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Band gaps and spin-orbit splitting of ordered and disordered Al„Gat „As and GaAs„Sb& „alloys

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Spontaneous long-range ordering of the otherwise disordered isovalent semiconductor alloys A B, C has been recently observed in numerous III-V alloy

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induced effects will be more pronounced.
This study is aimed at (i) developing an understanding
of the basic differences between the electronic structure
of ordered and disordered alloys, and (ii) aiding in the
identification of ordered ternary compounds, through their electronic fingerprints.
The two systems we have chosen to study are the or-dered common-anion Al„Ga4 „As system, exhibiting near lattice match, and the lattice-mismatched common-cation Ga4As„S14 „system. Both systems have been ob-served to form spontaneously ordered phases when grown on lattice-matched substrates.
Using self-consistent first-principles band-structure cal-culations we find that (i) the fundamental band gaps of the ordered ternary compounds are usually smaller than the linear average of the band gaps of the binary constitu-ents due to symmetry-enforced level repulsion; (ii) the bowing parameter of the spin-orbit splitting Ao is nega-tive for common-cation ordered ternary compounds due to repulsion-induced

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here as "CA"}and at 25%-75% or 75%-25%%uo composi-
tion the luzonite-like structure (denoted here as "L").
(iii) For the (1,0,—,

) ordering vector [sometimes denoted
(2,0, 1)], we have at 50%-50% composition the chalcopy-
rite structure (denoted here as "CH"), whereas for 25%-
75% or 75%-25% composition we have the famatinite

structure (denoted here as "F"). Finally, (iv) for the (—, ', —, ', —, ') ordering vector [sometimes denoted (l, l, l)], we

have at the 50%-50% composition the layered trigonal CuPt-like structure (denoted here as "CP").
Since the information on these structures is scattered
in numerous literature sources, and since different nota-tions are used in these sources, we give in Fig. 1 a comprehensive list of notations. For instance, to find the space group operations, use the International Tables for Crystallography under the space-group number given in Fig. 1. To find other compounds belonging to these
structure types and the atomic-position coordinates, use
Pearson's Handbook of Crystallographic Data for Interme
tallic Phases according to the "Pearson symbol" given
in Fig. 1. Information regarding the structural chemistry of these phases can be found in Ref. 44, designated ac-
cording to the Struckturbericht symbol given in Fig. 1.
Statistical-mechanics descriptions of configurational or-
dering in these Landau-Lifshitz structures (e.g., Refs. 40,
41, and 45) are designated according to the "ordering
vectors.

" Figure

1 also indicates (where applicable) the

in

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TABLE I. Mapping (folding relationships) of the zinc-blende (ZB) I and X states into ABC& CuAu-I-like states and A, BC4 luzonite-like states at the center of the Brillouin zone. The origin of the coor-dinate system in ZB is at the anion site.

ZB

ABC„CuAu
Common anion Common cation

A 3BC4, luzonite
Common anion Common cation

r,
I ls
X,
X3
Xs

I I
I s+I 4

I,
r,
r,

I.
,

I,+I.

,

II
I4
Is

I I
I is
I ls

I (+I)2
I is+I zs

I)
I is
I I+I 12
I is
I is+I zs

turbed" crystal potential for the ternary structure such

that the states I „(Ii,) and 1 „(Xi,) would be degen-erate with the average energies (I „)and (X„),respec-

tively. Despite the fact that a seemingly new direct state

I, (X„)is generated in the ternary structure, in the ab-
sence of a potentia1 perturbation the no-phonon transi-tion matrix element from valence-band I states is, of
course, still precisely zero, just as is the case for transi-
tions to the X&, state from folded.
In reality, however, there ordering potential perturbation b,

r)

distinguishing energy levels of the
ternary system

from the

average energy levels

of the

binaries. potential not have zinc-blende sym-metry, but belongs instead to the totally symmetric repre-sentation of the space group of the ternary structure. In general, b,

r)

has a electronic piece, contribut

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TABLE III. Mapping (folding relationships) of the zinc-
blende (ZB) I and L states into ABC2 CuPt-like states at the
center of the Brillouin zone. Origin at the anion site.

(a) Linearly Averaged
band gaps
(LDA)

b
Linearly Averaged
band gaps
(Experimental)

ZB

r,
I ls

L,
L3

Common anion Common cation 2.2—

o 1.8

CA

1.4

LLJ

1.0

0.6

1.

