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Interpolation of quaternary III-V alloy parameters with surface bowing estimations

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We present the weighted-sum expression of quaternary parameter estimations via balancing surface bowing estimation errors. Expressions have been derived for both quaternary alloys $A_xB_yC_zD$ and $A_xB_{1-x}C_yD_{1-y}$ and applied to the band gaps of AlGaInAs, AlGaInP, InAsPSb, GaInAsP, and GaInAsSb. Approximation can be improved by fitting known data of quaternary alloys and maximum deviations are less than 20 meV with respect to the lattice-matched polynomial expressions. © 2007 American Institute of Physics. [DOI: 10.1063/1.2404791]

I. INTRODUCTION

The III-V compound semiconductor alloy parameters have been investigated for decades, e.g., see Ref. 1 as a summary, whereas their explicit approximate expressions in molar fractions are expected by scientific researchers, device designers, and material growth engineers. Little argument exists on the interpolation of ternary parameters, which incorporates a bowing term to the linear Vegard law employing known binary compound parameters.² However, the task is far more complicated for quaternary III-V alloys and there is no standard interpolation scheme. Under a special condition, i.e., lattice matching, this problem can be eased by interpolation between two end points with a bowing term, e.g., Eq. (5.4) in Ref. 1. However, for some work such as intermixing³ on quaternary alloys whose compositions may become lattice mismatched as a result of interdiffusion at nonunity V/III diffusion length ratio, it is still of interest to have a molar fraction expression. In an early empirical interpolation scheme, a quaternary alloy parameter is expressed as a weighted sum of four ternary parameters for the $A_xB_{1-x}C_yD_{1-y}$ type of quaternary alloys,⁴ or three ternary parameters for the $A_xB_yC_zD$ type of quaternary alloys.⁵ An interpolation scheme of polynomial expression was proposed to incorporate the ternary bowing parameters for the $A_xB_{1-x}C_yD_{1-y}$ type of quaternary alloys, showing better performance.⁶ Recently, a systematic analysis on polynomial interpolation was presented based on the standard multivariable quadratic interpolation.⁷

Although the bowing of a surface function inside a boundary cannot be inferred from the values of the function on the boundary,⁷ with some known data, especially those at lattice-matched (LM) compositions, it is still possible to further improve the approximation. In this article, we derive the weighted-sum expression of a quaternary parameter via balancing surface bowing estimation errors for both types of quaternary alloys $A_xB_yC_zD$ and $A_xB_{1-x}C_yD_{1-y}$. Connections of several schemes are demonstrated in this context and interpolated results of several quaternary alloys are presented in the discussion.

II. QUATERNARY ALLOY $A_xB_yC_zD$

A material parameter function $Q(x, y)$ of a quaternary alloy $A_xB_yC_zD$ (or $DA_xB_yC_z$) can be plotted as a surface in an equilateral triangular domain as shown in Fig. 1. $Q(x, y)$ reaches binary parameters $B_{A(D)}$, $B_{B(D)}$, and $B_{C(D)}$ at the three corners and deviates from the linear Vegard law with ternary bowing parameters $C_{AB(D)}$, $C_{AC(D)}$, and $C_{BC(D)}$ along the three boundaries. The molar fractions can be represented by the distances to the boundaries so that they satisfy $x + y + z = 1$. To estimate $Q(x, y)$ in a path, e.g., $A-T_{BC}$ in Fig. 1, with a surface bowing, the quaternary alloy is expressed in the form of $A_x(B_wC_{1-w})_{1-x}D$ with $w = y/(1-x)$ such that

$$Q_{A-BC} = xB_A + (1-x)T_{BC} - x(1-x)C_{A-BC}, \quad (1)$$

where

$$T_{BC} = wB_B + (1-w)B_C - w(1-w)C_{BC}, \quad (2)$$

and in a similar consideration to a ternary parameter expression, the bowing parameter in Eq. (2) is written as

$$C_{A-BC} = wC_{AB} + (1-w)C_{AC} - w(1-w)D_A. \quad (3)$$

Substituting Eqs. (2) and (3) in Eq. (1) results in

FIG. 1. Equilateral triangular domain for a parameter surface $Q(x, y)$ of a quaternary alloy $A_xB_yC_zD$.

