

Photoemission Study of the Electronic Structure of ZnTe*

J. L. SHAY† AND W. E. SPICER

Stanford Electronics Laboratories, Stanford University, Stanford, California 94040

(Received 18 June 1968)

We report the results of ZnTe photoemission studies over a range in photon energy extending from 6.0 to 11.6 eV. Single crystals were cleaved and studied at pressures less than 10^{-9} Torr. The energy distributions of the photoemitted electrons are rich in structure, and contain as many as six distinct peaks in the electron-energy distribution at a single photon energy. By studying the motion and changes in strength of this structure as the exciting photon energy is varied, we demonstrate that all of it is due to direct transitions. By comparing the experimental data with recent pseudopotential and orthogonalized-plane-wave calculations, we determine the energies of many states over a range extending from 4 eV below to about 10 eV above the top of the valence band. Transitions are assigned primarily to Γ , X , and L and along Σ , Δ , and A on the basis of the one-electron band structure. Experimental values for energy states are generally within a few tenths of an eV of theory, particularly for valence bands, although there may be a discrepancy at the conduction-band level L_3 .

I. INTRODUCTION AND CONCLUSIONS

IN the present work, we have used photoemission experiments to study the electronic structure of ZnTe over a range of 14 eV extending from about 4 eV below to 10 eV above the top of the valence band. Whereas conventional optical experiments determine only the energy differences between quantum levels, photoemission measurements determine the absolute energies of the levels involved in an electronic transition. Another feature of photoemission experiments is the ability to decompose the total electronic transition probability for a given photon energy according to the different possible final energies. For example, in a later section we show that whereas the reflectivity of ZnTe near 11.0 eV displays only one broad peak, the energy distributions of the photoemitted electrons at the same photon energies show six distinct pieces of structure. Hence these six different transitions, taking place for the same photon energy, can be observed simultaneously in the photoemission experiment.

After studying this structure over a broad range of photon energy, we have concluded that the major features of the photoemission from ZnTe are due to direct transitions in which the electronic wave vector k is conserved. In this respect the photoemission¹⁻⁴ from ZnTe and that from CdTe are very similar. However, previous studies of CdSe^{4,5} and CdS⁴⁻⁶ showed

that in addition to several direct transitions, in these materials there were a significant number of transitions for which k conservation was unimportant. We notice that both CdTe and ZnTe crystallize in the zinc-blende structure, whereas CdSe and CdS normally have the wurtzite structure. At present there is no detailed explanation for this correlation between crystal symmetry and the relative importance of k conservation.

As already mentioned, most of the photoemission structure results from direct transitions. With the assistance of the orthogonalized-plane-wave band calculation of Herman⁷ and the pseudopotential calculation of Cohen and Bergstresser,⁸ we have identified the regions of the Brillouin zone responsible for most of the structure. These identifications allow us to determine the absolute energies of many levels in the band structure of ZnTe. It must be emphasized that the identification of a given peak in the energy distributions as resulting from a specific interband transition relies heavily upon the correctness of the theoretical band calculation. If there are gross errors in the band structure, portions of the photoemission data may need reinterpretation.

On the whole, our experimental results and the theoretical band calculations agree within about 0.5 eV although some discrepancies are as large as 1.1 eV. Since the estimated uncertainty in the photoemission data is ± 0.2 eV, either some of our identifications are incorrect, or the theoretical band structures are in error in some regions. Nonetheless, there appears to be sufficient agreement between theory and experiment in many regions of the Brillouin zone to demonstrate the essential validity of the theoretical one-electron band structure for ZnTe even for states which are as much as 10 eV from the top of the valence band.

* Experimental work supported by the National Aeronautics and Space Administration, the Advanced Research Projects Agency through the Center for Materials Research at Stanford University, and the National Science Foundation.

† Present address: Bell Telephone Laboratories, Holmdel, N. J.

¹ J. L. Shay, W. E. Spicer, and Frank Herman, *Phys. Rev. Letters* **18**, 649 (1967).