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the wave function on difFerent sublattices in A„B4 „C4.
The degree of this localization is strongly correlated with
the energy shift 5E of Eq. (l). This repulsion-induced
level localization is analogous to that described in tight-
binding models for zinc-blende compounds, where level
repulsion causes the bonding states (shifting to lower en-ergies) to localize on the anion sites, whereas the anti-bonding states (shifted to higher energies) localize on the cation site (since the anion has lower s- and p-orbital en-ergies than the cation). This repulsion-induced localiza-tion will have important efFects on the spin-orbit splitting which depends primarily on the degree of wave-function localization near a given atomic site.
(vi) Since in ternary tetrahedral semiconductors the I »-like top of the valence band (p-like) is usually repelled
upwards

top 117i4 Td (This)Tj-l0.8 Tf 57.33 549.44 Td (IWielled) (This)Tjing

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TABLE VII. Calculated semirelativistic LDA band

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rameters need to be established through total-energy
minimization. In addition to the unit-cell lattice vectors,
the ternary

A „84 „C4 structures of Fig.

1 have cell-
internal structural parameters which determine the equi-librium bond lengths R z c and R~ z

and the bond angles.
We have minimized for Ga4As„Sb4

„ the total energy of
each of the ordered structures, with respect to both the
unit-cell lattice parameters and the cell-internal degrees
of freedom. The resulting equilibrium structural parame-ters are given in Table IX. We see that the cubic lattice constants of these ordered compounds follow closely
Vegard's rule, i.e.

, a,'q'

= —, '[na, „(AC)+(4

—n)a,q(BC)].
In contrast, bond lengths do not average, but instead ex-
hibit a bimodal distribution' ' whereby the Ga—As and
Ga

—Sb bond lengths in the ternary compounds are
closetructural

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2.5—

2.0—

I „(L)

CuPt

(d)

1.98; X] 1.43

1.81; Llc
1.09

0.81
JI

GaAs GaSb
(a) (b)

2.30 X3c

CuAu

(c)
1.„(X,)-

1.71 I4 (X~)

0.86 1-, (1-,)-

ii

[111]direction; see Fig. 1), we will compare their energy levels to the well-center and well-bottom energies of Fig. 5, to discuss wave-function localization. Note that for lattice-mismatched system like Cxa(As, Sb), the energy alignment of Fig. 5 pertinent to thick superlattices will be modified for ultrathin superlattice, particularly for those conduction states which are sensitive to the

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TABLE XI. LDA and LDA-corrected band energies with respect to the valence-band maximum (in eV) of the three ordered Ga4As2Sb2 structures at I.

ZB
label State

Ga4As2Sb~ (CA)
LDA
corrected State

Ga~As2Sb2 {CH)
LDA
corrected State

Ga4As2Sb2 (CP)
LDA
LDA corrected

I is.
~lsv

r„
Xs„
X),
X3,
L3v

Lj,

I s„
r„,

r„
r,.
r„
I4,

0.0' —0.09 —0.316 —2.76

1.42
1.03

0.0' —0.09
0.86 —3.09
1.98
1.71

r„,

r„
r„
r„
I3,
I2,

—0.01
0.0'

—0.050 —2.57
1.23
1.14

—0.01
0.0'

1.13 —2.90
1.79
1.82

0.0'

—0.23 —0.781

—1.51 —0.90

0.0'
—0.23
0.37

—1.74 —1.79
'Valence-band maximum.

TABLE XII. LDA and LDA-corrected band energies with respect to the valence-band maximum (in eV) of the three ordered A1~Ga2As4 structures at I.

A12Ga2As4 (CA) A12Ga2As4 (CH) A1~Ga2As4 (CP)
ZB
label

I is.
~1sv

r„
Xs,
X),
X3,
L3,
Li,

State

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l

er

C$

e

0
bo

a$

05

ch

GC

Q

Q

ch

g~ ~

C

C3
CP

CA
r/0

V

*

bo

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(a)[GaAs

2.

4

-

2,30

X3C 2.2-
20

X

198

1.8-
1.6-
14

- 1.52

1.2-
1.0-
0.8-
0.6-
0.4-
0.2-
00 YBM

- 2.

