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Electronic energy levels in $Ga_{1-x}Al_xSb$ alloys

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Piezomodulation spectra are reported for $Ga_{1-x}Al_xSb$ alloys. E_0, $E_0+\Delta_0$, E_1, $E_1+\Delta_1$ direct transitions and the lowest indirect transitions are observed as a function of temperature and alloy composition. At room temperature, the direct-indirect crossover occurs at $x = 0.4 \pm 0.02$. The bowing parameters for the E_0 and E_1 gaps are 0.69 and 0.28 eV, respectively, and that for $\Gamma \rightarrow X$ indirect transitions is 0.48 eV. The spin-orbit splittings Δ_0 and Δ_1 are found to vary linearly with composition. Between 100 and 300°K the temperature dependence of various gaps is found to be linear. The coefficient of temperature dependence varies nearly linearly with composition. Theoretical calculations of bowing are examined. It is concluded that uncertainties in the material parameters of the pure compounds are the main cause which makes theoretical prediction of bowing parameters and crossover compositions rather uncertain.

I. INTRODUCTION

The composition dependence of energy gaps in substitutional semiconducting alloys has attracted considerable attention in recent years.[1] Of particular interest are alloys in which the lowest band edge changes from direct to indirect transitions at a certain composition. The variation of a given energy gap E_g with fractional composition x is generally expressed by a parabolic equation

$$E_g(x) = a + bx + cx(x-1), \qquad (1)$$

where the parameter c depicting the departure from linearity is called the bowing parameter. Theoretical calculations based on the empirical pseudopotential method[2,3] (EPM) and the dielectric method[4] (DM), both reproduce the basic trend in the bowing parameters of various alloys. In both calculations, as well as in experiments, the alloys with one of the two intersubstituted atoms belonging to the fourth row show a larger departure from linearity. The two theoretical approaches, however, differ remarkably in their conclusions. The EPM calculations are based on the virtual-crystal approximation (VCA), and their success is taken to indicate the validity of VCA. Van Vechten and Bergstresser (VB) using the DM, however, concluded that most of the bowing was due to chemical disorder in these alloys. This disorder-induced or "extrinsic" bowing was argued to be proportional to $(1/r_F - 1/r_G)^2$, where r_F and r_G are the covalent radii of the two intersubstituted atoms. Using EPM, Hill[5] found a similar relation between bowing and covalent radii in the VCA. In view of this importance of the covalent radii in determining the bowing, alloys like $Ga_{1-x}Al_xAs$ and $Ga_{1-x}Al_xSb$ attain special importance because for these the difference between r_F and r_G is negligible. $Ga_{1-x}Al_xAs$ has been studied in conventional absorption[6] as well as modulation spectroscopy.[7] The experimental bowing was found to be 0.26 eV compared to 0.03 eV predicted by VB. The bowing in $Ga_{1-x}Al_xSb$ has been estimated[8] to be larger than this, but no definitive results have so far been available.[9] The present work reports a detailed study of the electronic energy levels in $Ga_{1-x}Al_xSb$ alloys.

In Sec. II, we present piezomodulation spectra of $Ga_{1-x}Al_xSb$ alloys over a wide range of energy and temperature. We analyze the composition dependence of various energy gaps in Sec. III, and show that even in the DM uncertainties in the material parameters of constituent compounds, as well as those in the depiction of VCA, are substantial. This is particularly important in view of the recent work[5,10-12] on the importance of chemical disorder in energy-gap bowing.

Finally, our conclusions are presented in Sec. IV where we also present a summary of our results.

II. EXPERIMENTAL RESULTS

The samples used in our work were p-type monocrystalline layers grown on p-type GaSb single-crystal substrates, by liquid-phase epitaxy.[13] The free surface of each layer was polished to optical flatness and was used for reflection studies. The GaSb substrate was polished away leaving a fairly reflecting back surface. The samples, about 30 to 60 μm thick and with a surface area 3×3 mm^2, were cemented onto a ceramic with a hole in the center to allow modulated transmission measurements. The composition of these crystals were determined by electron microprobe as well as by x-ray diffraction on powder samples. The two results agreed quite well and gave $x=0.16$, 0.28, 0.40, and 0.56 for the first four samples. Only x-ray results could be obtained for the fifth one, to obtain $x=0.68$. Generally, the values of x are expected to be accurate to within 0.02. Details of the experimental setup for piezomodulation stud-

