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Spin-orbit splitting in $Mg_xZn_{1-x}Te$ alloys

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The spin-orbit splitting Δ_0 and Δ_1 of electron energy levels at the Γ and Λ points are obtained for the alloys $Mg_xZn_{1-x}Te$ in the cubic range $x < 0.5$ from wavelength-modulation reflectivity measurements at both $T = 300$ K and $T = 82$ K. Within experimental scattering, Δ_0 and Δ_1 do not depend on x and T. These data are explained by semiempirical developments on the basis of the well-known time-sharing relation following the Berolo-Woolley analysis. An evaluation of the multiplicative factor in this relation is made for Δ_0 by introducing the metallicities of end compounds. In addition, we give a new set of a parameters for the calculation of Δ_0 in II–VI binary compounds from the time-sharing relation. Finally, an estimation is made of Δ_0 and Δ_1 for MgTe: $\Delta_0 \sim 0.95$ eV and $\Delta_1 \sim 0.6$ eV. The 2/3 law is roughly observed for end compounds and ternary alloys.

I. INTRODUCTION

The composition dependences of spin-orbit splittings (SOS) in alloys are of great interest since they can be analyzed on the basis of semiempirical models. In this paper we report the first observation of SOS transitions in $Mg_xZn_{1-x}Te$ ternary alloys. The incorporation of magnesium in ZnTe leads to ternary semiconductor alloys having higher band-gap energy and allows for blue luminescence.[1-5] Ternary alloys exist in all the composition range.[1] A change from zinc-blende to wurtzite structure is observed near $x = 0.5$. Due to the reactivity of Mg and change in structure, the Mg-rich alloys are difficult to grow. The optical properties of MgTe are almost unknown. Even its fundamental band-gap value is controversial: 4.7 eV (Ref. 1), 3.6 eV (Refs. 2–4). In contrast ZnTe has been extensively investigated and our results shall be easily compared with those found by different techniques. The previous studies of the optical properties of MgZnTe alloys are related to the transition near the E_0 fundamental gap.[1-4] In cubic materials, Parker[1] has obtained by cathodoluminescence a quasilinear variation of $E_0(x)$. From wavelength-modulated-reflectivity measurements (WMR) and cathodoluminescence a parabolic law has been recently inferred.[2-4]

In this paper we give an accurate determination of $E_0 + \Delta_0, E_1, E_1 + \Delta_1$ gaps from WMR spectra $\Delta R/R = f(\lambda)$ at 300 K and 82 K for high-purity $Mg_xZn_{1-x}Te$ alloys. The results, especially the laws $\Delta_0(x)$ and $\Delta_1(x)$ for spin-orbit splitting at Γ and Λ points, are analyzed and discussed within recent theories. Finally, an estimation of Δ_0 and Δ_1 is given for MgTe on the basis of experimental and theoretical data. In addition we provide a new set of parameters for the calculation of Δ_0 in II-VI binary compounds from the well-known time-sharing relation.

II. EXPERIMENT

A. Sample preparation

The samples were synthetized by the Bridgman technique using the method described in Refs. 3 and 5. The starting materials are 99.9999%-purity grade. The crystals have high purity and good homogeneity. Their compositions were determined by electron-beam microprobe analysis with an accuracy of $\Delta x/x \sim 5\%$. They were not intentionally doped. Before the WMR, the surfaces of the crystals were polished mechanically, then etched in a solution of bromine in methylalcool, rinsed in distilled water and then in methanol.

B. Wavelength-modulated-reflectivity measurements

We used the wavelength-modulation technique because of its nondestructive character and of the fact that no particular preparation is needed for low-temperature measurements. Moreover, magnesium makes it difficult to use the electrolytic method in electroreflectance. The wavelength-modulation spectrometer used in our experiments is derived from apparatus described by Shen.[6] A double-beam method cancels the background contribution to the derivative signal. The optical detector is a Hamamatsu R376 photomultiplier. The signals are analyzed by analog electronics in order to plot the logarithmic derivative $\Delta R/R$ of the reflectivity versus the wavelength. The monochromatic beam at 122 Hz. The wavelength modulation can be adjusted continuously within the 4–24-Å range. The detectivity of the spectrometer is better than 10^{-4}.