8

-

-2.92
30 X5y
-3.2—

GasAsSb
"CuAut"

(001)

1.98; Pic

1.71;

I 4C

0.86;

I 1C

0.0;

I 5y
f

-0.09;

I 4y

-3.09;

I 5y

I GaSb

t

1.66
X1C

X3~ 1.43

I1C 0.81

VBM

-2.94
X5y

I GSAs

1

2.30
X3C

1.98 XIC

1C

VBM 0.

0

-2.92 Xsy

GaqAsSb
Chstcopyrlte
(201) IGSSb

l

X1C
X3C-

0.0; r4y
-0.01; P5y

-2.90;

I 5y -2,94

VBM

X5y

1.82;l pc 1

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small (L„)—(I „) energy denominator of 0.49 eV.
Hence, the I „(I „) wave-function amplitude is

enhanced significantly on the GaAs sublattice [Fig.
11(a)]. This leads to "indirectness in real space" whereby
the I 3, VBM is localized preferentially on VaSb, whereas
the 1 &, CBM is localized on the GaAs sublattice.
The upward shift of the VBM (by 0.19 eV) and the
downward shift of the CBM (by 0.605 eV) leads to a very
large bowing coefficient of 4(0.19+0.605)=3.18 eV. The large crystal-Geld splitting leads to different localization
patterns: while the I 3, (1 i~,) state [Fig. 11(b)] is local-
ized preferentially on Sb, the I i„(I »,) state [Fig. 11(c)]
has a preferred localization on As. Compared with the binaries, the total charge density at the VBM [Fig. 11(d)] is, however, more localized on Sb.

4. General trend for bowing
in ordered GaSb-GaAs compounds

We see that because of the different identities of the re-pelling states and the different symmetry properties, the

bowing coefficients for Ga4As„Sb4 „depend strongly on the crystal structure with b(CP) &b(CA) &b(CH). Simi-larly, we find b(L) & b(F) for n =1,3 luzonite and famatin-ite structures. In all cases we find that the top of valence band at I tends to localize on the Sb sublattice and that the bottom of conduction band tends to localize on the As sublattice.

Since the differences (I i,)—(Xi,) and (I i,) —(L„) in the average energies (Fig. 3) as well as

(I », }—(X5„) and (1,5,)—(L3„) depend on composi-tion weakly and linearly, we find that b (X„)is only weak-ly composition dependent for the structures with the (0,0, 1) ordering vector (i.e.,

the ordered CuAu-I-like and

"luzonite"-like structures). We find that b (X„)is slightly larger in As-rich compounds since the energy difference

(I I,)—(Xi,) is slightly smaller in the As-rich side re-gion (Fig. 3). The larger bowing of the two famatinite structures relative to the chalcopyrite structure is due to symmetry-allowed coupling between the X-derived and I -derived I state (Table II).

(a) i GaqAsSb (CA) i

(a) ~ GasAsab (CH)(

[I 4C (I qC)]

F1C (I 1C)

A;)

Itsy (I ~sv) I

[I sv (I 4sv) (

(c)

As I

Sb E 3. As

(I 4v(I 4sv)I)r4v (I csv)[

~

I sv + I 4v] Psy+I 4y

EGa

FICx. 9. Electronic charge-density contour plot of Ga2AsSb in CuAu-I-like structure (in units of 10 e/a. u.) for (a) the I „ (I I,) state, (b) the I 5, (I »,) state, (c) the I 4, (I »,) state, and (d) the weighted sum of (b) and {c).

FICx. 10. Electronic charge-density contour plot of Csa2AsSb

in chalcopyrite structure {in units of 10 e/a. u. ') for (a) the I „ (I I,) state, (b) the I 5„(I»,) state, (c) the I &, (I »,) state, and (d) the weighted sum of (b) and (c).

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IGa2Assb (CP))

/'

j,// (((rr'
/i' I(j' Ga

)~1C (t 1C)I

Since GaAs, GaSb, and their alloys are direct-gap ma-terials (cf. Fig. 3), folding of X states in the ordered ter-nary compounds does not change this basic situation. This is not the case in A1As-GaAs alloys, as discussed
next.