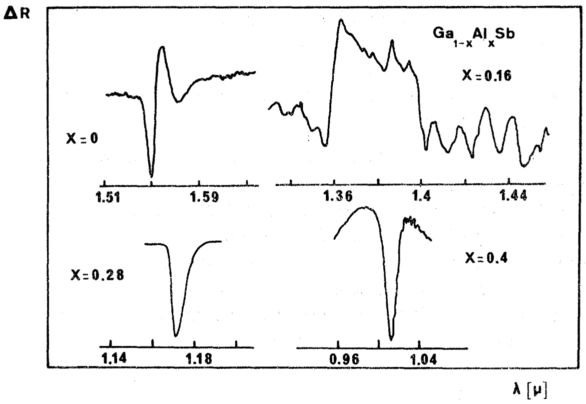

FIG. 1. Piezoreflection spectra at 100 °K in the range of the E_0 gap.

ies have been described elsewhere[14] and will not be repeated here.

The piezoreflection spectra at 100 °K are shown in Fig. 1 for $x=0$, 0.16, 0.28, and 0.4, respectively. The lowest structures shown here are due to direct transitions to the lowest conduction band at Γ. This assignment is based on the spectral shapes, and was confirmed by studying the temperature dependence of these line shapes. The corresponding spectra at 300 °K have been shown in a brief communication[9] earlier. It was found that all these spectra shift and broaden slightly but retain their shape as the temperature is increased. The spectrum for GaSb ($x=0$) is typical for a steplike excitonic edge in the absorption coefficient. After accounting for the binding energy this gives $E_0=0.799$ eV at 100 °K. Owing to a larger concentration of holes the spectra for alloys do not show an excitonic edge. The line shape here corresponds to that for an M_0 critical point for $x=0.28$ and $x=0.4$. The transition energy is then close to the peak in $\delta R/R$. The correction due to broadening of the structure is $\frac{1}{4}\sqrt{3}\Delta E$, where ΔE is the half-width of the structure at half-maximum[15] on the higher-energy side. This correction, typically of the order of a few meV, is ignored in our analysis. In the spectrum for $x=0.16$ the low-energy side is masked by electronic transitions involving impurities. The results obtained from piezotransmission measurements were found to be in agreement with those reported above. The temperature dependence for the E_0 gap for several x is shown in Fig. 2.

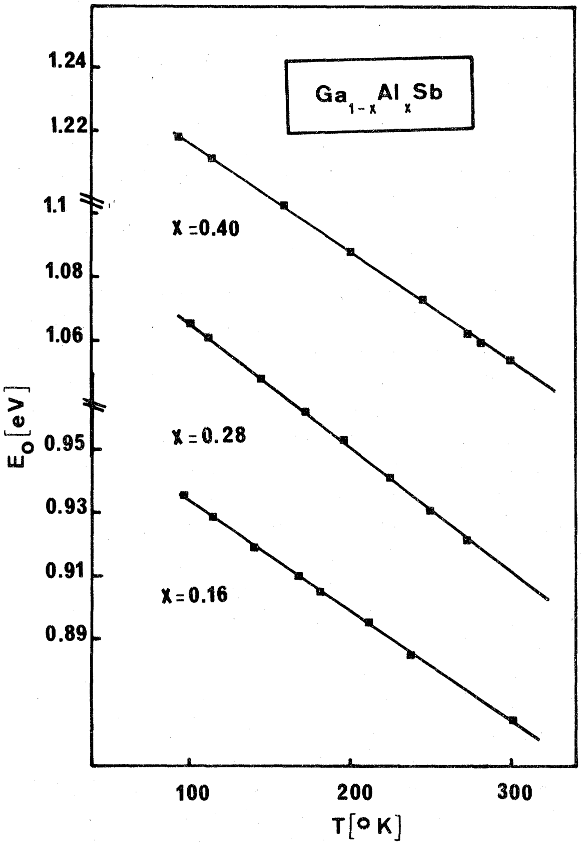

FIG. 2. Temperature dependence of the E_0 gap.