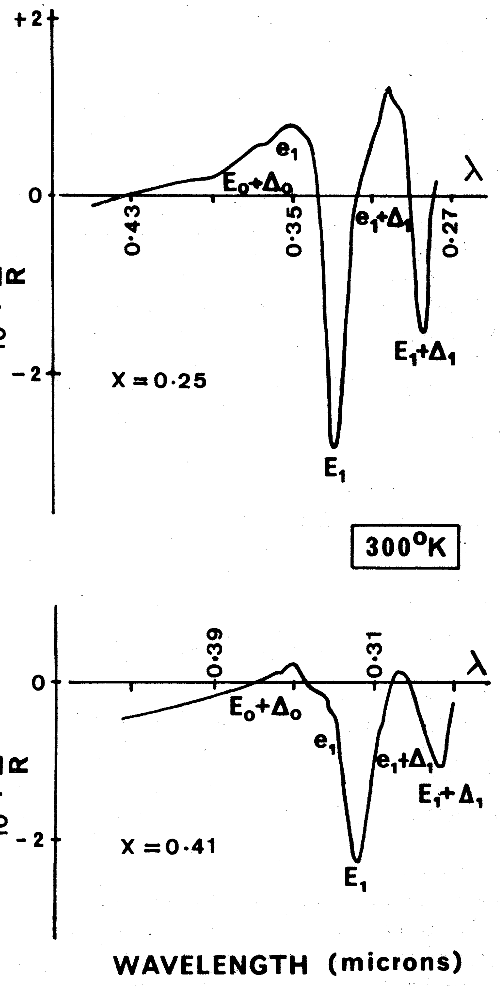

FIG. 1. WMR spectra $\Delta R/R = f(\lambda)$ of the ternary alloys $Mg_xZn_{1-x}Te$ for $x = 0.25$ and 0.41 at 300 K.

III. EXPERIMENTAL DATA

The spectra $\Delta R/R = f(\lambda)$ of $Mg_xZn_{1-x}Te$ have been investigated systematically between 300 K and 82 K for five concentrations x: 0, 0.05, 0.097, 0.25, and 0.41. Figures 1 and 2 show some spectra at these temperatures within the range 3–4.7 eV for various x. They are roughly similar to those obtained previously for ZnTe using electroreflectance or thermoreflectance techniques.[7,8] The energy position and amplitude of various structures change regularly with x. The tentative assignment of peaks is made by comparison with ZnTe on the basis of the literature data.[7-13] The $E_0 + \Delta_0$ structure is resolved at 300 K and 82 K. For ZnTe, its energetic midposition gives a spin-orbit splitting in good concordance with previous results (see Table I). As the energy increases, two large peaks appear attributed to the E_1 and $E_1 + \Delta_1$ doublet in the [111] direction of the band structure (Λ point). For ZnTe, the values for Δ_1 deduced from the difference between the energy positions E_1 and $E_1 + \Delta_1$ are compared with data in the literature in Table I. We can also identify near this doublet a weakly resolved structure attributed to e_1 and $e_1 + \Delta_1$ transitions for the L point at the boundary of the first Brillouin zone. The energy positions of e_1 and $e_1 + \Delta_1$ are obtained with poor precision. The value deduced for Δ_1 from this structure agrees with the one obtained from the $(E_1, E_1 + \Delta_1)$ doublet. The energies of the E_0' and E_2 transitions are located outside the spectral operating range of our apparatus.

Figure 3 shows the variations of the E_0, $E_0 + \Delta_0$, E_1, and $E_1 + \Delta_1$ bandgaps as a function of x at 300 K and 82 K. We can remark that the $E_0(x)$ and