² J. L. Shay and W. E. Spicer, *Phys. Rev.* **161**, 799 (1967).

³ J. L. Shay and W. E. Spicer, in *Proceedings of the International Conference on II-VI Semiconducting Compounds*, Providence, 1967 (W. A. Benjamin, Inc., New York, 1967), p. 651.

⁴ J. L. Shay, Ph.D. dissertation, Stanford University, 1966 (unpublished); Solid State Electronics Laboratories Technical Report No. 5216-1 (unpublished).

⁵ J. L. Shay and W. E. Spicer, *Phys. Rev.* **169**, 650 (1968).

⁶ N. B. Kindig and W. E. Spicer, *Solid State Commun.* **2**, 13 (1964); *Phys. Rev.* **138**, A561 (1965).

⁷ F. Herman, R. L. Kortum, C. D. Kuglin, and J. L. Shay, in *Proceedings of the International Conference on II-VI Semiconducting Compounds*, Providence, 1967 (W. A. Benjamin, Inc., New York, 1967), p. 503.

⁸ M. L. Cohen and T. K. Bergstresser, *Phys. Rev.* **141**, 789 (1966).

II. EXPERIMENTAL METHODS

Most of the experimental methods used in this work are identical to those reported¹⁻⁶ in similar studies of CdTe, CdSe, and CdS, so a detailed discussion will not be repeated here. The experiments have been performed at room temperature on ZnTe single crystals cleaved in ultrahigh vacuum in the (110) plane using the cleaving chamber and vacuum system described by Powell.⁹ The experiments were performed at pressures less than 10^{-9} Torr and required about 4 to 5 h to obtain a complete set of energy distributions. Energy distributions measured at the end of this period were indistinguishable from others at the same photon energy measured immediately (~ 5 min) after the crystal had been cleaved. The largest single crystals were $4\text{ mm} \times 4\text{ mm} \times 4\text{ mm}$, and on account of the very small sample size, it was necessary to introduce an aperture between the crystal and the exit slits of the monochromator to prevent light from striking parts of the equipment other than the crystal under study.

The Cs₃Sb phototube used to measure light intensity has been calibrated¹⁰ against a vacuum thermopile with an estimated accuracy of $\pm 10\%$. Since it was difficult to measure the intensity of the incident radiation with the aperture in place, the absolute values of the quantum yield may be in error by as much as 30%. Nonetheless, relative values of the yield at different photon energies should be accurate within about 10%. Hence, the relative heights of energy distributions at different photon energies should be correct within about 10%.

In our previous studies¹⁻⁶ of CdTe, CdSe, and CdS, we performed experiments both in ultrahigh vacuum and also at pressures of about $0.1\ \mu$. These latter experiments produced an electron affinity about 1 eV less than that for a crystal cleaved in ultrahigh vacuum. After verifying that this was the only major effect of the poorer vacuum, we proceeded to deduce a great deal of information from the photoemission from the conduction bands uncovered by the lower electron affinity. In ZnTe we find that similar low-vacuum experiments do not appreciably reduce the electron affinity. However, in one experiment in which the crystal was cleaved at the very high pressure of several microns, the resulting electron affinity was much lower than for a crystal cleaved in ultrahigh vacuum. This experiment was briefly reported elsewhere.³ For photon energies greater than those previously reported, the inelastic scattering smeared out all of the structure. Hence, these data are not included in this paper.

III. PHOTOEMISSION DATA

In this section we present the results of photoemission experiments performed using single crystals of

⁹ R. J. Powell, Ph.D. dissertation, Stanford University, 1967 (unpublished); Solid State Electronics Laboratories Technical Report No. 5220-1 (unpublished).

¹⁰ Much of this work has been performed in our laboratory by R. Koyama.