For $x>0.4$, the lowest interband transitions are indirect. For $x=0.56$ and $x=1$, the piezotransmission spectra at 100 °K are shown in Fig. 3. At higher temperatures[9] the structures become broader and show phonon emission as well as absorption. In AlSb the lowest indirect transition is from

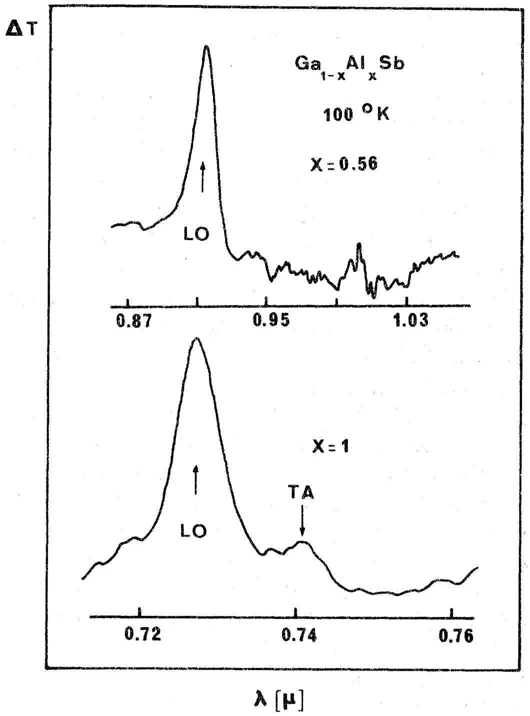

FIG. 3. Piezotransmission spectra at the indirect band edge at 100 °K.

top of the valence band at Γ to the conduction-band minimum near X. From band-structure calculations, the $\Gamma\rightarrow L$ transition energy is also close to the $\Gamma\rightarrow X$ transition energy, but no definite experimental verification of this, has been possible[16] because $\Gamma\rightarrow L$ transitions are relatively weakly modulated in piezotransmission. Since the $\Gamma\rightarrow X$ transition energy is higher than the $\Gamma\rightarrow L$ one in GaSb, one might expect $\Gamma\rightarrow L$ transitions to be the lowest indirect edge in a small range of x around $x\sim 0.5$. We identify the main peak in the $x=0.56$ and $x=0.68$ spectra as the phonon-assisted transitions from $\Gamma\rightarrow X$ because these transitions are expected to be the strongest. For a coplanar (111) stress, the ratio of the piezomodulation parameters for the $\Gamma\rightarrow X$ and $\Gamma\rightarrow L$ indirect transitions via the conduction state Γ_{1C} may be written

$$\frac{\Delta(\Gamma\rightarrow L)}{\Delta(\Gamma\rightarrow X)}\simeq\frac{2\alpha_L+\delta}{2\alpha_X+\delta},$$

where $\alpha_{X(L)}=a_{X(L)}(S_{11}+2S_{12})$ and $\delta=dS_{44}/2\sqrt{3}$. Here $a_{X(L)}$ denotes the hydrostatic deformation potential, d is the shear deformation potential, and S_{ij} is the compliance tensor. Using the values given in literature[17] for deformation potentials in pure compounds and linearly interpolating them for the alloys, we find this ratio to about $\frac{1}{5}$ in $Al_{0.5}Ga_{0.5}Sb$. Since α_L and δ have opposite signs whereas α_x and δ have the same sign, it is clear that the above ratio is much smaller than one, even though we cannot estimate it very accurately. The structures in $x=0.56$ spectrum on the lower-energy side of the main peak, could be identified as phonon-assisted $\Gamma\rightarrow L$ transitions. Unfortunately, a verification of this was not possible due to the lack of samples in the required composition range. In GaSb and AlSb, indirect transition $\Gamma\rightarrow X$ involves only LO (X) phonons. Thus, we attribute the main structure in the spectra in Fig. 3 to indirect transitions involving emission of LO phonons. In the spectrum for AlSb, the other structure is attributed to the emission of TA phonons. Knowing the excitonic binding energy and the LO phonons frequencies (10 and 36 meV, respectively, for[18] AlSb) one can obtain the band gap $\Gamma\rightarrow X$ in these alloys. As these corrections are small and their interpolation uncertain, the transition energies corresponding to the main peak are used for determining the bowing for indirect $\Gamma\rightarrow X$ transitions. At 300 °K, these are 1.02, 1.264, 1.313, and 1.630 eV for $x=0$, 0.56, 0.68, and 1, respectively. These values are well represented by the equation

$$E_{\text{ind}}(x)=1.02+0.61x+0.48x(x-1)\,, \qquad (2)$$

with a maximum probable error of 0.04 eV in the bowing parameter.

