

Indirect-energy-gap dependence on Al concentration in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys

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The energy of the indirect X exciton in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ epitaxial layers (with $0.38 < x < 0.81$) has been determined by photoluminescence measurements at a sample temperature of 12 K; from these data, the dependence on the Al concentration x of the energy of the Γ - X gap has been obtained: $E_g^X(x) = 1.988 + 0.207x + 0.055x^2$ eV. This relation is characterized by a very small bowing. From our data we deduce also the coordinates of the Γ - X crossover point: $x_c = 0.385 \pm 0.016$ and $E_c = 2076 \pm 4$ meV at $T = 0$ K and $x_c = 0.396 \pm 0.016$ and $E_c = 1997 \pm 4$ meV at $T = 300$ K.

I. INTRODUCTION

The knowledge of the fundamental energy gap of III-V compounds and alloys is of great importance for the study and fabrication of heterostructures and devices. The conduction-band minima of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ are at the Γ , X , and L points of the Brillouin zone. In Ga-rich compounds ($x < 0.4$) the absolute minimum is at Γ and the alloys are characterized by a direct gap; at higher Al concentrations the system becomes indirect, the absolute conduction-band minimum being at (or close to) the X point of the Brillouin zone. The L minimum is never the absolute conduction-band minimum for the system.¹ The maximum of the valence band remains at Γ for all the Al concentrations.

Recently we determined the dependence of the direct Γ - Γ energy gap on the Al concentration x .² In Ref. 2 we showed that the $E_g^\Gamma(x)$ dependence that was commonly used for the optical determination of the alloy concentration³ must be revised. The main modification we found is a strong reduction of the value of the bowing parameter: 0.22 ± 0.06 (Ref. 2) instead of 1.147 (Ref. 3); a small quadratic term in the $E_g^\Gamma(x)$ dependence is also expected from theoretical considerations.⁴⁻⁷ The problem of a correct measurement of the Al concentration in the alloy emerged as the main point in the determination of a consistent $E_g(x)$ relation.

The situation for the indirect gap Γ - X is still unclear,¹ despite the fact that for high Al concentration the indirect transitions related to this minimum become detectable by spectroscopic techniques. The indirect exciton photoluminescence (PL) has been measured at low temperatures for Al concentration x up to 0.98;⁸ transitions related to the indirect minimum are reported also in Ref. 9 for $0.35 < x < 0.90$ and in Ref. 10 for $0.4 < x < 0.6$. In

these papers, however, no analytical dependence of the gap energy on the Al concentration is proposed. Other systematic PL measurements^{11,12} have been performed on a limited Al concentration range. Formulas for $E_g^X(x)$, valid at room temperature, are reported in Refs. 3, 13, and 14.

Very little is known on the indirect Γ - L gap.¹ An evaluation of $E_g^L(x)$ in the range $0.2 < x < 0.55$ has been obtained from photoluminescence excitation measurements;¹⁵ other estimations have been proposed from the analysis of high-temperature transport measurements.^{13,14} A discussion of the available data is reported in Ref. 15.

From the situation described in this synthetic summary, it results that an accurate determination of the energy of the Γ - X and Γ - L gaps in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ is highly desirable. Here we present a determination of $E_g^X(x)$ obtained through low-temperature PL measurements performed on samples whose composition has been determined on the basis of our previous work.²

II. EXPERIMENTAL METHODS

We performed PL measurements in the temperature range between 12 and 300 K on $\text{Al}_x\text{Ga}_{1-x}\text{As}$ samples grown by liquid-phase epitaxy and having Al concentrations x between 0.38 and 0.81. The characteristics and the quality of the samples have been discussed in our previous paper.²