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$$Q_{A-BC} = xB_A + yB_B + zB_C - \frac{yz}{1-x}C_{BC} - xzC_{AC} - xyC_{AB} + \frac{xyz}{1-x}D_A. \quad (4)$$

Similar expressions can be derived for the estimations in the other two paths,

$$Q_{B-AC} = xB_A + yB_B + zB_C - yzC_{BC} - \frac{xz}{1-y}C_{AC} - xyC_{AB} + \frac{xyz}{1-y}D_B, \quad (5)$$

$$Q_{C-AB} = xB_A + yB_B + zB_C - yzC_{BC} - xzC_{AC} - \frac{xy}{1-z}C_{AB} + \frac{xyz}{1-z}D_C. \quad (6)$$

In sorting out an expression for $Q(x, y)$, it is considered that the Q function is required to be unique,⁷ i.e., $Q_{A-BC} = Q_{B-AC} = Q_{C-AB}$. However, the argument is that the real expressions for D_A , D_B , and D_C might never be known, whereas Eqs. (4)–(6) are just estimations, which have to be combined to give an optimal estimation for $Q(x, y)$. Since the estimated D_A , D_B , and D_C results in errors in the quaternary parameter via the factors $xyz/(1-x)$, $xyz/(1-z)$, and $xyz/(1-y)$, respectively, it is natural to adopt a weighted sum of Q_{A-BC} , Q_{B-AC} , and Q_{C-AB} by considering these weighting factors,

$$Q(x, y, z) = \frac{[(1-x)/xyz]Q_{A-BC} + [(1-y)/xyz]Q_{B-AC} + [(1-z)/xyz]Q_{C-AB}}{(1-x)/xyz + (1-y)/xyz + (1-z)/xyz} = Q_0(x, y, z) + \Delta Q(x, y, z), \quad (7)$$

which comprises a term of fundamental estimation,

$$Q_0(x, y, z) = xB_A + yB_B + zB_C - xyC_{AB} - yzC_{BC} - xzC_{AC}, \quad (8)$$

and a term of correction,

$$\Delta Q(x, y, z) = \frac{1}{2}xyz(D_A + D_B + D_C - C_{AB} - C_{BC} - C_{AC}). \quad (9)$$

Since it is not necessary to take D_A , D_B , and D_C as constants, Eq. (9) can be written in a general form for better approximation,

$$\Delta Q(x, y, z) = xyz(d_0 + d_1x + d_2y). \quad (10)$$

It has to be noticed that the quaternary parameter expression, i.e., Eqs. (7)–(10), is different from the other expression which is a weighted sum of ternary parameters at feet of perpendiculars,⁵

$$Q(x, y, z) = \frac{xyT_{A_{1-u}B_uD} + yzT_{B_{1-v}C_vD} + xzT_{A_{1-w}C_wD}}{xy + yz + zx}, \quad (11)$$

where $u = (1-x+y)/2$, $v = (1-y+z)/2$, and $w = (1-x+z)/2$.

We evaluate the above equations for Γ band gaps of the three quaternary alloys $\text{Al}_x\text{Ga}_y\text{In}_z\text{As}$, $\text{Al}_x\text{Ga}_y\text{In}_z\text{P}$, and $\text{InAs}_x\text{P}_y\text{Sb}_z$. All adopted binary parameters and ternary bowing parameters are recommended in Ref. 1. To simplify the discussion, we only examine their maximum deviations from the polynomial expressions [Eq. (5.4) in Ref. 1] at LM compositions. As seen in Table I, the fundamental estimation [Eq. (8)] has much better performance than the weighted sum of ternary parameters at feet of perpendiculars [Eq. (11)]. The

main reason is that the latter does not take surface bowing into account although the ternary bowing has been adopted along the ternary boundaries.

