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Electronic Raman scattering and infrared absorption spectra of shallow acceptors in ZnTe

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Electronic Raman scattering and infrared absorption have been measured for shallow acceptors As, P, and Li in ZnTe. The spectra are interpreted in terms of electronic transitions between bound hole states, some of them being phonon assisted. The valence-band parameters of ZnTe have been deduced by fitting the calculation of Baldereschi and Lipari to the experimental results. Estimated values are close to those obtained from cyclotron resonance experiments. Broadening of some electronic transition bands has been observed in both Raman and infrared spectra, which can be attributed to the mixing of the two-phonon state and the electronic excitation. TO- and LO-phonon sidebands of the electronic transition from the 1*s* to 2*s* state have been observed for the three acceptors. The energy of the LO phonon associated with this transition has been found to be smaller than that corresponding to the Γ point. The mechanism of this energy lowering is discussed.

I. INTRODUCTION

Zinc telluride is a cubic semiconductor (zinc-blende structure) with a direct band gap of 2.3 eV at 300 K. It normally shows *p*-type conduction and has only been made moderately *n*-type in heavily Al-doped Zn-saturated specimens. Shallow acceptor states are introduced by a group V*b* element substituting for tellurium or by a group I*a* element substituting for zinc.

Electrical properties of ZnTe containing shallow acceptors have been investigated by Aven[1] and Crowder and Hammer.[2] Absorption bands have been observed near 1 eV for several impurities, and these bands were attributed to transitions between the impurity level and the splitoff valence band.[3] A number of investigations on photoluminescence have been made in order to identify the energy levels associated with these shallow-acceptor impurities.[4-8] However, general agreement on the assignment of the observed levels is still lacking.

Excited states of the shallow-acceptor levels have recently been determined from electronic Raman scattering and infrared absorption on crystals containing P, Li, and As.[9,10] A rough estimation of the binding energies of the acceptors was made from photoluminescence experiments involving donor-acceptor pairs. However, some ambiguity remains in the values of the binding energies due to the lack of knowledge about donor levels in ZnTe.

Recently, Baldereschi and Lipari (BL) have applied the angular momentum theory to formulate the effective-mass theory in a simple spherical approximation and calculated the excited states of shallow acceptors.[11,12] Extension of Schechter's work[13] to materials with diamond or zinc-blende-type structure has been examined by Lin-Chung and Henvis,[14] who included the cubic symmetry term in its exact form in the Hamiltonian. The advances in these effective-mass treatments of acceptor states in semiconductors have led to the availability of accurate calculated eigenvalues for many materials.

In a very recent work we have presented an experimental determination of the binding energies of these acceptor levels by means of photoluminescence excitation spectroscopy.[15] This allows a quantitative comparison of the calculated energies of the excited states with the experimentally determined ones. The present investigation is a continuation of our previous work. Its purpose is to obtain complete and reliable excitation spectra of shallow-acceptor impurities in ZnTe. Improvement of the signal-to-noise ratio by use of a krypton or a dye laser enabled us to determine precisely the energies of the electronic transition bands as well as the phonon-assisted transition bands.

From these measurements more accurate values than the previous results[10] were obtained for the excited states, which were then compared to the theoretical calculation of BL. They use the valence-band parameters γ_1, γ_2, and γ_3 given by Lawaetz[16] in their calculation. However, Lawaetz parameters give too much anisotropy to the heavy-hole mass and too much difference between light and heavy holes. The most reliable values seem to be those obtained from cyclotron resonance experiments, which have been performed on ZnTe by Stradling.[17] Making use of his result, we have determined the band parameters by fitting the experimental results to the model of BL.

Bound hole-phonon interaction has also been studied by analyzing the phonon sidebands associated with the electronic transition.

II. EXPERIMENTAL

Single crystals of ZnTe were grown from melts containing excess Te and the desired doping agent (As, P, or none, as the case may be). Doping with Li was obtained by the diffusion method at 500°C; ZnTe crystals were dipped in $LiCO_3$ solution and then dried and heated in Zn vapor.