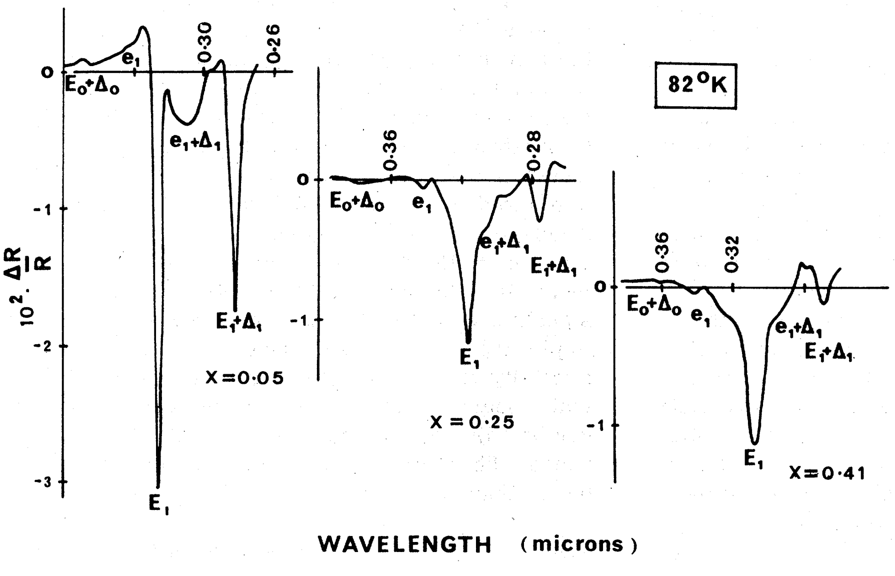

FIG. 2. WMR spectra $\Delta R/R = f(\lambda)$ of $Mg_xZn_{1-x}Te$ for $x = 0.05$, 0.25, and 0.41 at 82 K.

TABLE I. Experimental values of spin-orbit splitting Δ_0 and Δ_1 for ZnTe at 300 K.

Spin-orbit splitting	Measured value (eV) (this work)	Recent literature data (with measurement techniques and references)
Δ_0	0.96 ± 0.01	0.93 (electroreflectance) (Ref. 7) 0.96 (reflectivity) (Ref. 9) 1.00 (electroreflectance) (Ref. 10) 0.92 (electroreflectance) (Ref. 11)
Δ_1 (deduced from E_1 and $E_1+\Delta_1$ structures)	0.59 ± 0.015	0.57 (electroreflectance) (Ref. 7) 0.57 (thermoreflectance) (Ref. 8) 0.57 (reflectivity) (Ref. 9) or 0.59

$E_0+\Delta_0(x)$ representations are quasiparallel, respectively. The eight curves have been fitted to parabolic relations of the form $E = a + bc + cx^2$ using the least-squares method.

The values of the experimental bowing parameter are reported in Table II. For the investigated temperature the $E(T)$ variations are linear. Table III shows the data for $E_0+\Delta_0$ transitions at 300 K and 82 K and the temperature coefficients $d(E_0+\Delta_0)/dT$. Within experimental scattering these coefficients are constant with x: $(-5 \pm 0.5) \times 10^{-4}$ eV K^{-1}. They agree with the values for dE_0/dT: -4.6×10^{-4} eV K^{-1}.[4] Table IV lists the data for E_1 and $E_1+\Delta_1$ versus x at 300 K and 82 K. It is interesting to remark that the temperature coefficients for E_1 and $E_1+\Delta_1$ are also nearly independent of x: $(-6 \pm 0.5) \times 10^{-4}$ eV. That suggests that (i) as for ZnTe and many semiconductors, the major contribution to the shift with T for E_0 and E_1 is due to electron-phonon interaction; (ii) Δ_0 and Δ_1 are nearly independent of temperature.

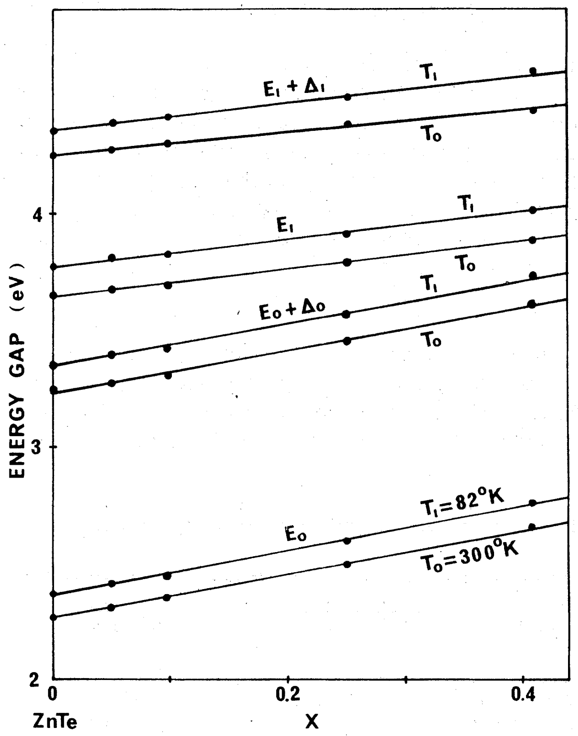

FIG. 3. Dependence on composition x of the energies of band gaps E_0, $E_0+\Delta_0$, E_1, and $E_1+\Delta_1$ in Mg$_x$Zn$_{1-x}$Te crystals at $T_0 = 300$ K and $T_1 = 82$ K.