The PL measurements were performed with a cryocooler; the sample, temperature was measured with an Au:Fe versus chromel thermocouple in good thermal contact with the sample. The PL was excited with different lines of an Ar^+ laser; an exciting power between 50 and 150 mW corresponding to an exciting power density up to several hundreds of W/cm^2 was used. At the highest excitation intensities some heating of the sample

is possible. To evaluate the sample heating, we measured the PL spectrum at different excitation intensities. At low temperatures ($T < 30$ K), the increase of the excitation intensity from a few W/cm^2 to $800 \text{ W}/\text{cm}^2$ did not induce any shift of the spectral structures; in fact, at these temperatures the energy gap dependence on T is very modest (see below). At $T > 80$ K for the same variation of the exciting intensity we observed a shift of about 1–2 meV, corresponding to a maximum heating of the excited region of about 5 K. In the following, however, the sample temperature as measured by the thermocouple is quoted in the text and in the figures. The PL was focused on the entrance slit of a 0.5-m grating monochromator and detected by a cooled GaAs cathode photomultiplier tube connected to a lock-in amplifier. The resolution was better than 0.4 nm for the low-temperature measurements and better than 3.2 nm for the high-temperature measurements. The shape and the noise of the spectra give rise to the uncertainties on the energy values that will be quoted hereafter together with the energy values. A few PL and transmission measurements have been made at $T = 2$ K using the experimental setup described in Ref. 2.

III. RESULTS

A. Low-temperature photoluminescence spectra

The low-temperature PL spectra of some samples are shown in Fig. 1. Their shape is typical of good-quality indirect-gap $\text{Al}_x\text{Ga}_{1-x}\text{As}$.^{16,17} Following Refs. 16 and 17, the peak at the highest energy is attributed to the recombination of bound excitons associated mainly with donors, and we call it (D^0, X) . At high Al concentrations

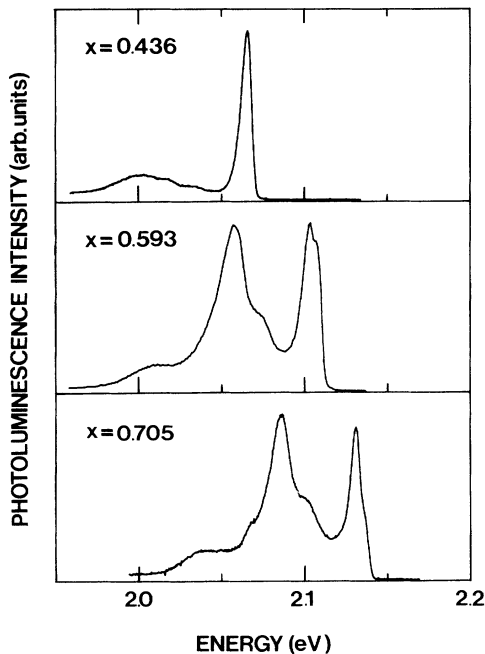


FIG. 1. Photoluminescence spectra at $T = 12$ K of different $\text{Al}_x\text{Ga}_{1-x}\text{As}$ samples; the Al concentration x is indicated. The exciting power density is $\sim 120 \text{ W}/\text{cm}^2$.

this peak splits into a doublet. A similar structure has also been observed in PL spectra of GaAs measured under high hydrostatic pressure and has been attributed to two different residual donor species.¹⁸ The (D^0, X) recombination is a zero-phonon process that becomes allowed in the indirect-gap alloy due to the influence of impurity and alloy scattering. The two other structures, distant ~ 30 and ~ 45 meV from the (D^0, X) transition, are ascribed to phonon replica of the donor bound exciton recombination. Finally, the broad structures appearing at low energy could be associated with impurity energy levels; they are possibly due to donor-acceptor pair recombinations and their phonon replica. We do not discuss the attribution of all the structures of the spectra, and we will concentrate on the exciton recombination characterizing the high-energy side of the PL spectra.