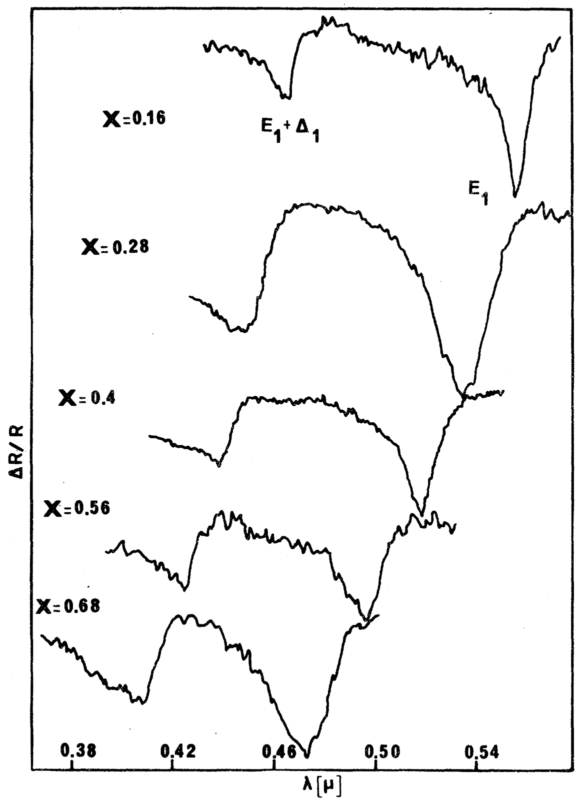

FIG. 4. Modulated reflectivity spectra corresponding to E_1 and $E_1+\Delta_1$ transitions, $T=100$ °K.

The direct transitions E_1 and $E_1+\Delta_1$ could be observed over the entire composition range owing to the large joint density of states associated with these transitions. The piezoreflection spectra in this energy range are shown in Fig. 4 at 100 °K. The widths of these structures are similar in alloys and in the pure compounds. Figure 5 shows the energy gap E_1 as a function of composition at 100 and 300 °K. The room-temperature data are

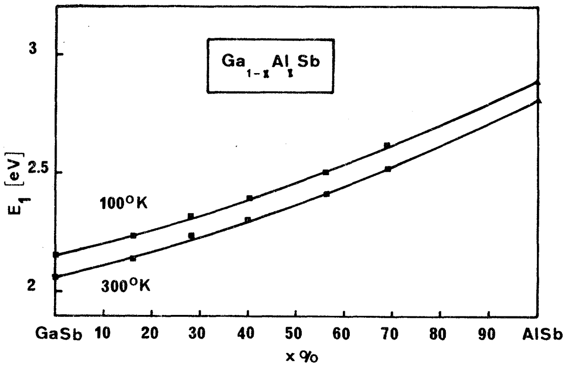

FIG. 5. Composition dependence of E_1 gap at 100 and 300 °K. Experimental points: ■, present work; ▲, M. Cardona, K. L. Shaklee, and F. H. Pollak, Phys. Rev. 154, 696 (1967).

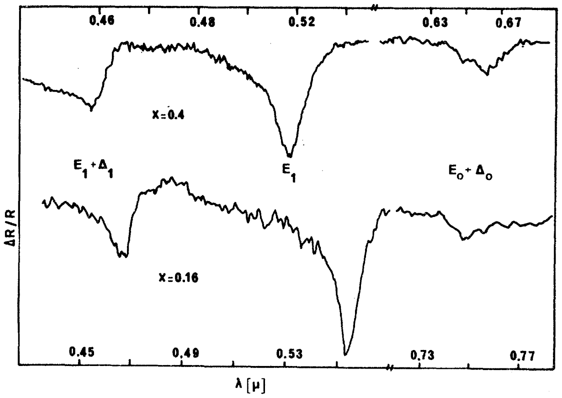

FIG. 6. Modulated reflectivity spectra showing the weak $E_0+\Delta_0$ transitions. For comparison E_1 and $E_1+\Delta_1$ transitions are also included. $T=100$ °K.

fitted by the equation

$$E_1(x)=2.06+0.75x+0.28x(x-1). \qquad (3)$$

The spin-orbit splitting Δ_1 is a linear function of x and will be discussed along with Δ_0.