IV. ANALYSIS

Within the experimental dispersion the spin-orbit splitting Δ_0, given by the energy difference between the E_0 and $E_0+\Delta_0$ gaps is not dependent on x and T (the average Δ_0 is 0.97 at 300 K and 0.98 K at 82 K). This result is interesting because it may be theorically guessed. We follow the well-known "time-sharing" principle which was applied by Kane for an estimate of SOS in crystals in terms of atomic splitting for constituent free atoms.[14]

The SOS Δ_0 of the valence band at Γ_{15}, using Cardona's notation,[12] is given by

$$\Delta_0 = \left(\frac{3\hbar i}{4mc^2}\right)\left\langle \Gamma_{15}(x) \left| \frac{\partial V}{\partial x} p_y - \frac{\partial V}{\partial y} p_x \right| \Gamma_{15}(y) \right\rangle , \quad (1)$$

where V is the self-consistent crystal potential

TABLE II. Bowing parameter from WMR for Mg$_x$Zn$_{1-x}$Te ternary alloys.

Transitions	Experimental bowing parameter (eV) 300 K	82 K
E_0	0.60 ± 0.10	0.43 ± 0.10
$E_0+\Delta_0$	0.55 ± 0.10	0.47 ± 0.10
E_1	0.24 ± 0.10	0.26 ± 0.10
$E_1+\Delta_1$	≈0 ± 0.10	0.21 ± 0.10

TABLE III. Values of $E_0+\Delta_0$ and shift-temperature coefficient for the ternary alloys $\mathrm{Mg}_x\mathrm{Zn}_{1-x}\mathrm{Te}$.

x	$E_0+\Delta_0$, 300 K (eV)	$E_0+\Delta_0$, 82 K (eV)	$\frac{d(E_0+\Delta_0)}{dT}$ (10^{-4} eV/°K)
0	3.245 ± 0.005	3.347 ± 0.005	−4.7 ± 0.5
0.05	3.283 ± 0.005	3.394 ± 0.005	−5.1 ± 0.5
0.097	3.307 ± 0.005	3.424 ± 0.005	−5.4 ± 0.5
0.25	3.466 ± 0.005	3.570 ± 0.005	−4.8 ± 0.5
0.407	3.616 ± 0.010	3.740 ± 0.010	−6.0 ± 1.0

and p is the linear-momentum operator. $|\Gamma(i)\rangle$ are the wave functions which define the Γ_{15} representation (without splitting). By examination of the contribution to the matrix element in Eq. (1) it is expected that Δ_0 be given by a contribution from the group-II atoms and by another from the group-VI atoms. Hence generalizing the Braunstein-Kane approximation[15] and the Berolo-Woolley analysis[16] in the spirit of the virtual-crystal approximation, the SOS Δ_0 of $\mathrm{Mg}_x\mathrm{Zn}_{1-x}\mathrm{Te}$ can be written

$$\Delta_0(x)=C_0(x)\{t(x)[x\Delta_{\mathrm{Mg}}+(1-x)\Delta_{\mathrm{Zn}}] + [1-t(x)]\,\Delta_{\mathrm{Te}}\}\,, \qquad (2)$$

where the Berolo-Woolley disorder parameter γ_t is assumed to be zero. Δ_{Mg}, Δ_{Zn}, and Δ_{Te} are the respective atomic splittings of Mg, Zn, and Te deduced from Ref. 12. t is a linear function of x determined from the extremal values $t_{\mathrm{ZnTe}}(x=0)$ and $t_{\mathrm{MgTe}}(x=1)$ related to Phillips compounds ionicities f_{ZnTe} and f_{MgTe},[17]

$$t(x)=t_{\mathrm{Mg}_x\mathrm{Zn}_{1-x}\mathrm{Te}}=t_{\mathrm{ZnTe}}(1-x)+t_{\mathrm{MgTe}}\,x \qquad (3)$$

with

$$t_{\mathrm{ZnTe}}=\tfrac{1}{2}(1-f_{\mathrm{ZnTe}})\;,\quad t_{\mathrm{MgTe}}=\tfrac{1}{2}(1-f_{\mathrm{MgTe}})\;.$$