The concentration of the impurities was determined from the peak energy of donor-acceptor recombination bands following the procedure of Morgan *et al.*[18] For this determination it is necessary to know the sum of the binding energies $(E_D + E_A)$ for the acceptor and the donor involved in the donor-acceptor pair. This value was estimated from the measurement of photoluminescence excitation spectra.[19] For Raman measurements, a He-Ne laser and a Kr laser as well as a dye laser were used as the exciting sources. Conventional cryostats were used for the measurement at low temperatures. The 90° scattered Raman radiation was focused on a Spex model 1400 double monochromator and analyzed. The weak Raman signal was detected by a cooled S-20 photomultiplier.

The infrared absorption was measured by using two spectrometers; one was a vacuum single-beam far-infrared grating spectrometer coupled to a germanium bolometer through a light pipe system, which covered the wave-number range from 20 to 500 cm^{-1}, and the other a single-beam middle-infrared prism-grating spectrometer coupled to a Ge:Cu photoconductor-detector, which was used in the higher wave-number region above 450 cm^{-1}. The electronic transition bands were determined by comparing the transmission obtained for a "doped" crystal with that obtained for an "undoped" one.

III. RESULTS AND DISCUSSIONS

A. Electronic Raman spectra

In Fig. 1 we show a typical Raman spectrum for arsenic-doped ZnTe, where the 6471-Å line of a krypton laser was used as the exciting light. The electronic Raman band at 59 meV (*A* band) is assigned to the electronic transition from acceptor $1s_{3/2}$ state to $2s_{3/2}$ state. The weak band (*B* band) at the high-energy side of the *A* band is ascribed to a $1s \rightarrow 3s$ transition band. Two weak bands are observed between the *B* and *D* bands, which are

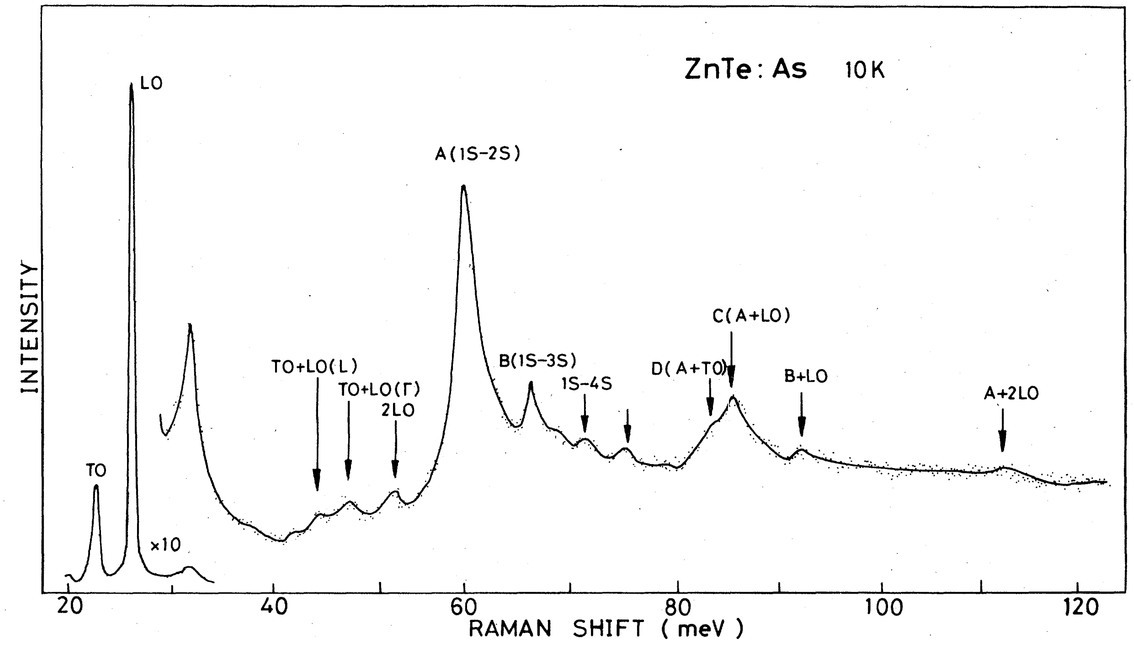

FIG. 1. Raman spectra for ZnTe:As.