B. Determination of the sample composition

The Al concentration x at the point where the PL has been measured was determined using the dependence of the direct exciton energy E_x^Γ on x determined in our previous work.² On some samples, photoluminescence and transmission spectra have been measured at $T = 2$ K on the same place. The Al concentration at this place was obtained from the energy of the direct exciton measured in absorption using Eq. (1) of Ref. 2 for $E_x^\Gamma(x)$. From the photoluminescence spectra, which are similar to those shown in Fig. 1, we read the energy of the donor bound exciton (D^0, X) . The dependence of these energies on the Al concentration x is within the experimental accuracy a straight line; a least-squares fit yields

$$E_{(D^0, X)}(x) = 1.957 + 0.25x \text{ eV} \quad (0.38 < x < 0.81). \quad (1)$$

The errors on the constant term and on the linear coefficient are ± 0.006 and ± 0.01 eV, respectively; the estimated standard deviation¹⁹ is 1.3 meV. On other samples we made only PL measurements. The Al concentration of these samples was determined by reading $E_{(D^0, X)}$ from the spectra and by using Eq. (1). The values of x obtained this way are reported in Table I together with the corresponding uncertainties.

The procedure we used for the determination of the sample composition is summarized in Fig. 2. Only one line for $E_{(D^0, X)}$ is reported in Fig. 2. In fact, the (D^0, X) doublet is not resolved in the PL spectra measured where the GaAs substrate has been etched away, probably due to stress on $\text{Al}_x\text{Ga}_{1-x}\text{As}$. In the case where a (D^0, X) doublet is present we used for $E_{(D^0, X)}$ the average of the energies of the two components.

Finally, we observe that a source of indetermination is intrinsically present in the method we used. The transmission measurements give transition energies average over the whole depth of the layer (typically 2–3 μm), whereas the PL measurements probe the layer mainly at the surface, for a depth of some tenths of μm . However, due to the small slope of the $E_{(D^0, X)}$ dependence on x , this indetermination is not important.

TABLE I. Aluminum concentration x and indirect exciton energy E_x^X at $T = 12$ K of our samples as determined by photoluminescence measurements. The calculated exciton Rydberg E_b^X [Eq. (2)] and the corresponding energy gap E_g^X are reported. Values of the energy gap for GaAs (Ref. 1) and for AlAs (see text) have been added. Experimental uncertainties Δx and ΔE_x^X are also given.

x (at.%)	Δx (at.%)	E_x^X (meV)	ΔE_x^X (meV)	E_b^X (meV)	E_g^X (meV)
0.0					1981 \pm 5
38.5	0.5	2062.8	0.2	12.2	2075.0 \pm 0.2
40.3	0.5	2068.4	0.2	12.4	2080.8 \pm 0.2
41.6	0.5	2072.6	0.3	12.6	2085.2 \pm 0.3
42.8	0.5	2073.9	0.2	12.8	2086.7 \pm 0.2
43.6	0.5	2075.7	0.4	12.9	2088.6 \pm 0.4
49.3	0.5	2089.5	0.3	13.6	2103.1 \pm 0.3
55.3	0.5	2104.2	0.4	14.3	2118.5 \pm 0.4
59.3	0.6	2115.9	0.3	14.9	2130.8 \pm 0.3
68.5	0.6	2138.1	0.4	16.2	2154.3 \pm 0.4
70.5	0.7	2143.9	0.4	16.5	2160.4 \pm 0.4
80.5	0.7	2169.4	0.3	18.1	2187.5 \pm 0.3
100.0		2229	1	21.4	2250 \pm 1

C. The indirect energy gap at X

At high excitation intensities ($P = 800$ W/cm²) when the temperature increases, some interesting modifications of the PL spectrum appear. As an example, we show in Fig. 3 PL spectra measured at different temperatures of the sample with $x = 0.493$. At $T = 13$ K a weak struc-