Again after Ref. 16, $C_0(x)$ is also a linear function of x with two constants C_{ZnTe} and C_{MgTe}:

$$C_0(x)=C_{\mathrm{ZnTe}}(1-x)+xC_{\mathrm{MgTe}}\;. \qquad (4)$$

C_{ZnTe} may be calculated using (2) for $x=0$ and then fitting C_0 to the experimental SOS for ZnTe. Unfortunately C_{MgTe} may not be determined because the experimental SOS for MgTe is unknown. Numerically, we obtain $t_{\mathrm{ZnTe}}=0.227$ and $t_{\mathrm{MgTe}}=0.223$; consequently, we suppose that $t(x)$ keeps a nearly constant value 0.225 as x varies. Elsewhere, the values of the atomic SOS are, respectively, $\Delta_{\mathrm{Mg}}=0.007$ eV, $\Delta_{\mathrm{Zn}}=0.071$ eV, and $\Delta_{\mathrm{Te}}=0.89$ eV. We see that $\Delta_{\mathrm{Mg}}, \Delta_{\mathrm{Zn}} \ll \Delta_{\mathrm{Te}}$. Then Eq. (1) becomes

$$\Delta_0 \sim C_0\times 0.755\Delta_{\mathrm{Te}}\;. \qquad (5)$$

As $C_0(x)$ varies linearly or remains constant, Eq. (5) shows that $\Delta_0(x)$ has a bowing parameter $c=0$. This is verified within experimental dispersion.

V. DISCUSSION

This analysis is not conclusive enough because it needs experimental fitting of the parameter C_{MgTe}, which is not possible because of lack of data relative to the SOS Δ_0 for MgTe. Below we give a means to evaluate this SOS by a semi-intuitive method for the study of trends in Δ_0 within the family of II-VI compounds.

A. Estimation of SOS Δ_0 for MgTe

Rustagi *et al.*[18] have shown that the time-sharing relation,

$$\Delta_0=\tfrac{1}{2}K[(1-p)\Delta_{\mathrm{III}}+(1+p)\Delta_{\mathrm{V}}]\;, \qquad (6)$$

is convenient in order to infer the SOS Δ_0 for III-V compounds. In this formula, in contrast to Berolo[16], p, which is a measure of the asymmetry in the wave function at Γ, is not identified with Phillips' ionicity but maintains the constant value $p=0.25$ throughout the III-IV family. Δ_{III} and Δ_{V} are atomic SOS for the elements of groups III and V. K is a constant given by the relation

$$K=1+\alpha_m^{2.65} \qquad (7)$$

in which α_m represents the metallicity of the compound, determined by Harrison and Ciraci.[19] We have tried to extend this approach to II-VI compounds. We have chosen $p=0.700$ for the II-VI

TABLE IV. Values of E_1, $E_1+\Delta_1$, and shift-temperature coefficient for the ternary alloys $\mathrm{Mg}_x\mathrm{Zn}_{1-x}\mathrm{Te}$ with cubic structure.

x	E_1 (eV) 300 K	E_1 (eV) 82 K	$E_1+\Delta_1$ (eV) 300 K	$E_1+\Delta_1$ (eV) 82 K	$\frac{dE_1}{dT}$ (10^{-4} eV/°K)	$\frac{d(E_1+\Delta_1)}{dT}$ (10^{-4} eV/°K)
0	3.650 ± 0.005	3.779 ± 0.003	4.245 ± 0.007	4.356 ± 0.005	−6.2 ± 0.3	−5.1 ± 0.5
0.05	3.676 ± 0.005	3.812 ± 0.003	4.274 ± 0.007	4.389 ± 0.005	−6.2 ± 0.3	−5.3 ± 0.5
0.097	3.694 ± 0.005	3.826 ± 0.003	4.298 ± 0.007	4.414 ± 0.005	−6.1 ± 0.3	−5.2 ± 0.5
0.25	3.793 ± 0.005	3.918 ± 0.003	4.383 ± 0.008	4.504 ± 0.007	−5.7 ± 0.3	−5.5 ± 0.7
0.407	3.888 ± 0.005	4.020 ± 0.003	4.446 ± 0.010	4.612 ± 0.008	−6.1 ± 0.3	−7.6 ± 1.0

TABLE V. Comparison of calculated and experimental spin-orbit splitting for II-VI compounds. References to experimental values are indicated by superscripts. The metallicities α_m (Ref. 19) and atomic spin-orbit splitting are also noted.