ZnTe cleaved and studied at pressures less than 10^{-9} Torr. In Sec. IV we deduce from the photoemission data the energies of several levels in the electronic band structure of ZnTe and compare these results with the pseudopotential band calculation of Cohen and Bergstresser⁸ and the orthogonalized-plane-wave (OPW) band calculation by Herman.⁷

A. Quantum Yield

In Fig. 1 we present the absolute quantum yield of ZnTe as a function of the exciting photon energy. Relative values of the yield at different photon energies should be accurate within about 10%. However, for the reasons discussed in Sec. II the absolute yield at a single photon energy may be in error by as much as 30%. It is apparent from Fig. 1 that the threshold for photoemission is slightly less than 6.0 eV. A more accurate value can be obtained from the zero intercept of the energy distributions in Sec. III B which we find to be 5.6 eV. Since the band gap¹¹ at room temperature is 2.25 eV, the electron affinity for a clean surface of ZnTe is 3.35 ± 0.2 eV.

B. Energy Distribution Curves

In Figs. 2 to 5 we present normalized energy distributions (NED) of the photoemitted electrons for various photon energies between 6.4 and 11.4 eV. All NED have been normalized to the yield in Fig. 1 so that the abscissa is calibrated in electrons per incident photon per electron volt. Electron energies are stated relative to the valence band maximum with an estimated accuracy of ± 0.2 eV. For a discussion of the calibration of the energy scale see Refs. 4 and 6.

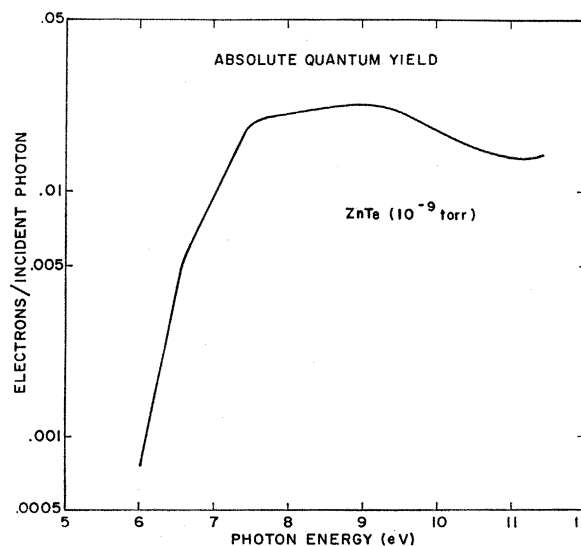


FIG. 1. Absolute quantum yield for a single crystal of ZnTe cleaved at a pressure less than 10^{-9} Torr.

¹¹ M. Cardona, K. L. Shaklee, and F. H. Pollak, Phys. Rev. 154, 696 (1967).

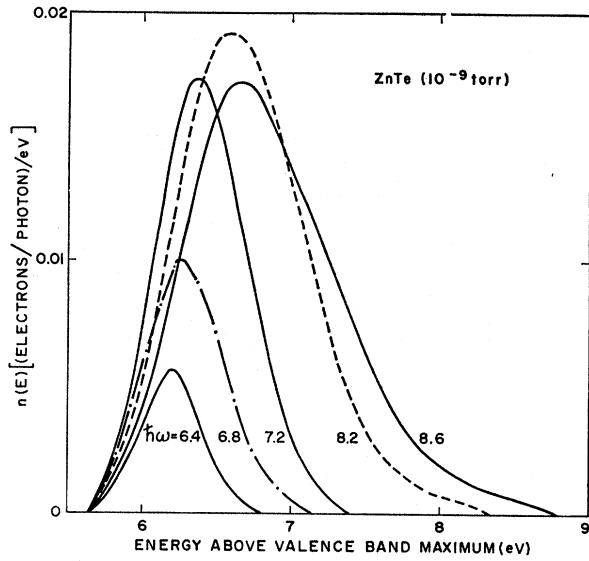


FIG. 2. Normalized energy distributions of the photoemitted electrons.