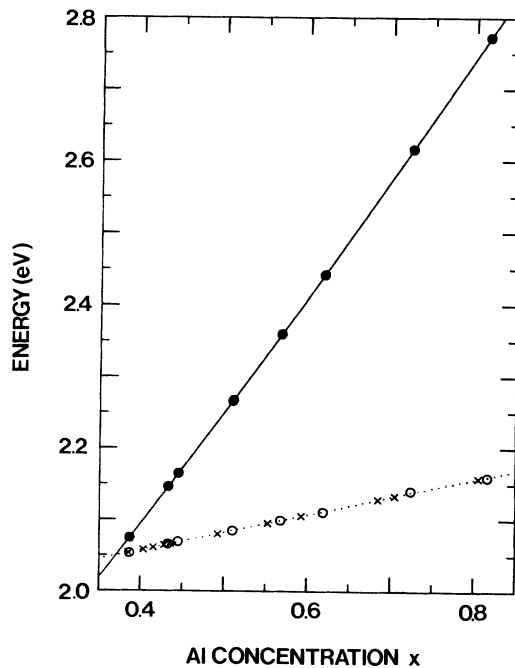


FIG. 2. Summary of the procedure we used to determine the Al concentration in our samples. The solid line is the $E_g^F(x)$ relation obtained at $T = 2$ K in Ref. 2. The closed and open circles are the energy of the direct free exciton measured in transmission and of the bound exciton (D^0, X) measured in photoluminescence at the same place, respectively. The dotted straight line is the least-squares fit through the open circles [Eq. (1)]. Finally, the crosses are the energies of (D^0, X) of the samples investigated by PL only; their Al concentrations, which are reported in Table I, have been determined using Eq. (1).

ture is present on the high-energy side of the bound exciton peak; with increasing temperature, the strength of (D^0, X) decreases while the strength of the high-energy transition increases and its high-energy tail becomes more and more pronounced.

This evolution indicates that the high-energy peak acquiring importance at $T > 12$ –15 K is the recombination of the free exciton (FX). It overlaps with the band-to-band transitions at higher temperatures. Observation of the indirect free exciton in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ has already been reported in Refs. 17 and 20. The difference between the free-exciton recombination energy and the (D^0, X) energy is the binding energy of the exciton to the impurities: at

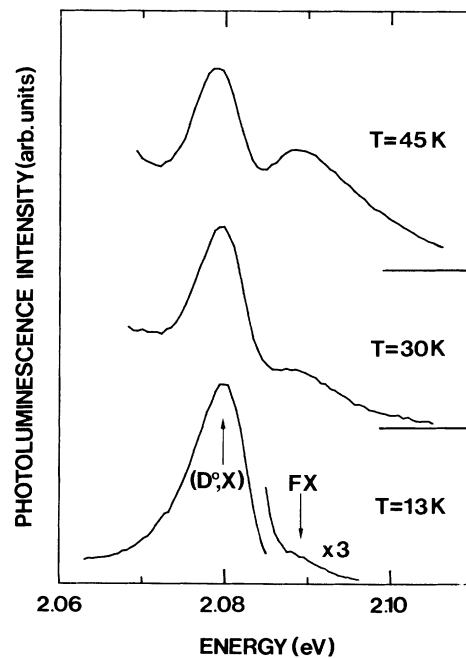


FIG. 3. Photoluminescence spectra at different temperatures of the sample having an Al concentration $x = 0.493$. The exciting power density is ~ 800 W/cm². (D^0, X) and FX refer to bound-exciton and to free-exciton recombination, respectively.

$x=0.593$, where the two (D^0, X) lines are clearly resolved, the binding energies are ~ 9 and ~ 13 meV, in good agreement with previous determinations.¹⁸

All the samples we studied show a behavior as a function of the temperature analogous to that reported in Fig. 3. Thus the energy E_x^X of the indirect exciton can be deduced directly from the PL spectra. We simply identify E_x^X with the peak energy of the zero-phonon exciton recombination line. This procedure yields E_x^X with an uncertainty of the order of kT: ~ 2 – 3 meV for $T \sim 20$ – 30 K. In Table I, the free-exciton energies E_x^X with their experimental errors are reported, as deduced from the spectra measured at $T=12$ K.