Compound	α_m (Ref. 19)	Δ_{II} (eV) Δ_{VI} from Ref. 12	Δ_0 (calc.) Cardona (Ref. 12)	This work	Δ_0 (exp.) (eV)
ZnTe	0.58	0.071 0.89	1.00	0.95	See Table I
CdTe	0.59	0.21 0.89	1.06	0.98	0.945 ± 0.005 (Ref. 10) (electroreflectance)
ZnSe	0.64	0.071 0.37	0.43	0.425	0.43 (reflectivity) (Ref. 20) 0.40 (reflectivity) (Ref. 9) 0.42 (electroreflectance) (Ref. 11)
ZnS	0.62	0.071 0.069	0.10	0.09	0.1 (reflectivity) (Ref. 21) 0.068 (reflectance modulation) (Ref. 22) 0.068 (from excitonic spectra) (Ref. 23)
CdS	0.64	0.21 0.069	0.14	0.12	0.07 (reflectivity) (Ref. 24) 0.066 (electroreflectance) (Ref. 7)
CdSe	0.66	0.21 0.37	0.47	0.46	0.42 (from excitonic spectra) (Ref. 24) 0.408 (reflectivity) (Ref. 25) 0.404 (electroreflectance) (Ref. 7)
HgTe	0.56	0.76 0.89	1.21	1.06	1.00 (magnetoreflection) (Ref. 26)
MgTe	0.59	0.007	1.00	0.945	0.97 (extrapolated, this work)

family by fitting the relation

$$\Delta_0 = \tfrac{1}{2}K[(1-p)\Delta_{II} + (1+p)\Delta_{VI}] \qquad (8)$$

to the experimental SOS for ZnTe, K being still given by Eq. (7). The calculations are in agreement with experimental results (Table V). This set of parameters K and p seems more convenient than the one given by Cardona[12] ($K = 1.4$ and $p = 0.6$). Consequently, such an estimate for MgTe could be valuable. As the metallicities for MgTe ($\alpha_m = 0.59$) and ZnTe ($\alpha_m = 0.58$) are close to each other and as $\frac{1}{2}(1+p)\ \Delta_{Te}$ is the dominant term in Eq. (8), we obtain nearly the same values for the SOS Δ_0 for these binary compounds ($\Delta_{0\,MgTe} \sim 0.945$ eV against $\Delta_{0\,ZnTe} \sim 0.95$ eV).

B. SOS $\Delta_0(x)$ for the MgZnTe alloys

For the ternary alloy MgZnTe we can then easily apply the Berolo-Woolley ideas with revised values taking account of the fact that the dominant term is still $\frac{1}{2}(1+p)\Delta_{Te}$ and that p remains constant for the II-VI family. Consequently, the relation (1) becomes

$$\Delta_0(x) \sim K(x)\tfrac{1}{2}(1+p)\Delta_{Te}\ . \qquad (9)$$

It is exactly similar to (5). The values for K_{MgTe} and K_{ZnTe} being the same within 1% (1.236 and 1.247), we can conclude that $\Delta_0(x)$ remains constant within experimental dispersion as x varies. Its value is about 0.95 eV. This result is in agreement with our experiments.

C. SOS $\Delta_1(x)$ for the MgZnTe alloys

The SOS Δ_1 is given by an equation similar to Eq. (1).[12] A multiplicative factor $\frac{2}{3}$ is to be used due to the fact that Γ is a triply degenerate orbital state and Λ is a doubly degenerate orbital state.[27] The matrix element is roughly the same as for Δ_0.[28,29] Hence, using the same reasoning as for Δ_0 within the Kane-Braunstein approximation and the Berolo-Woolley analysis, we assume theoretically a bowing parameter $c \sim 0$ and a nearly constant value for $\Delta_1(x)$ if the $\frac{2}{3}$ law is valid for the binary compound MgTe. (It is roughly true for ZnTe: $\Delta_1/\Delta_0 \sim 0.59/0.95$.) Experiments, especially at 82 K, confirm this assumption. The mean value $\Delta_1(x)$ for Δ_1 is about 0.59 eV. It is also an estimate of the SOS for MgTe.

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