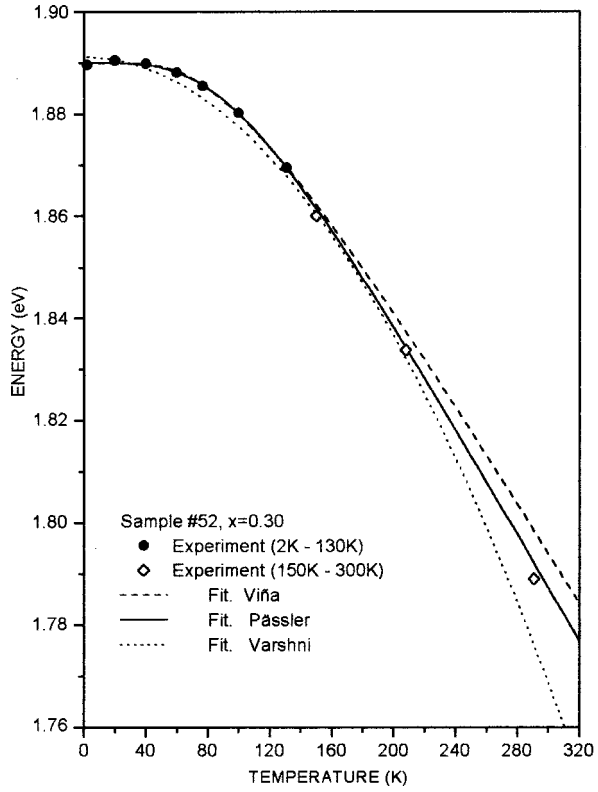

FIG. 3. The temperature dependence of excitonic transitions to AlGaAs with $x=0.3$ fitted by Varshni (dotted line), Viña (dashed line), and Pässler (solid line) models in the range 2–130 K extrapolated to 300 K. The experimental values at 2–130 K and 150–300 K are represented by filled circles and empty squares, respectively.

Our results obtained for $E_{\mathrm{Var}}(T=0,x)$, $E_B(x)$, and $E(T=0,x)$ displayed in Figs. 4(a), 4(d), and 4(g) [and also by Eqs. (5a), (5d), and (5g)] show a linear dependence on x. These results are in good agreement with the experimental data obtained by Casey and Panish[27] (represented by continuous lines in Fig. 4) that show a linear dependence on x of the fundamental band gap of the AlGaAs alloy in the direct gap range ($0<x<0.45$). The dependence of other adjusted parameters with the aluminum composition $[(\alpha_{\mathrm{Var}}(x),\beta(x)), (a_B(x),\Theta_B(x)), (\alpha(x),\Theta(x))]$ shown, respectively, in Figs. 4(b), 4(c), 4(e), 4(f), 4(h), and 4(i) are linear to Viña model and quadratic to Varshni and Pässler models. This behavior could be associated with the different choice of the electron–phonon spectral function for each model.[17] Viña *et al.*[12] for example, adopted the spectral function as a singular function while Pässler adopted the spectral function as a power law.[17,20] A set of properties of the $Al_xGa_{1-x}As$ alloy system as the band gap energy, effective masses and phonon energies are composition dependent and increases as the composition increases. Thus, the composition dependence of the optimized parameters in the different models obtained by fitting the optical transitions experimental data with the temperature seems in principle obvious. Therefore, the composition dependence of the optimized parameters used to fit the experimental data in the three models is expected, despite the unclear physical significance of these parameters.

The principal mechanism responsible for the temperature dependence of band-gap energy is the electron–phonon interaction.[28,18] Calculations of electron–phonon spectral functions in Si and Ge have shown that the energy shift due to electron–phonon interaction includes contributions from both acoustic as well as optical phonons.[29] Then Θ_B in expression (2) should correspond to the average frequency of the phonon spectra in the temperature scale. To the GaAs, the electron–acoustical longitudinal phonon interactions give the dominant contribution to the band gap or exciton transition energy shift.[5,17,30] In the $Al_xGa_{1-x}As$, the energy values associated to Θ_B in the Viña model increases due to the variation of the aluminum concentration.[15] However, these works have not explained whether this increase is related to the optical or acoustical branches of the phonon spectra or to other mechanisms. Pavesi and Guzzi[31] have shown that the energy of the acoustical longitudinal phonons are not dependent on the aluminum concentration in the AlGaAs alloy.

TABLE I. Parameters of the Varshni, Viña, and Pässler expressions. Var, Vin, and Pass are abbreviations of Varshni, Viña, and Pässler, respectively.