In order to get the value of the indirect energy gap, we add the free-exciton binding energy (E_b^X) to the exciton energy. The width and the weakness of the free-exciton structure prevent us from observing structures due to the excited states of the free exciton, and thus an experimental evaluation of the exciton Rydberg from our data is impossible. Furthermore, the value of E_b^X is not known either for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ or for AlAs; only some estimations have been published.^{20,21} An evaluation of E_b^X can be obtained using the hydrogenic effective-mass approximation:²²

$$E_b^X = \frac{\mu}{\epsilon_0} E_H, \quad (2)$$

where

$$\begin{aligned} \mu &= \frac{m_e m_h}{m_e + m_h}, \\ m_e^{-1} &= \frac{1}{3} \left[\frac{1}{m_{le}} + \frac{2}{m_{te}} \right], \\ m_h^{-1} &= \frac{1}{2} \left[\frac{1}{m_{hh}} + \frac{1}{m_{lh}} \right]. \end{aligned} \quad (3)$$

E_H is the ionization energy of the hydrogen-atom ground state, ϵ_0 is the static dielectric constant, and m_{le} and m_{te} are the longitudinal and transverse components of the electron effective-mass tensor, respectively. Finally, m_{lh} and m_{hh} are the light-hole and the heavy-hole effective mass, respectively. In Table I we report E_b^X values calculated using Eqs. (2) and (3) and the following relations for the electron and the hole effective masses and the static dielectric constant as a function of the Al concentration x :

$$\begin{aligned} m_{le}/m_0 &= 1.9 - 1.02x, \\ m_{te}/m_0 &= 0.19 + 0.06x, \\ m_{lh}/m_0 &= 0.082 + 0.078x, \\ m_{hh}/m_0 &= 0.51 + 0.20x, \\ \epsilon_0 &= 12.40 - 2.79x. \end{aligned} \quad (4)$$

The $m_{lh}(x)$, $m_{hh}(x)$, and $\epsilon_0(x)$ relations are taken from Ref. 1; for $m_{le}(x)$ and $m_{te}(x)$ we propose a linear interpolation between the values for GaAs reported in Ref. 1 and

the recently published experimental values for AlAs.²³

The effects of the anisotropy of the X minimum are included in our determination of E_b^X through the mean electron effective mass given in Eq. (3). However, our evaluation of E_b^X does not take into account the possibility of camel's-back structure at the X minimum in $\text{Al}_x\text{Ga}_{1-x}\text{As}$; if, as in GaP, the band extremum is not at the zone boundary, the exciton binding energies could be different from those reported in Table I.²⁴ The main error the calculated exciton Rydberg bears is mainly the poor knowledge of the hole masses,¹ which cannot be quantified. The validity of the approximation we used in the calculation of E_b^X can be tested in the case of GaP, which has a band structure similar to that of AlAs. For GaP, the exciton Rydberg calculated using Eq. (2) with the parameters reported in Ref. 25 (18 meV) agrees well with the experimental value (20.5 meV).²⁶ The values of the exciton Rydberg calculated this way are lower than the estimations published in the literature: 23 ± 2 meV for $x=0.60$ (Ref. 20) and 25.8 meV for AlAs.²¹

The indirect Γ - X energy gap $E_g^X(x)$ we obtained is shown in Fig. 4 together with the corresponding values for GaAs and for AlAs deduced from the literature. In GaAs, $E_g^X = 1.981 \pm 0.005$ eV, as deduced from the analysis of intraconduction-band optical-absorption spectra.²⁷ In AlAs the energy of the indirect exciton has been obtained from spectroscopic measurements:^{28,29} 2.229 ± 0.001 eV; with the exciton Rydberg calculated us-