The $E_0+\Delta_0$ transitions are associated with a relatively small joint density of states and are somewhat difficult to observe. Piezoreflection spectra recorded at 100 °K show these transitions quite clearly for $x=0.16$ and $x=0.4$ and are reproduced in Fig. 6. For higher x this transition becomes too close to the E_1 gap to allow clear resolution. The composition dependence of Δ_0 and Δ_1 is shown in Fig. 7 where Δ_1 is seen to vary linearly with x. Since for $x=0$, 0.16, and 0.4, Δ_0 is very close to $\frac{3}{2}\Delta_1$, and since the bowings in Δ_0 and Δ_1 are generally related to each other, it seems reasonable to extrapolate Δ_0 also linearly with x. This gives $\Delta_0\simeq 0.6$ eV for AlSb, a value considerably lower than 0.75 eV obtained by Braunstein and Kane[19] from measurement of absorption due to interband transitions between valence bands in a p-type material. On a reanalysis of their data, however, we found[20] that $\Delta_0\simeq 0.6$ eV would be more consistent with their data than $\Delta_0=0.75$ eV. The argument is based on the observation that Δ_0 is the *short-wavelength* limit of $V_3\rightarrow V_2$ and the *long-wavelength* limit of $V_3\rightarrow V_1$ transition, where V_1, V_2, and V_3 denote the heavy-hole, the light-hole, and the split-off band, respectively. Because, the heavy-hole band has a much larger density of states, the $V_3\rightarrow V_1$ band is expected to be much stronger than the $V_3\rightarrow V_2$ band. This was indeed found to be the case in GaAs where both bands could be identified. In AlSb, however, only one band was observed and it would be reasonable to assume this to be the $V_3\rightarrow V_1$ band. Δ_0 would then be close to the onset of this band, i.e., at about 0.6 eV. We have now confirmed this interpretation by observing these intervalence-band transitions in piezotransmission spectra.[20] We found $\Delta_0(\mathrm{AlSb})=0.63\pm 0.02$ eV. This would indicate a very-small bowing on Δ_0 which is negligible within the experimental errors of 0.04 eV.

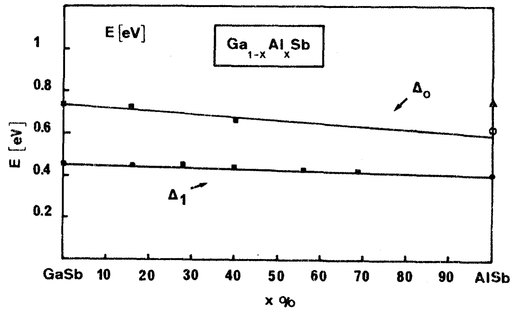

FIG. 7. Composition dependence of spin-orbit splittings Δ_0 and Δ_1. Experimental points: ▲, Ref. 19; ○, Ref. 20; ●, M. Cardona, K. L. Shaklee, and F. H. Pollak, Phys. Rev. 154, 696 (1967). ■, present work. The linear interpolation of Δ_0 is indicated.

The composition dependence of E_0 gap and the $\Gamma\rightarrow X$ indirect transition energies are shown in Fig. 8 at 100 and 300 °K. The E_0 gap at 300 °K satisfies the relation

$$E_0(x)=0.721+1.479x+0.69x(x-1). \qquad (4)$$

Within the quoted experimental errors the bowing was found to be temperature independent, but the crossover composition would change from 0.40 at 300 °K to 0.44 at 100 °K. It would be interesting to verify this directly.

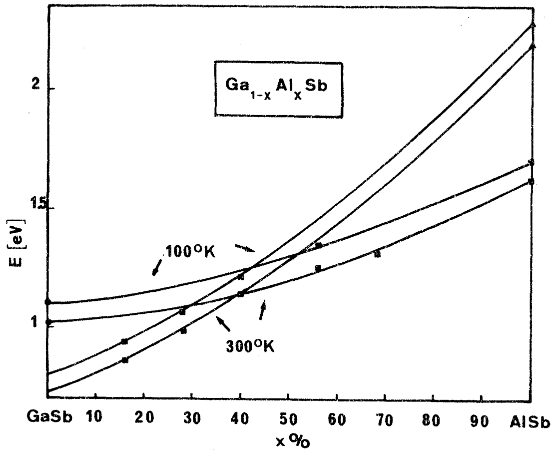

FIG. 8. Composition dependence of the E_0 direct gap and the $E_{\mathrm{ind}}(\Gamma\rightarrow X)$ indirect gap. Experimental points: ■, present work; ●, B. B. Kosichi, A. Jayaraman, and W. Paul, Phys. Rev. 172, 764 (1968).