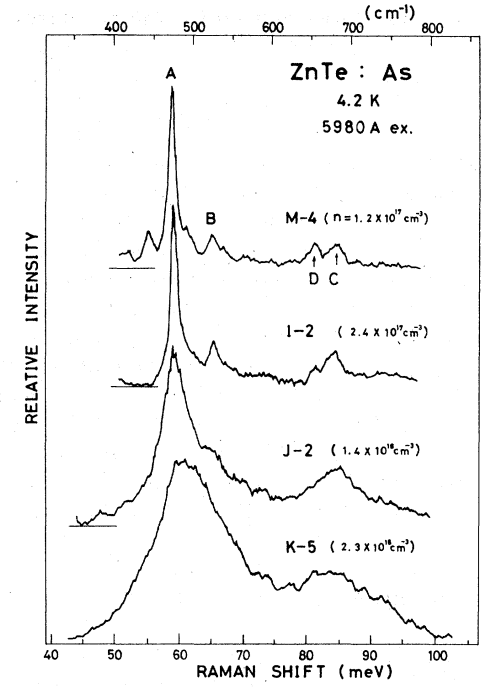

FIG. 2. Electronic Raman spectra ZnTe:As for several impurity concentrations.

located at 70.1 and 73.8 meV. The band at 70.1 meV is assigned as a $1s \rightarrow 4s$ transition. This energy is nearly equal to the value calculated by applying a simple hydrogen model to the acceptor. The binding energy of the As acceptor, 73.5 meV, is close to the value 73.8 meV. However, the band at 73.8 meV is too sharp to identify it as $1s \rightarrow$ continuum transition band. We ascribed it to a two-phonon sideband of the A band, as the calculated two-phonon density of state has a maximum at 14.4 meV in ZnTe.[20]

Electronic Raman spectra in As-doped ZnTe for several impurity concentrations are shown in Fig. 2. The linewidth of the A band increases with increasing acceptor concentration. The observed linewidth is 0.7 meV for the sample (M-4) with acceptor concentration of 1.2×10^{17} cm^{-3}. The B band broadens and merges into the A band for the samples with higher concentration. At the high-energy side of the A band, two bands are observed (C and D bands) which are separated from the A band by about 22 and 26 meV, respectively. We attribute the C and D bands to the $1s \rightarrow 2s$ transitions assisted by LO- and TO-phonon emission (phonon sidebands), respectively. While these two bands are observed separately for the sample with lower concentration, these bands broaden with the concentration and overlap each other for higher concentration. As the intensity of the LO-phonon sideband is stronger than the TO-phonon sideband, the peak of the composite band is located at the position of the LO-phonon sideband. In Fig. 1, the D band appears as a shoulder of the C band, and two other very weak bands can be observed which correspond energetically to B + LO and A + 2LO.

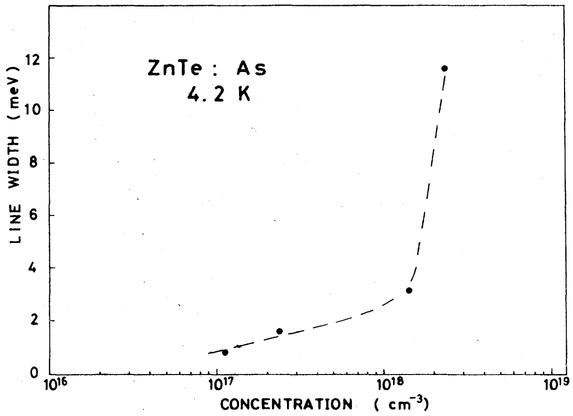

FIG. 3. Linewidth of the A band as a function of arsenic concentration.