As a general procedure to improve the approximation, the fundamental estimation can be calculated first at given molar fractions and its difference from the known data is fitted to obtain the correction expression. Here coefficients in the correction expressions are determined by fitting the LM polynomial expressions in adoption of bowing parameters recommended in Ref. 1. With the single coefficient in Eq. (10), the deviation is suppressed significantly to less than 20 meV for the three alloys, which is not bad for an interpolation scheme giving explicit expressions for quaternary alloys of arbitrary compositions. The contour diagrams for the three quaternary alloys are plotted in Fig. 2. The deviations can be further reduced down to several meV with three fitting coefficients. However, fitting with three coefficients along a LM condition leaves a free coefficient d_0 and data at molar fractions other than the LM condition are required in order to determine it.

III. QUATERNARY ALLOY $A_xB_{1-x}C_yD_{1-y}$

Several schemes of interpolation for a quaternary alloy $A_xB_{1-x}C_yD_{1-y}$ have been studied in literatures,^{4,6-8} and in the context of the weighted-sum expression with surface bowing consideration their connections will be seen. The function $Q(x, y)$ can be plotted as a surface in a square domain enclosed by four boundaries A , B , C , and D as shown in Fig. 3. It reaches the binary material parameters B_{AC} , B_{AD} , B_{BC} , and B_{BD} at the four corners, and is reduced to the ternary parameter expressions with bowing parameters C_{ABC} , C_{ABD} , C_{ACD} , and C_{BCD} along the boundaries,

TABLE I. Maximum $|E_g(x,y) - E_g^{(LM)}|$ for $\text{Al}_x\text{Ga}_y\text{In}_z\text{As}$, $\text{Al}_x\text{Ga}_y\text{In}_z\text{P}$, and $\text{InAs}_x\text{P}_y\text{Sb}_z$ ($T=300$ K) using (i) weighted sum of ternary parameters at feet of perpendiculars [Eq. (11)] and weighted sum considering surface bowing [Eq. (7) and (10)] with (ii) none (fundamental estimation), (iii) single, and (iv) three fitting coefficients. (a) $\text{Al}_x\text{Ga}_y\text{In}_z\text{As}$ LM to InP: $(\text{Al}_{0.48}\text{In}_{0.52}\text{As})_z(\text{Ga}_{0.47}\text{In}_{0.53}\text{As})_{1-z}$. (b) $\text{Al}_x\text{Ga}_y\text{In}_z\text{P}$ LM to GaAs: $(\text{Al}_{0.52}\text{In}_{0.48}\text{As})_z(\text{Ga}_{0.51}\text{In}_{0.49}\text{As})_{1-z}$. (c) $\text{InAs}_x\text{P}_y\text{Sb}_z$ LM to InAs: $(\text{InSb}_{0.31}\text{P}_{0.69})_z(\text{InAs})_{1-z}$.

Equation and condition				$ \Delta E_g ^{\max}$ (meV)
(a)	(i)	Eq. (11)		70
	(ii)	$\Delta Q=0$		36
	(iii)	$\Delta Q=-0.9272xyz$		16
	(iv)	$\Delta Q=xyz[d_0+(0.9312-2.0833d_0)x+(-4.8806-2.1277d_0)y]$		4
(b)	(i)	Eq. (11)		54
	(ii)	$\Delta Q=0$		46
	(iii)	$\Delta Q=-1.4446xyz$		0.2
	(iv)	$\Delta Q=xyz[d_0+(-2.8068-1.9231d_0)x+(-2.8034-1.9608d_0)y]$		$9E-4$
(c)	(i)	Eq. (11)		93
	(ii)	$\Delta Q=0$		32
	(iii)	$\Delta Q=-1.0613xyz$		14
	(iv)	$\Delta Q=xyz[d_0+(-2.4259-d_0)x+(-0.3512-1.4493d_0)y]$		8

$$T_{ABV} = xB_{AV} + (1-x)B_{BV} - x(1-x)C_{ABV}, \quad V = C, D,$$

$$T_{IIICD} = yB_{IIIC} + (1-y)B_{IIID} - y(1-y)C_{IIICD}, \quad \text{III} = A, B. \quad (12)$$