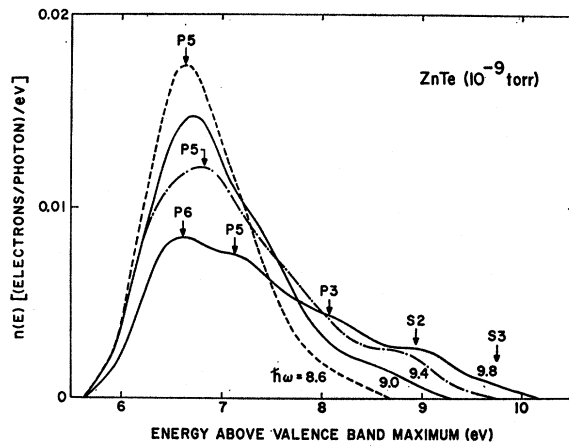


FIG. 3. Normalized energy distributions of the photoemitted electrons.

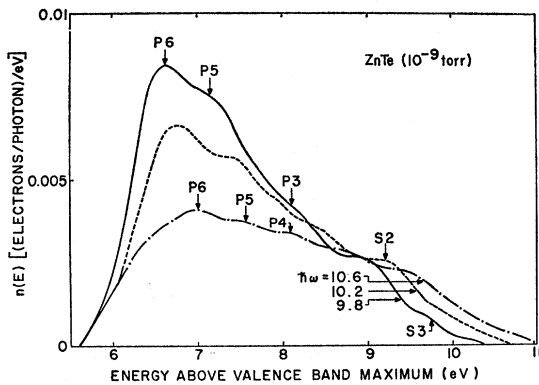


FIG. 4. Normalized energy distributions of the photoemitted electrons.

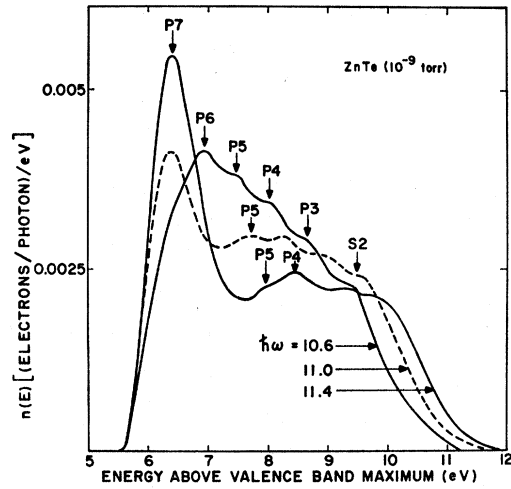


FIG. 5. Normalized energy distributions of the photoemitted electrons. The peak *P7* is due to the escape of inelastically scattered electrons which have created electron-hole pairs.

The shoulders *S2* and *S3* and the peaks *P3* through *P7* are distinct pieces of structure in the NED. In order to study the systematics of this rich collection of structure, we present in Fig. 6 a structure plot which summarizes the location in energy of all the structure observed for $6.4 \leq \hbar\omega \leq 11.6$. Each "p" represents a peak in the NED at the electron energy of the ordinate for the photon energy given by the abscissa. Similarly each "s" represents the occurrence of a shoulder in the NED. The 45° lines are lines of constant initial-state energy.

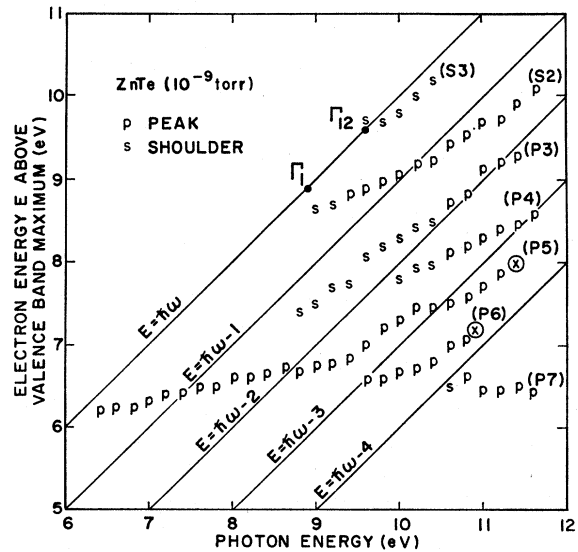


FIG. 6. Structure plot summarizing the structure in the ZnTe energy distributions. Each *p* indicates the occurrence of a peak in the energy distribution at the energy of the ordinate for the photon energy of the abscissa. Similarly, each *s* indicates the occurrence of a shoulder in the energy distributions. The labels in parentheses correspond to the structure in Figs. 2-5. The circled *x*'s represent the abrupt disappearance of peaks. The 45° lines are lines of constant initial-state energy.