Figure 3 shows the linewidth of the A band as a function of acceptor concentration. Here, the acceptor concentration deduced from the luminescence spectra of the donor-acceptor pairs is not the uncompensated one, but the net concentration. The compensation ratio of the sample studied here is not known. But it is considered to be in the range from 0.3 to 0.7.[21] The linewidth shows a marked increase at about 1×10^{18} cm^{-3}. This may be due to the impurity-band formation. The minimum linewidth obtained for the A band is 0.7 and 0.4 meV for As, and Li acceptors, respectively. These values are large compared to the value of ~0.1 meV, which has been observed in photoconductivity measurements in GaAs.[22] The linewidth of the electronic transition at low temperatures is considered to arise predominantly from inhomogeneous Stark shift of the energy levels by random electric fields arising from ionized donors and acceptors around the impurity of interest.[23] Our rather large values of the linewidth could indicate that the inhomogeneous broadening is a dominant mechanism of the linewidth for the concentration studied here.

The electronic Raman spectra for ZnTe:P and ZnTe:Li show a peculiar feature compared to that of As acceptor, as shown in Fig. 4. The B band corresponding to the transition $1s \rightarrow 3s$, is always broad and interferes with the 2LO-phonon band which is close to it. The linewidth of the purely electronic A band in ZnTe:Li is narrow and equal to 0.4 meV. However, the B band which is located near the 2LO phonon band is broad.

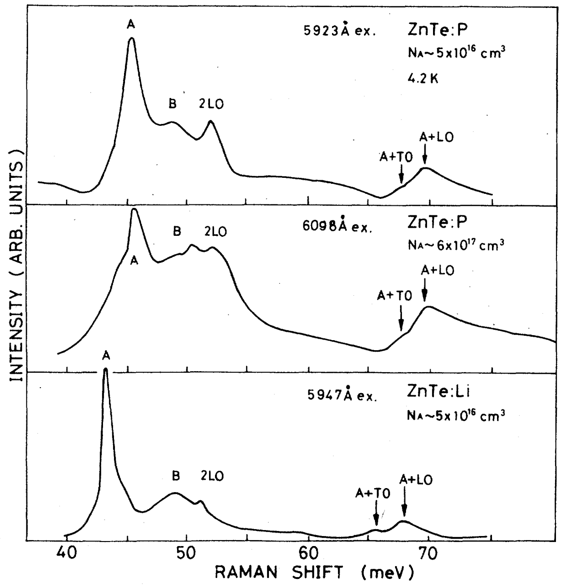

FIG. 4. Typical electronic Raman spectra for phosphorus and lithium acceptors in ZnTe.

Such a broadening effect is also observed in the infrared absorption spectra of Si (Ref. 24) when the excitation energy is comparable to that of the optical phonon. These results suggest that the $1s \rightarrow 3s$ excitation is mixed with the 2LO-phonon excitation. For P and Li acceptors, TO- and LO-phonon sidebands are not well resolved. Attempts to observe two separated bands in the sample with low impurity concentration did not succeed, probably because of the very weak intensity of the phonon sidebands.

B. Infrared absorption

Infrared absorption spectra associated with electronic transitions between impurity levels have been measured at liquid-He temperature, results of which are shown in Fig. 5. In the low-energy region, a sharp band (denoted as *a* band) is observed at 39.6 meV for P and 37.6 meV for Li acceptors. These bands are assigned to $1s_{3/2}$-$2p_{3/2}$ transitions. For As acceptor, we have observed weak bands at 51.0 and 52.9 meV, and a broad band at 47.4 meV, which was ascribed to the $1s_{3/2}$-$2p_{5/2}$ transition band in the previous paper.[9] However, for the reason described later the band at 52.9 meV is identified as this transition. The spectra for As acceptor reveal two strong bands (*b* and *c* bands) at 61.2 and 64.7 meV which are readily assigned to $1s_{3/2}$-$2p_{5/2}(\Gamma_8)$ and $1s_{3/2}$-$2p_{5/2}(\Gamma_7)$. The strong absorption bands observed in the region around 45 meV are due to two-phonon excitation.