The quaternary parameter can be estimated in two paths with surface bowing consideration,

$$Q_{AB} = xT_{ACD}(y) + (1-x)T_{BCD}(y) - x(1-x)C_{AB},$$

$$Q_{CD} = yT_{ABC}(x) + (1-y)T_{ABD}(x) - y(1-y)C_{CD}, \quad (13)$$

where the surface bowings C_{AB} and C_{CD} incorporate errors with the factors $x(1-x)$ and $y(1-y)$, respectively, in Eqs. (13). By taking these two weighting factors into consideration, the weighted-sum expression is written as

$$Q(x,y) = \frac{Q_{CD}/y(1-y) + Q_{AB}/x(1-x)}{1/y(1-y) + 1/x(1-x)}. \quad (14)$$

It is noted that Eq. (14) is reduced to the expression of a weighted sum of four ternary alloys⁴ by ignoring the surface bowing in Eq. (13),

$$Q(x,y) = \frac{x(1-x)[yT_{ABC}(x) + (1-y)T_{ABD}(x)] + y(1-y)[xT_{ACD}(y) + (1-y)T_{BCD}(y)]}{x(1-x) + y(1-y)}, \quad (15)$$

whereas Eq. (15) is further reduced to the expression of a weighted sum of binary parameters by ignoring the ternary bowing along the boundaries,⁸

$$Q(x,y) = xyB_{AC} + x(1-y)B_{AD} + (1-x)yB_{BC} + (1-x)(1-y)B_{BD}. \quad (16)$$

Obviously, Eq. (16) is the poorest as it adopts the data at the four farthest points in the domain and ignores the nonlinear terms.

The surface bowing parameters can be estimated as

$$\begin{aligned} C_{CD} &= xC_{ACD} + (1-x)C_{BCD} - x(1-x)D_1, \\ C_{AB} &= yC_{ABC} + (1-y)C_{ABD} - y(1-y)D_2, \end{aligned} \quad (17)$$

so that $Q(x,y)$ is further deduced to

$$Q(x,y) = Q_0(x,y) + \Delta Q(x,y), \quad (18)$$

which comprises a term of fundamental estimation,

$$\begin{aligned} Q_0(x,y) &= xyB_{AC} + x(1-y)B_{AD} \\ &\quad + (1-x)yB_{BC} + (1-x)(1-y)B_{BD} - xy(1-y) \\ &\quad \times C_{ACD} - (1-x)y(1-y)C_{BCD} - x(1-x)yC_{ABC} \\ &\quad - x(1-x)(1-y)C_{ABD}, \end{aligned} \quad (19)$$

and a term of correction,

$$\Delta Q(x,y) = x(1-x)y(1-y) \left[\frac{x(1-x)D_1 + y(1-y)D_2}{x(1-x) + y(1-y)} \right]. \quad (20)$$

In Ref. 6, the surface bowing estimation [Eq. (17)] contains no terms of D_1 and D_2 , so only the fundamental estimation

FIG. 2. (Color online) Contours of the Γ band gap energy ($T=300$ K) for (a) $\text{Al}_x\text{Ga}_y\text{In}_z\text{As}$, (b) $\text{Al}_x\text{Ga}_y\text{In}_z\text{P}$, and (c) $\text{InAs}_x\text{P}_y\text{Sb}_z$ calculated using (i) the fundamental estimation, [Eq. (8)], (ii) Eqs. (7)–(10) with the single fitting coefficients listed in Table I, and (iii) weighted sum of ternary parameters at feet of perpendiculars [Eq. (11)]. The binary parameters and the bowing parameters (in brackets) of ternary and LM quaternary alloys are labeled in the diagrams.