E. Ordered GaAs-AlAs compounds

'=.1 0/,

(b)

Ga

~Sb)

Ga &, =-== 20 &--10-

I&3v (&i5v)I

Figure 12 depicts the LDA-corrected energy levels at 1 for GaAIAsz in the CuAu-I-like [Fig. 12(a)], chalcopy-
rite [Fig. 12(b)], and CuPt-like [Fig. 12(c)] structures,
along with the energy levels of the binary constituents from which folding occurs. Table XV gives the angular-momentum decomposition of the wave functions at I. We find for this system that the VBM shows only negligible level repulsion and localization eA'ects since Ga and Al have very similar p-orbital energies (Table IV). Therefore, the optical bowing for this system is almost entirely due to the bowing of the conduction states.

1. CuAu-I

b)

+riv I

FIG. 11. Electronic charge-density contour plot of Ga&AsSb in CuPt-like structure (in units of 10 e/a. u.) for (a) the I &,

(I"&,) state, (b) the I 3, (I"»,) state, (c) the I „(I»,) state, and

(d) the weighted sum of (b) and (c).

The I &, (I 1,) state is repelled downward by I"

&, (X3,)
by 0.152 eV. The bowing coe%cient for this transition is, therefore, 4X0.152=0.61 eV. Since Ga has a more

tightly bound s orbital than Al (Table IV), the I „(I„) state tends to localize on the GaAs sublattice. The I 4, (X„)state is repelled upward by I 4„(I »,) by 0.07 eV

but is still lower in energy than the I"1, (I &,) state.
Study of the energy levels away from the Brillouin-zone center show the conduction-band minimum for this sys-tem at R &, (L &,) (at E, +1.92 eV) is due

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10

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CP V
V

ch

c5 (Q

nfl

g

U

Zry$

8
~ tW

+

q5 c5
cn

V3

c5

Q Pe
X

C4

cn

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2. Chalcopyrite
As is the case for all other

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4

E (x)= g P„(x)e'"'[a (x)]. (14)

Clearly, the ordered CuAu-I structure forms a

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1.8 3.1

14

~~ 1.0

(Xl 0.6

0.2
GaAs

2,7)
2.3

19 Pa

I I 1.5
025 050 075 AIAs GaAs 025 050 o,75 AIAs Concentration (mole fraction)

FIG. 14. Calculated concentration variation of the energy
gap Eg(I &,—I ») of the Al Ga& As alloy (solid line).
Dashed lines give the linear concentration-averaged band gap; band gaps of the ordered ternary compound are shown as solid triangles. Using (a) the LDA and (b) LDA-corrected results {see text).

B. Bowing of the spin-orbit splitting bo in disordered alloys

In analogy with Eq. (12), the composition variation of the 60 spin-orbit splitting has been fitted to the form

bo(x) =ho(x) b(ho)x (1——x), (19)

where b(b, o) is the bowing parameter. For all ordered GaAs-GaSb structures we find (Table XIII) b(b,)&00. For the disordered alloy b(b, o) of Eq. (19) is —0.08 eV.
The experimental situation pertaining to the magnitudes
of b (b.o) can be summarized as follows.
(i) Epitaxially grown lattice-matched samples show
b (b,o) &0, i.e., an enhancement of the spin-orbit splitting
over the linear average value. This is the case for
InGaAsP/InP, where b (b,o)=—0.09 eV (Refs. 113—117)
and in GaA1InAs/InP, where b (bo) =—0.10 eV."
(ii) Bulk grown alloys are said"

'
to exhibit

b(b,))o0, whereas our calculation for GaAsSb shows b(b, o) &0. The experimental situation here deserves fur-
ther discussion.

Most of the experimental studies on b (b,o) in bulk III-
V alloys were carried out in the 1960s by Woolley and
co-workers. ' " '

In any early study, " Berolo and
Woolley found that numerous III-V systems show posi-
tive b(bo) values, in excess of tn2103.89 Td (systems)Tj ET B7.56 184.33 d (of)Tj E BT /Xi21 13

to(f)Tj ET BT /Xi21 9.52 Tf 108.56 93.7sam8(bo))g BT /Xi21 8.13 Tf 75.11 189.sam8 (')Tj ET BT /Xi21 9.71 Tf1 164.44 297 Td d (alloy)T BT /Xi21 9.65 Tff 19 innfor bspin-or1 BT /Xi21 16.84 T 122.67 154 7 (spin-orT BT /Xi21 9.54 Tf 233 114.38.33 Td(for)Tj ET BT /Xi21 9.86 Tf 534.33 566.33 Td(b)Tj ETP,)TST BT /Xi21 10.22 Tf 225.78 617.22 72(for)Tj T BT /Xi21 9.49 Tf 262.67 170.22 72(for)Tj T0 BT /Xi21 13.23 T of 0.