It is reasonable to consider that these absorption bands are not associated with impurity-induced phonons but with the electronic transition bands, although there have been no data on impurity-induced phonon bands for ZnTe containing Li, P, and As; if we apply the mass defect model,[25] we do not expect that the frequencies of these impurity modes lie in the region where the electronic bands are observed in Fig. 5. Localized

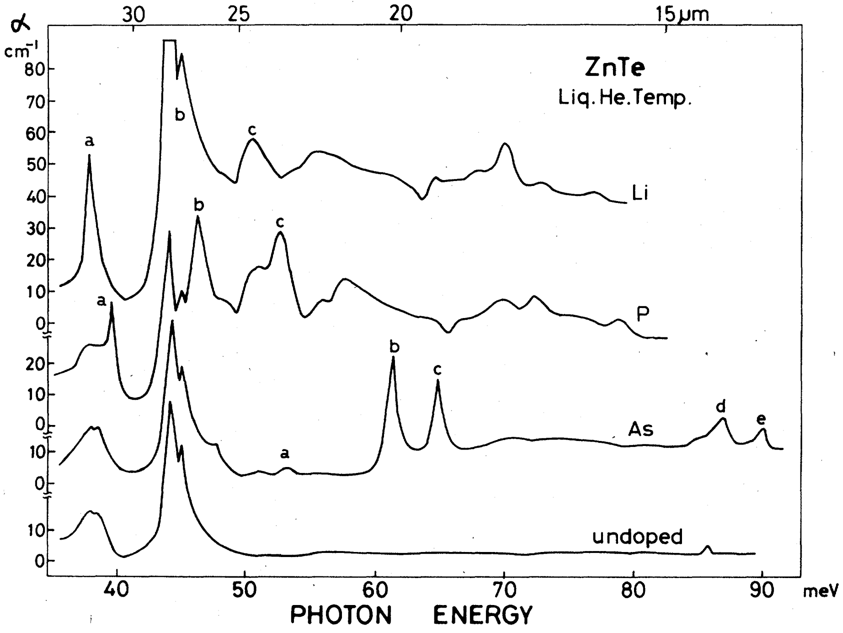

FIG. 5. Infrared absorption spectra for Li, P, and As acceptors in ZnTe.

modes in 2–6 compounds were generally observed in highly doped samples, e.g., for ZnTe containing Al impurity[26] of 10^{18} cm^{-3}. The absorption coefficients of the observed bands in Fig. 5 are too large to interpret these bands as the impurity-induced phonon modes.

For P and Li acceptors the energies of the transitions, $1s$-$2p_{5/2}$, are comparable to the two-phonon states.[27] These transition bands (*b* and *c* bands) for both of the acceptors are broad compared to each *a*-band. These facts suggest a resonant interaction of the two-phonon states and the $1s_{3/2} \rightarrow 2p_{5/2}$ transition, as in the Raman spectra of these acceptors. Chandrasekhar *et al.*[24] have measured the excitation spectra of the Ga acceptor in silicon. They have found that the band corresponding to the $1s(\Gamma_8) \rightarrow 2p(\Gamma_8)$ transition energy is comparable to the optical phonon energy and shows anomalous broadening. This feature has been interpreted as the mixing of the electronic excitation and the optical phonon state. While the relevant phonon state is the one-phonon state in Si, it will be possible for two-phonon states to couple with the electronic excitation if the electron-phonon interaction is strong. For As-doped crystals the LO-phonon sidebands of the *b* and *c* bands are observed, and are denoted by *d* and *e* bands, respectively, in Fig. 5, whereas we could not observe TO-phonon sidebands. Because of the strong background for Li- and P-doped crystals, we could resolve neither LO- nor TO-phonon sidebands in these crystals.

C. Electron-phonon interaction

We compare the Raman spectra for three different acceptors in Fig. 6, where a portion of the optical-phonon sideband of the *A* band is shown. Although the TO-phonon sideband is not resolved for Li- and P-doped samples in spite of their low concentration, it is clearly observed as a shoulder on the low-energy side of the LO-phonon sideband for Li acceptor. The energy separation between the $1s \rightarrow 2s$ zero-phonon transition (*A* band) and its LO-phonon sideband is 25.4 ± 0.5 meV for the As acceptor. The separation for P and Li acceptors is 23.8 and 24.7 ± 0.5 meV, respectively, which is slightly smaller than $\hbar\omega_l(\Gamma) = 26.1$ meV at 4.2 K. Judging from the band shape of the optical-phonon sideband of the *A* transition, it is not considered that the lowering of the vibrational energy is due to the shift resulting from the overlap of the TO- and LO-phonon sidebands.