III. DISCUSSION

As mentioned earlier, two reasonably successful methods have been used so far to calculate the bowing parameters in substitutional alloys. The EPM calculations by Richardson[3] using a reasonable method for obtaining the form factors gives a bowing parameter of 0.05 eV for the E_0 gap. Our experimental value 0.69 eV is about fourteen times larger. Since in most other cases Richardson obtains good agreement, we feel that this disagreement does not indicate a failure of the VCA or the method of interpolation of form factors. The most likely cause appears to be the inaccurate form factors for GaSb. Richardson[3] used the form factors given earlier by Cohen and Bergstresser[21] which are known to be unsatisfactory.[22] This might also be the cause of the EPM result, in GaInSb (0.24 eV) being smaller than the experimental value[15] 0.42 eV.

Using the empirical dielectric theory VB predicted an intrinsic bowing of 0.19 eV for the E_0 gap in $Ga_{1-x}Al_xSb$. The "extrinsic" bowing is estimated to be negligible because the covalent radii of Al and Ga are very nearly equal. The fact that Δ_0 does not show any appreciable bowing $Ga_{1-x}Al_xSb$ and $Ga_{1-x}Al_xAs$ appears to support this. Nevertheless, the total bowing predicted by VB for both these alloys is much smaller than the experimental values. We investigated two possible reasons for this disagreement. First, the material parameters of the pure compounds used as input may be inadequately known. Second, the average potential in the VCA is ill-defined. We found that the DM suffers from these uncertainties as much as the EPM does. We note that the correction due to the presence of d electrons is treated in a rather arbitrary manner. It is difficult to understand why D_{av} and D, introduced by Van Vechten[23,24] should differ. Since the values of D were anyway obtained from a prescription,[23] we decided to use only one D parameter which is adjusted to reproduce the E_0 band gap in pure compounds. With this stipulation we examined the other two uncertainties mentioned above.

The material parameters of relevance for AlAs, GaAs, AlSb, and GaSb are listed in Table I. The values of E_0, $E_{ind}(\Gamma\rightarrow X)$, and $E_{ind}(\Gamma\rightarrow L)$ gaps are those at room temperature. Whenever, reliable refractive index measurements below the band edge are available, they are used to obtain $\epsilon(0)$. The effects of dispersion are accounted for by using the single oscillator fits by the method of Wemple and Di Domenico,[25] the goodness of such a fit is indicative of the accuracy of the data. $\epsilon(0)$ obtained in this way often differs from that obtained by the restrahlen measurements by as much as 10%. Since the fits to the restrahlen data are made using a somewhat oversimplified classical-oscillator model with Lorentzian broadening, these

TABLE I. Physical parameters of pure compounds used in calculations of bowing parameters. Energy gaps are given at room temperature.

Compound	Lattice constant (Å)	$\epsilon(0)$	E_0 (eV)	Δ_0 (eV)	E_1 (eV)
	a			k	
AlAs	5.6622	8.16^b	2.95^g	0.28	3.9^g
AlSb	6.1355	9.9^c 8.2^d	2.22^h	0.63	2.81^h
GaAs	5.6532	10.89^e	1.42^i	0.34	2.89^l
GaSb	6.0954	$13.7^{d,f}$	0.721^j	0.74	2.059^m

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deviations are not surprising. And yet, we found that such uncertainties in $\epsilon(0)$ of pure compounds cause considerable uncertainties in the calculation of bowing parameter in the DM. For AlSb, we have given two values of $\epsilon(0)$. We obtained the value 8.2 by measuring the reflectivity in the range of 2–3 μm. This is given as a tentative value because the accuracy of our measurement was only about 1% in reflectivity, i.e., not enough to detect the effects of dispersions and because the sample was relatively heavily doped (~10^{18} holes cm^{-3}).