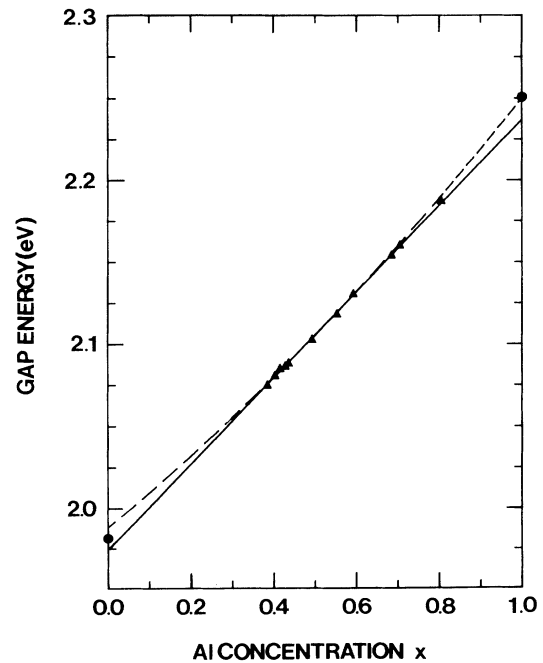


FIG. 4. Indirect energy gap at X vs Al concentration x ($T=12$ K). We also report data from the literature (see text) for GaAs and AlAs (closed circles). Errors are given in Table I. The solid and the dashed lines are the fits of the data with Eqs. (5) and (6), respectively.

ing Eq. (2) and the parameters (4), we get $E_g^X(\text{AlAs}) = 2.250 \text{ eV}$.

We made a weighted fit of the energy gap versus Al concentration x using polynomial functions. Our data and the literature value for GaAs (Ref. 27) can be fitted very well by a straight line:

$$E_g^X(12 \text{ K}, x) = 1.974 + 0.263x \text{ eV} . \quad (5)$$

The errors on the constant term and on the linear coefficient are ± 0.002 and $\pm 0.003 \text{ eV}$, respectively; the estimated standard deviation¹⁹ is 1.2 meV . The extrapolated value for the energy of the indirect gap of AlAs (2.237 eV) is too small compared with the value quoted in Table I. In order to also fit the AlAs indirect gap, we need a small quadratic dependence of E_g^X on x :

$$E_g^X(12 \text{ K}, x) = 1.988 + 0.207x + 0.055x^2 \text{ eV} . \quad (6)$$

The errors on the constant term and on the linear and quadratic coefficients are ± 0.003 , ± 0.011 , and $\pm 0.008 \text{ eV}$, respectively; the estimated standard deviation¹⁹ is 1.4 meV . The parabola given by Eq. (6) is reported in Fig. 4 together with the straight line (5) and the experimental points.

We can also determine the dependence of the energy gap on x at room temperature. The expression usually employed for the energy-gap dependence on the temperature is Varshni's semiempirical relation:^{30,31}

$$E_g(T) = E_g(0) - \alpha T^2 / (\beta + T) = E_g(0) - \Delta E_g(T) . \quad (7)$$

We use the values for α and β proposed in Ref. 1:

$$\alpha = 4.6 \times 10^{-4} \text{ eV/K}^2, \quad \beta = 204 \text{ K} . \quad (8)$$

These values have been assumed to be independent of the Al concentration. Thus we get

$$\Delta E_g^X(300 \text{ K}, x) = 0.082 \text{ eV} . \quad (9)$$

We note that, as discussed in Ref. 1, this implies an additional uncertainty of the order of 10 meV .³²

Combining Eqs. (7), (8), and (9), we obtain the dependence of the indirect gap on x at room temperature:

$$\begin{aligned} E_g^X(300 \text{ K}, x) &= E_g^X(12 \text{ K}, x) - \Delta E_g^X(300 \text{ K}, x) \\ &= 1.906 + 0.207x + 0.055x^2 \text{ eV} . \end{aligned} \quad (10)$$

IV. DISCUSSION

In this section our data, measured at $T = 12 \text{ K}$, are compared and combined with literature data that normally have been measured at $T = 2$ or 4 K . This small difference in the temperature does not constitute a problem, because the energy shift corresponding to this ΔT is practically zero.³¹