The LO-phonon sideband of the electronic transition in donors,[28] acceptors,[10] and excitons[29] have been observed in a number of crystals whose vibrational energy of these modes are lower than the LO-phonon energy at the Γ point. Toyozawa and Hermanson[30] have shown that the electron-phonon interaction leads to an attractive interaction which lowers the energy of the LO-phonon sideband of the exciton. They predicted that in some circumstances a bound state consisting of an exciton-LO-phonon complex could occur. In the subsequent paper Toyozawa[31] pointed out that the exciton-phonon quasibound state could be formed even if the phonon energy is greater than the binding energy of the exciton.

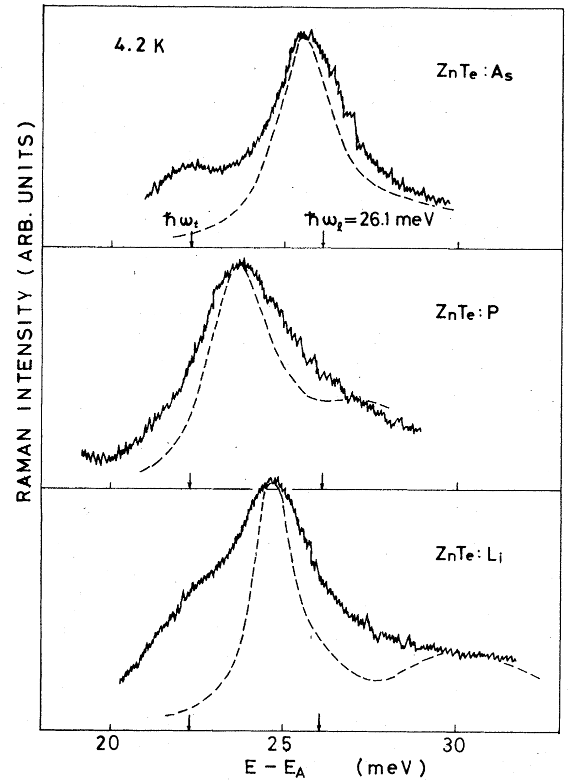

FIG. 6. LO-phonon sidebands of the *A* band for the three acceptors. The line shapes of the zero-phonon band (dashed lines) normalized in intensity to the sideband are shown in comparison. The abscissa is the energy shift from the position of each zero-phonon band.

The LO-phonon local modes bound to neutral donors have been observed in Raman scattering, luminescence,[32] and infrared reflection[33] spectra of GaP crystals. Dean, Manchon, and Hopfield[32] have shown that the interaction of the donor electron with the LO phonon is attractive and that a local mode is formed when the LO-phonon energy is less than the energy of the first excited state of the donor electron.

Henry and Hopfield[34] have shown that the LO-phonon sideband of the I_2 line (decay of exciton bound to a neutral donor) in CdS and CdSe is shifted to lower energy compared to the nominal

LO-phonon energy $\hbar\omega_l$. In these crystals the LO-phonon energy is greater than the donor binding energy E_D. A bound state can not be expected because such a state would be degenerate with the free-electron continuum of the donor. The attractive interaction which is necessary to produce the quasibound state is caused by the virtual excitation of the donor electron to the continuum state whose energy is greater than the energy of the 1*s* electron plus the LO- phonon, i.e., E(continuum) $>E_{1s}+\hbar\omega_l$. This quasibound state is closely related to the exciton-phonon quasibound state previously proposed by Toyozawa.