remains for $Q(x, y)$. With $D_1=D_2$, it can be seen that Eq. (18) will be reduced to Eq. (7) in Ref. 7. The factors for D_1 and D_2 in Eq. (13) are both $x(1-x)y(1-y)$, which may lead to a different weighting consideration. However, the same fundamental estimation will still be obtained except for a different correction expression, which is also equivalent to Eq. (20) for $D_1=D_2$. In a similar consideration to Eq. (10), the following expression is also possible for a better approximation with three adjustable coefficients:

$$\Delta Q(x, y) = x(1-x)y(1-y)(d_0 + d_1x + d_2y). \quad (21)$$

A good example of interpolation using only a fundamental estimation [Eq. (19)] without correction is the band gap of $\text{Ga}_x\text{In}_{1-x}\text{As}_y\text{P}_{1-y}$. Figure 4 shows various interpolating curves

FIG. 3. Schematic diagram of a quaternary parameter surface $Q(x, y)$ of the quaternary alloy $A_xB_{1-x}C_yD_{1-y}$.

and experimental results for the band gap energy E_g of LM $\text{Ga}_x\text{In}_{1-x}\text{As}_y\text{P}_{1-y}$ ($x=0.47y$). Curve (d) is calculated using the LM formula that has compromised experimental data [Eq. (5.4) in Ref. 1]. Curve (a), i.e., using Eq. (15), apparently has the largest deviation from experimental data, whereas curve (c), i.e., the fundamental estimation using Eq. (19), is in excellent agreement to experimental data with only a maximum deviation of 2 meV from the LM formula. The band gaps for the binary compounds GaAs, InAs, GaP, and InP are 1.4225, 0.3521, 2.777, and 1.3529 eV, respectively, and the bowing parameters for the ternary alloys GaInAs, GaInP, GaAsP, and InAsP are 0.477, 0.65, 0.19, and 0.1, respectively.¹

Similar to the discussion in Sec. II, the Γ band gap energy of $\text{Ga}_x\text{In}_{1-x}\text{As}_y\text{Sb}_{1-y}$ is fitted to the three LM formulas

FIG. 4. (Color online) Various formulas for $\text{Ga}_x\text{In}_{1-x}\text{As}_y\text{P}_{1-y}$: (a) weighted sum of four ternary alloys [Eq. (15)]; (b) $E_g = 1.35 + 0.668x - 1.068y + 0.758x^2 + 0.078y^2 - 0.069xy - 0.332x^2y + 0.03xy^2$ from Ref. 9; (c) fundamental estimation [Eq. (19)]; (d) $E_g = 1.353(1-y) + 0.737y - 0.13y(1-y)$, Eq. (5.4) in Ref. 1; (e) $E_g = 1.35 + (0.758x + 0.642)y + (0.101x - 1.101)y - (0.28x - 0.109y + 0.159)xy$ in Ref. 6; (f) $E_g = 1.35 + 0.668x - 1.17y + 0.758x^2 + 0.18y^2 - 0.069xy - 0.322x^2y + 0.03xy^2$ in Ref. 10 in contrast to experimental data collected in (g) Ref. 11, (h) Ref. 10, and (i) Ref. 12 under LM condition $x=0.47z$ and $y=z$. The base line has been subtracted from the linear Vegard law $E_g = 1.353(1-y) + 0.736y$.