val5d (i.)Tj ET BT /Xi21 9.06 Tf 26.44 351.56 Td (i.)Tj ETned

i.

i.

i.

the &,

for

for

(b5d (case)TjresponsiET BT /Xi21 10.95 T 249.56 123 Td 5d (for)Tj ET BT /Xi21 9.57 Tf 296 276.54.22 Td (for)Tj ET BT /Xi21 9.07 Tf 115.11 19 (3)TTd (for)Tj anomalET 0. for436.22 Tj Edemons-ave BT /Xi21 9.53 Tf 281.67 214.78 436.for 'orderedE14T BT /Xi21 9.26 Tf 78.89 236.22 478(carried)T BT /Xi21 8.89 Tf 206.44 351.11 478(carriedadequve T BT /Xi21 8.86 Tf9128.33 555.89 47 (show 'anFor EpitaxiavT BT /Xi21 8.89 Tf 173.22 211.44 4d (ther)Tj Eoug BT /Xi21 10.1 Tf 241.11 205.89 44he show For (of)436r i.and

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Adjustment of the tight-binding matrix elements describ-
ing this perturbation led to a good fit of the experimental
data. Ling and Miller' argued that this perturbation is
too large for perturbation theory to be valid. They have calculated this mixing using

a superposition approach,
showing that it leads

b (b,o) values which are considerably
smaller than the experimentally reported values.
Our discussion above suggests that there is no rigorous
quantitative theory currently available to describe the large nonlinear deviation of bo(x) observed in experi-
ment. "

Further study on this subject both experimen-
tally and theoretically appears desirable. Since alloying may also split the heavy-hole and light-hole states at the VBM, crystal-field splitting should also be

split

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a[.(X„)—a.(X„)]
ad (A4)

dc/dp)0 (A6)

so that E(Xi,)—E(X3,) is a monotonic function of d.
Combined with the discussion surrounding Eq. (A2), this
shows that

E(X„))E(X3,) for all d)0, (A5)
in obvious conflict with other calculations (e.g., Table
VII).
Since the pressure derivative dc. /dp has the opposite sign of t)EIBd, Eq. (A3) also shows that

for both Xi, and X3„in conflict with experiment (Table VI).
Equations (Al) and our discussion provides, however, simple guidance for understanding the order of X&, and X3 states: for systems with sufficiently large lattice pa-rameters (hence, sufficiently small V,), sufficiently shal-low anion s states and sufficiently deep cation s states X3, will be below X&,. We find this to be the case for GaSb and HgTe.

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34, n3 9.gf 61.67 121.56 Td (3199)TjBT /Xi23 9.6 Tf 115.67 111.56rj ET j Eo5676 141.96 Tf 127.56 181.rTj ET BT /Xi23 8.98 Tf 112 201.4i 141.1 2433 Td (7T /X). 34,

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50Td (in)Tj4i 141.1 2433 Td (7TTf 434.169.560Td (in)Tj10.7 (900)89 131.3723 8.98 111.1120589 Td (S.27by)Tj ET BT /Xi23 8.96 Tf 22 26260Td (in)Tjd)Tj ET BT /Xi2 Res.pn58 Td (n3 9.gf 61.67Tf 1914 61.564867 Td (dii23 9.6 Tf 115.67 111.56rj4329.22 485.11 Lett.)Tj ET BT /Xi23 8.53 Tf 468.89 48184.67 49Tj ET BT /Xi23 9.07 Tf 1 T Lett. 56,

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of andCr Lett.

35.11 Td 83).

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Tj.

of L. P. ChKlel.

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Sh36.1.89 Td (Lett.)Tj ET BT /Xi23 883.56 3 183.1Tf 401.4 Td585G. MartiET B

pa-Sh327167 Td (,)T 52, 6,ITj ET BT /Xi23 8.85 19 Tf 3544.2 00230.

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39d (27,)Tj E (198