In ZnTe, the LO-phonon sideband of the *A* band corresponds to the bound state hole at the 2*s* level plus LO-phonon state. This state is degenerate with the continuum state of the acceptor for all three acceptors. We might expect that the LO-phonon–2*s* bound hole complex would form a quasibound state in analogy with the exciton-LO-phonon quasibound state discussed by Henry and Hopfield. Their calculation indicates that the line shape of the I_2 line LO-phonon replica is structured and broadened compared to the zero-phonon line. We observed no structure in the optical-phonon sideband of the *A* transition except for the TO-phonon sideband. However, the line shape of the LO-phonon sideband is broadened as is seen in Fig. 6, where the zero-phonon band normalized to the one LO-phonon band in its peak intensity is plotted by dashed lines at the same position as its sideband. After subtracting the contribution of the TO-phonon sideband, the LO-phonon sideband is still broader than its zero-phonon band. The broadening of the phonon sideband may be due to the decay of the hole-LO-phonon quasibound state to the continuum state of the acceptor.

The intensities of the electronic transition bands accompanying *n*th LO-phonon emission are in general described by the Poisson distribution function $I_n=I_0(S)^n/n!$, where S is the electron-LO-phonon coupling constant and I_0 is the intensity of the zero-phonon band. The coupling constant S for As, Li, and P acceptors is estimated from the intensity ratio of the zero-phonon band to the one LO-phonon sideband in the Raman spectra. It seems to depend somewhat upon the impurity concentration. A rough estimation of the coupling constant gives us the values of 0.24–0.15, 0.18–0.14, and 0.30–0.17 for As, Li, and P acceptors, respectively. These values are comparable to those obtained from the intensity ratio of the LO-phonon sidebands of the donor-acceptor pair luminescence, which are 0.3, 0.16, and 0.19 for As, Li, and P, respectively. It is to be noted that the energy shift of the LO-phonon sideband in the Raman spectra does not show a dependence on the strength of the coupling between the bound hole and the LO phonon. Of these three acceptors the shift for the As acceptor is the least, whereas its coupling constant is not the least. This fact may suggest that the energies of excited and ground states for the acceptor relative to the LO-phonon energy dominate the magnitude of the energy shift for these quasibound states. However, a quantitative explanation of these energy shifts awaits a more detailed theoretical study.

As is seen in Figs. 2 and 6, the intensity of the TO-phonon sideband of the *A* transition is somewhat comparable to the LO-phonon sideband for As-doped crystals. Such strong optical sidebands associated with shallow levels have not yet been observed in other materials. Weak zone-boundary-phonon sidebands were observed in GaP.[35-37] The TO-phonon sidebands associated with the electronic transition have also been observed in the photoluminescence excitation spectra[19] of donor-acceptor pairs in ZnTe. It is to be noted that the TO-phonon sidebands in both cases are associated with the excited levels of the electronic states.

D. Determination of the valence-band parameters

Excited states experimentally determined for the three acceptors in ZnTe are listed in Table I. The 1*s*-2*s* separation for As and Li acceptors is smaller than the previous results[10] by 0.2 and 0.4 meV, respectively. The results of the Raman scattering and infrared absorption give us only the energy difference between the excited states and the ground state, and hence the binding energies cannot be obtained from either experiment. From photoluminescence excitation spectra for donor-acceptor pairs in doped ZnTe, we have estimated the values of these binding energies to be 73.5, 60.2, and 58.3 ± 0.5 meV for As, P, and Li acceptors, respectively. The chemical shift for the 2*s* state is very small; 2*s*-state energies for these acceptors coincide

TABLE I. Observed energies of the acceptor states. Fitting values are obtained using the theoretical treatment of Baldereschi and Lipari. The experimental error is ±0.5 meV.