A. The indirect energy gap at X

The amount of published data to which our results on the $E_g^X(x)$ dependence can be compared is limited, as already mentioned in the Introduction. A detailed analysis of the recombination in indirect-gap $\text{Al}_x\text{Ga}_{1-x}\text{As}$ at low

temperature is given in Ref. 8, and some values of $E_g^X(x)$ for alloys with $0.4 < x < 0.8$ can be deduced from these experimental data (see Fig. 5 in Ref. 8). We report in Fig. 5 these values together with our $E_g^X(x)$ relation valid at low temperatures [Eq. (6)]. It is easy to see that Dingle's data are systematically below our curve (with a discrepancy up to 0.045 eV in energy or 0.16 in x) and that their dependence on x has definitely more bowing than our data. A similar situation was already found for $E_g^\Gamma(x)$.² Such a discrepancy might be due to the particular method Dingle, Logan, and Arthur used to determine x , as discussed by Kuech *et al.*¹²

Three relations for $E_g^X(x)$ at room temperature have been published in the past; two of them were deduced from the analysis of high-temperature transport data^{13,14} and the third one is reported in the well-known book by Casey and Panish.³ These relations are compared with our results [Eq. (10)] in Fig. 6. Once again, the bowing of the literature equations is higher than that of the $E_g^X(x)$ relation we propose.

Different theoretical works have been concerned with the dependence of the gap on the aluminum concentration x . Empirical pseudopotential calculations⁶ predict that the electron band structure of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ should in good approximation be a linear function of the alloy composition. Our experimental finding of a small quadratic coefficient in $E_g^X(x)$, $c = 0.055 \pm 0.008 \text{ eV}$, confirms this prediction. This point is consistent with the results we recently obtained² for the direct Γ - Γ gap. The results of first-principles calculations are contradictory; while a linear dependence of the gaps on Al concentration x has also been found in Ref. 33 [however, the linear coefficient

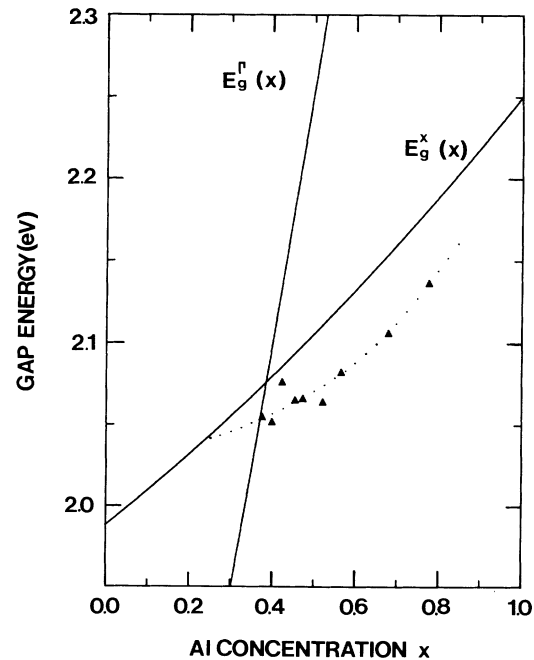


FIG. 5. Direct (E_g^Γ ; Ref. 2) and indirect [E_g^X ; Eq. (6)] energy gaps at low temperatures vs Al concentration (solid lines). Data published by Dingle, Logan, and Arthur (triangles and dotted line) are also shown (Ref. 8).