TABLE II. Maximum $|E_g(x, y) - E_g^{(LM)}|$ for $\text{Ga}_x\text{In}_{1-x}\text{As}_y\text{Sb}_{1-y}$ ($T=300$ K) using (i) weighted sum ignoring surface bowing [Eq. (15)] and considering surface bowing [Eq. (18)] with (ii) none (fundamental estimation), (iii) single, (iv) two [Eq. (20)], and (v) three fitting coefficients. LM to InP: $(\text{GaAs}_{0.5}\text{Sb}_{0.5})_z(\text{Ga}_{0.47}\text{In}_{0.53}\text{As})_{1-z}$. LM to InAs: $(\text{GaAs}_{0.08}\text{Sb}_{0.92})_z(\text{InAs})_{1-z}$. LM to GaSb: $(\text{GaSb})_z(\text{InAs}_{0.91}\text{Sb}_{0.09})_{1-z}$.

Equation and condition		$\max(E_g(x, y) - E_g^{(LM)})$ (meV)		
		LM to InP	LM to InAs	LM to GaSb
(i)	Fundamental estimation [Eq. (19) and $\Delta Q=0$]	10	68	57
(ii)	$\Delta Q=0.6215x(1-x)y(1-y)$	31	42	37
(iii)	$\Delta Q=x(1-x)y(1-y)\left[\frac{0.0141x(1-x)+1.2282y(1-y)}{x(1-x)+y(1-y)}\right]$	30	40	38
(iv)	$\Delta Q=x(1-x)y(1-y)(2.5276-0.1755x-3.3367y)$	9	17	19
(v)	Eq. (15)	169	136	151

adopting bowing parameters recommended in Ref. 1. Table II lists the maximum deviations from the LM formulas at three LM conditions using single, two, and three fitting coefficients in Eqs. (18)–(21) in contrast to the results using the weighted-sum approach. Again, we see that the fundamental estimation [Eq. (19)] is far superior to the weighted-sum of four ternary alloys [Eq. (15)]. There remains 30–40 meV deviation after the corrections with either single or two fitting coefficients [see (ii) and (iii) in Table II], whereas three adjustable coefficients have to be used to reduce the deviation below 20 meV, which is comparable only to the results of single fitting coefficients listed in Table I. This shows that the $A_xB_{1-x}C_yD_{1-y}$ type of quaternary alloys have more complicated alloy parameter surfaces than $A_xB_yC_zD$ due to the relatively larger domain and one more corner. The fitting curves with the corresponding conditions listed in Table II

are plotted for the three LM cases as shown in Fig. 5 in contrast to the experimental results available in the literatures.^{13–18} The experimental data points are marked on the contour diagram shown in Fig. 6. It can be seen that although the fundamental estimation overestimates the quaternary bowing,^{1,19,20} it still approaches the experimental data better than the weighted-sum method ignoring surface bowing.

IV. SUMMARY

In summary, the weighted-sum expression of quaternary parameter estimations via balancing surface bowing estimation errors is shown to be effective for both types of quaternary alloys $A_xB_yC_zD$ and $A_xB_{1-x}C_yD_{1-y}$. Without further data fitting, the fundamental estimation is a better approximation than the weighted-sum schemes^{4,5} that were ever reported. With the correction terms, explicit molar fraction expressions of the band gaps of AlGaInAs, AlGaInP, InAsPSb, GaInAsP,

FIG. 5. (Color online) Fitting curves (i)–(v) calculated using the corresponding conditions listed in Table II in contrast to the experimental data.

FIG. 6. (Color online) Contours (i)–(iv) of the Γ band gap energy ($T=300$ K) of $\text{Ga}_x\text{In}_{1-x}\text{As}_y\text{Sb}_{1-y}$ calculated using conditions listed in Table II. The binary parameters and the bowing parameters (in brackets) of ternary and LM quaternary alloys labeled in the diagram are recommended in Ref. 1. The experimental data points used for comparison in Fig. 5 are marked as well.

and GaInAsSb can be obtained within less than 20 meV deviation to the LM polynomial expressions for LM compositions.

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