States	Acceptors			Fitting value
	As	P	Li	
$1s_{3/2}$ (Γ_8)	73.5	60.2	58.3	51.4
$2p_{3/2}$ (Γ_8)	20.7	20.6	20.7	20.6
$2s_{3/2}$ (Γ_8)	14.5	14.8	14.7	14.6
$2p_{5/2}$ (Γ_8)	12.3	13.7	13.5	13.8
$2p_{5/2}$ (Γ_7)	8.8	7.2	7.7	10.4
3*s*	8.41	9.62	9.07	···

with each other within experimental error. The $2p_{3/2}$-state energies for Li and P acceptors show excellent agreement. For As acceptor there was an ambiguity in the identification of this state; there are two candidates[9] (the bands at 47.4 and 52.9 meV) for this state. Since the noticeable chemical shift is not observed for the $2s$ state, it is not expected for the $2p_{3/2}$ state, either. We choose in the present work the absorption band at 52.9 meV to correspond to the $1s \rightarrow 2p_{3/2}$ transition. There is a slight discrepancy in the $2p_{5/2}$ states for P and Li acceptors. As was discussed in Sec. III B, it may originate from the mixing or the interference effect of the two-phonon state and the $1s$-$2p_{5/2}$ transitions. The same will occur for the $1s \rightarrow 3s$ transition in both of the acceptors. The energy shift of the $3s$ state for Li and P might be due to this interference effect.

In their treatment of the problem of acceptors in cubic semiconductors, BL rewrote the acceptor Hamiltonian as the sum of two terms, one having a cubic symmetry and the other a spherical symmetry. The energies of the acceptor states were calculated by treating the cubic term as a perturbation. Their calculation was made for various sets of the band parameters, μ and δ, where the spherical coupling parameter μ describes the strength of the spherical spin-orbit interaction and the cubic coupling parameter δ represents the strength of the cubic contribution. These two parameters are related to the Luttinger valence-band parameters γ_1, γ_2, and γ_3 by

$$\mu = (6\gamma_3 + 4\gamma_2)/5\gamma_1 ,$$

$$\delta = (\gamma_3 - \gamma_2)/\gamma_1 .$$

Using their procedure, we have determined the Luttinger band parameters so that the theoretical energies fit the experimental values shown in Table I. In this fitting procedure we have chosen the energies of $2p_{3/2}(\Gamma_8)$ and $2s_{3/2}$ states as the most reliable values for the determination because the mixing of the phonon and the electronic transition seems to be absent for these states.

The fitted energies of the acceptor states are listed in Table I. The obtained valence-band parameters γ_1, γ_2, and γ_3 are compared with other authors' values in Table II. Our valence-band parameters are comparable to the values obtained from cyclotron resonance experiment by Stradling. Taking into account the experimental errors, the agreement seems to be quite satisfactory.

TABLE II. Comparison of the valence-band parameters.

	Valence-band parameters		
	γ_1	γ_2	γ_3
Lawaetz[a]	3.74	1.07	1.64
Stradling[b]	4.00	1.15	1.29
Present work[c]	$4.20^{+0}_{-0.2}$	$0.91^{+0.1}_{-0.4}$	1.54 ± 0.1

[a] See Ref. 16.
[b] See Ref. 17.
[c] Static dielectric constant $\epsilon_0 = 9.86$ was used for the calculation.

However, it is to be noted that our γ_2 value determined in this procedure is rather small compared to Stradling value. As is shown in Table I, the calculated energy of the $2p_{5/2}(\Gamma_7)$ state deviates slightly from the experimental values. If we try to fit the energy of this state to the experimental values, γ_2 falls down to 0.6–0.7. A small value of γ_2 means that the strength of the cubic contribution δ is rather big. Although such values of γ_2 are beyond the experimental error of cyclotron resonance, the possibility of a smaller γ_2 value would not be ruled out.

More recently Dean *et al.*[38] have estimated the binding energies of shallow acceptors in ZnTe by the measurements of the photoluminescence due to the two-hole transition from the bound exciton and the bound-to-acceptor transition. Their estimated values are ~60 meV for Li, 63.5 meV for P, and ~77 meV for As, which are close to our values.

In conclusion, electronic Raman scattering as well as infrared absorption seem to be very good tools to study the energy levels associated with impurities in semiconductors and eventually the electron-phonon interaction. The effective-mass treatment describes quite satisfactorily the excited states of shallow acceptors in ZnTe, with the result that the position of the states is independent of the nature of the impurity. A central cell correction is needed to explain the chemical shift of the ground state of the impurity.

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