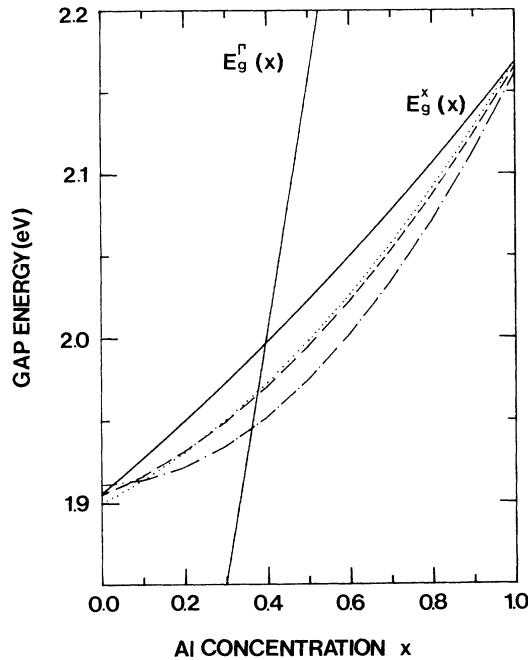


FIG. 6. Direct (E_g^Γ ; Ref. 2) and indirect [E_g^X ; Eq. (10)] energy gaps in function of the Al concentration at $T=300$ K (solid lines). $E_g^X(x)$ dependences published in Ref. 3 [$E_g^X(300$ K, $x)=1.900+0.125x+0.143x^2$ eV, dotted line], in Ref. 13 [$E_g^X(295$ K, $x)=1.911+0.005x+0.245x^2$ eV, dash-dotted line], and in Ref. 14 [$E_g^X(300$ K, $x)=1.905+0.26x-0.16x(1-x)$ eV, dashed line] are also shown.

of $E_g^X(x)$ calculated there is negative], the bowing of the direct gap $E_g^\Gamma(x)$ obtained in Ref. 34 is stronger than the one we found.²

B. The direct-indirect crossover

The dependence of the energy of the Γ - X gap E_g^X at $T \sim 0$ on the Al concentration [Eq. (6)] can be combined with that of the direct gap:²

$$E_g^\Gamma(x) = 1.5194 + 1.36x + 0.22x^2 \text{ eV} . \quad (11)$$

This way we obtain new values for the Γ - X crossover point at low temperature:

$$x_c = 0.385 \pm 0.016 , \quad E_c = 2.076 \pm 0.004 \text{ eV} . \quad (12)$$

At $T=300$ K the crossover point is obtained similarly, combining Eq. (10) with the dependence of the energy of the Γ - Γ gap:²

$$E_g^\Gamma(300 \text{ K}, x) = 1.423 + 1.36x + 0.22x^2 \text{ eV} . \quad (13)$$

We get

$$x_c = 0.396 \pm 0.016 , \quad E_c = 1.997 \pm 0.004 \text{ eV} . \quad (14)$$

The crossover concentrations at low and high temperatures are practically coincident with those determined by Kuech *et al.*¹² [$x_c = 0.37 \pm 0.015$ ($T=2$ K)] and by Oelgart *et al.*¹¹ [$x_c = 0.374 \pm 0.005$ ($T=38$ K) and $x_c = 0.41 \pm 0.01$ ($T=300$ K)]. They support also the older evaluation by Monemar, Shih, and Pettit⁹ [$x_c = 0.35 \pm 0.01$ ($T=4$ K) and $x_c = 0.37 \pm 0.01$ ($T=293$ K)]. All these results confirm that the crossover concentration is definitely lower than the often-used value of $x_c \approx 0.43$.⁸

V. CONCLUSIONS

This work on the determination of the indirect Γ - X gap follows our previous paper² on the Γ - Γ direct gap in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys. The accurately measured Al concentrations of Ref. 2 have been used to determine the composition of the samples in the present work. The registration of the photoluminescence spectra at higher temperatures enabled us to measure directly the free indirect exciton energy in the range $0.38 < x < 0.81$. Using the exciton binding energy calculated in the effective-mass approximation we got the value of the indirect energy gap $E_g^X(x)$. The dependence on aluminum concentration of this indirect gap has practically no bowing; a similar result had been found in our previous work for the direct gap. The Γ - X crossover has been determined and found to be in agreement with earlier evaluations.

We think that this direct determination of $E_g^X(x)$ settles the problem of the poor knowledge of the indirect Γ - X gap in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys. The main information that is still absent from our knowledge of the conduction-band structure in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ is the energy of the L minimum.

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