The second type of uncertainties, i.e., ambiguities in the prescription of the average potential, refer mostly to the parameter D and the average heteropolar gap C. For $D-1 \ll 1$, inaccuracies in the interpolation of D may be assumed unimportant, and D may be interpolated linearly. To obtain C then, one may interpolate either C, or the ionicity[26] f_i or $\epsilon(0)$. The values of bowing parameter for the E_0 gap calculated using these three methods of interpolation are listed in Table II. We observe that, at least in the cases considered here, the effects of uncertainties in the method of interpolation are much less important than those in the parameters of the pure compounds. This may not be the case for many other alloys. For $Ga_{1-x}Al_xAs$, accurate measurements of refractive index have recently become available.[27] The accuracy of the experiments do not permit us to distinguish clearly between the three alternatives in this case. Accurate measurements of $\epsilon(0)$ for $Ga_{1-x}Al_xSb$ alloys would clearly be of interest. In particular we note that a small upward bowing in $\epsilon(0)$ will be enough to make the calculated bowing agree with experiment. In principle, the dielectric theory needs no new adjustment to predict the E_1 gap for all alloys. However, E_1 gap in AlSb is the worst failure of this theory,[23] and the disagreement becomes even worse if the lower value of $\epsilon(0)$ is used. The values of bowing for E_1 gap listed in Table II are obtained when D has been readjusted to obtain the E_1 gaps in the pure compounds. For the indirect gap $E_{ind}(\Gamma \rightarrow X)$, no calculation of the bowing was attempted because this gap is not well represented in the dielectric method.

TABLE II. Experimental and calculated bowing parameters in $Ga_{1-x}Al_xSb$ and $Ga_{1-x}Al_xAs$. (i), (ii), and (iii) refer to calculations with linear interpolation of C, $\epsilon(0)$, and f_i, respectively. (a) and (b) correspond, respectively, to the two values 8.2 and 9.9 of $\epsilon(0)$ of AlSb. The experimental data for Al-GaAs are from Refs. 6 and 7.

	Bowing parameter (eV)			
		Theory		
Transition	Expt.	i	ii	iii
E_0 ($Ga_{1-x}Al_xSb$)	0.69	(a) 0.33	0.45	0.42
		(b) 0.18	0.22	0.19
E_1 ($Ga_{1-x}Al_xSb$)	0.28	(a) 0.05	0.28	0.31
		(b) 0.08	0.17	0.15
E_0 ($Ga_{1-x}Al_xAs$)	0.26± 0.02	0.19	0.25	0.22
E_1 ($Ga_{1-x}Al_xAs$)	0.45± 0.1	0.11	0.20	0.17

Using the experimentally determined energy gaps and linearly interpolated momentum matrix elements, effective masses and g factors can be calculated using the $\vec{k}\cdot\vec{p}$ perturbation theory. Following Cardona,[28] we include the lowest two conduction bands and the highest (p-type) valence bands in the $\vec{k}\cdot\vec{p}$ basis set. We find effective-mass and g-factor values which are well represented by the equations

$$m_c = 0.042 + 0.08x + 0.048x(x-1),$$

$$g_c = -6.67 + 7.38x - 6.2x(x-1),$$

$$m_{1h} = 0.046 + 0.058x + 0.012x(x-1).$$

We note here that the effect of including the higher conduction band is not negligible in these alloys. Except for alloys with very-small effective masses, this effect is of the same order of magnitude as that due to the disorder-induced mixing as proposed recently by Berolo *et al.*[29]

IV. CONCLUSIONS

We have measured piezomodulated optical spectra of $Ga_{1-x}Al_xSb$ alloys. The composition dependence of energy gaps can be well represented by the usual parabolic equation. The bowing parameters for the E_0, E_1, and $E_{ind}(\Gamma \rightarrow X)$ gap are 0.69, 0.28, and 0.48 eV, respectively. Since AlSb and GaSb have almost the same lattice parameters, these alloys along with those of the $Ga_{1-x}Al_xAs$ system are ideal to explore the application of the VCA in dielectric method. We have shown that even in the dielectric method, as in the EPM, the calculation of bowing parameter is rather sensitive to the accuracy in material parameters, especially $\epsilon(0)$, for the pure compounds. The uncertainty as to which parameters may be interpolated would be considerably smaller if experimentally determined $\epsilon(0)$ were available. For example, a small convex bowing in $\epsilon(0)$ would suffice to reproduce the experimental bowing in $Ga_{1-x}Al_xSb$. It follows that accurate determinations of refractive indices for alloys, until now studied mainly for technological applications, would also help to define the virtual crystal approximation better.

ACKNOWLEDGMENTS

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*Associé au Centre National de la Recherche Scientifique.

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