Samples	$E_g(T=0)$ (meV) E_B (meV) $E_g(T=0)$ (meV)	$\alpha_{\mathrm{Var}}(10^{-4}$ eV/K) or a_B (meV) or $\alpha(10^{-4}$ eV/K)	β (K) or Θ_B (K) or Θ (K)	p	S^2 (meV)^2
GaAs	1523.1±1.2	5.4±0.5	173±45	···	S^2(Var)=2.83
$x=0$	1576.6±1.6	59.7±7.4	241±7	···	S^2(Vin)=0.91
	1517.6±1.1	4.9±0.2	203±8	2.85	S^2(Pass)=0.87
AlGaAs	1728.7±1.3	8.1±1.3	392±102	···	S^2(Var)=7.4
$x=0.17$	1802.4±4.1	76.9±4.4	275±11	···	S^2(Vin)=1.7
	1725.1±0.6	5.1±0.1	208±9	4.0	S^2(Pass)=1.3
AlGaAs	1892.8±1.0	10.5±1.9	573±159	···	S^2(Var)=4.6
$x=0.30$	1975.7±2.2	85.9±2.3	289±6	···	S^2(Vin)=0.31
	1890.0±0.3	5.5±0.1	220±7	3.5	S^2(Pass)=0.28
AlGaAs	1991.5±1.2	14.2±2.5	941±250	···	S^2(Var)=5.6
$x=0.40$	2081.2±4.6	94.4±4.8	317±10	···	S^2(Vin)=0.77
	1989.7±0.6	5.9±0.1	275±9	2.9	S^2(Pass)=0.88

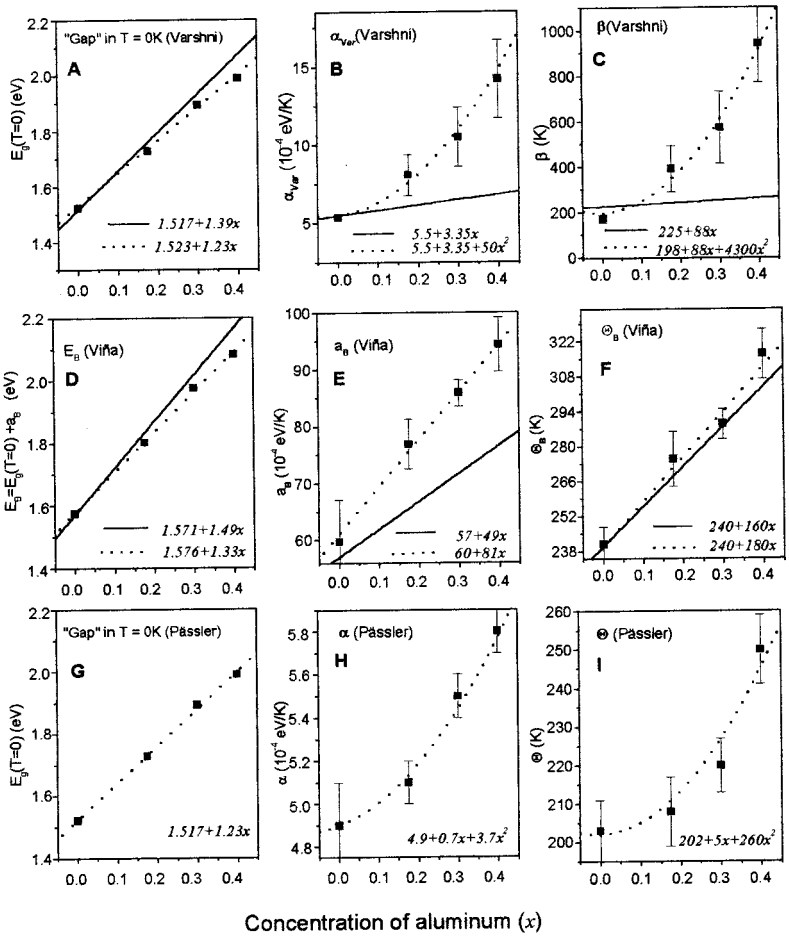

FIG. 4. Composition dependence of the parameters of Varshni (A,B,C), Viña (D,E,F), and Pässler (G,H,I) models.

Then the increase of $\Theta_B(\Theta)$ due to the aluminum concentration can be related only to the optical branches in the phonon spectra. Our results for the Viña model give: $k_B\Theta_B = 20.7$ meV ($x=0.0$), 23.7 meV ($x=0.17$), 24.9 meV ($x=0.30$), and 27.3 meV ($x=0.40$) showing an increase in the mean phonon energy with the aluminum concentration. For the Pässler model, we have: $k_B\Theta = 17.5$, meV ($x=0.0$), 17.9 meV ($x=0.17$), 18.9 meV ($x=0.30$), and 23.7 meV ($x=0.40$) showing also an increase in the mean phonon energy with aluminum concentration but with smaller values when compared to the Viña model. This suggests that the Pässler model acoustical phonons give a more important contribution to the reduction of energy when the temperature increases. More systematic works are necessary to elucidate this question.

V. CONCLUSIONS

In conclusion we have shown in this work that the temperature dependence of the parameters obtained by fitting the experimental data of the optical transitions in the AlGaAs alloy system is basically the same as that found in the GaAs. The Pässler model gives the best adjustment to experimental points for the AlGaAs system. And lastly, the optimized parameters in the three different models are aluminum concentration dependent and increase as the Al concentration increases in the AlGaAs system.

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