They are included so that one can determine at a glance the energy of the initial state for any of the entries. Structure plots were first introduced by Eden and Spicer¹² in their studies of GaAs, GaP, and Si.

In our study¹³ of CdTe, we demonstrated the power of photoemission experiments by comparing the reflectivity¹⁴ curve for all photon energies with a single NED for $\hbar\omega=9.9$ eV. Whereas the reflectivity of CdTe near $\hbar\omega=9.9$ eV displayed only one broad peak, an NED at the same photon energy had *five* distinct pieces of structure. The reflectivity spectrum gave little or no indication of the variety of different transitions taking place in CdTe for photon energies near 9.9 eV. The situation is even more dramatic in ZnTe. Whereas the reflectivity¹⁴ near 11.0 eV displays only *one* broad peak, the NED for photon energies near 11.0 eV (Fig. 5) show *six* distinct pieces of structure (*S2*, *P3* through *P7*). A study of the photon energy dependence of the locations and shapes of the photoemission structure provides information about key features of the band structure even in this absence of sharp structure in the optical data.

The peaks *P5* and *P6* are unique in that after appearing in the NED for a range of photon energy, they abruptly disappear from the NED. The points at which these peaks disappear are indicated by circled x's in Fig. 6. When *P6* is last seen for a photon energy of 10.9 eV, it is due to transitions from initial states at -3.7 eV in the valence band to final states near 7.2 eV in the conduction band. When *P5* is last seen for a photon energy of 11.4 eV, it is due to transitions from initial states near -3.4 eV to final states near 8.0 eV. In Sec. IV we show that these peaks disappear since they have reached the bottom of certain valence bands. We locate the regions of the band structure responsible for these peaks by comparing the energies at which they disappear with theoretical band calculations.

The disappearance of peaks *P5* and *P6* is one indication that they result from direct transitions,^{2,4,5,15} i.e., transitions in which the electronic wave vector \mathbf{k} is conserved. The strength of a direct transition to an energy E is proportional to the number of \mathbf{k} -conserving transitions possible to that energy. When the details of the band structure no longer permit \mathbf{k} -conserving transitions, the corresponding feature of the NED (and optical data) disappears. Another characteristic of direct transitions is that structure moves to higher energy in the NED with increases in energy different from the increases in photon energy, i.e., the energy of the initial state is a function of $\hbar\omega$. Inspection of Fig. 6 shows that the initial-state energies for most of the

structure vary considerably. We are thus led to conclude that the structure in the photoemission from ZnTe results principally from direct transitions.

When the shoulders *S2* and *S3* first appear in the NED for $\hbar\omega=8.9$ and 9.6 eV, respectively, they lie at the high-energy side of the NED. Hence, they are due to transitions from initial states near the top of the valence band. Since it seems well established¹⁶ that the valence-band maximum in ZnTe lies near Γ ($\mathbf{k}=0$), these shoulders locate two Γ conduction bands at 8.9 and 9.6 eV above the valence-band maximum. This is one situation in which the important region of the Brillouin zone can be inferred directly from the photoemission data completely independently of any theoretical calculations.

For $\hbar\omega \geq 10.8$ eV, the peak *P7* appears in the NED at an energy of about 6.5 eV. This stationary peak of low-energy electrons is characteristic¹⁷⁻¹⁹ of a peak due to the escape of inelastically scattered electrons which have created electron-hole pairs. Similar scattering peaks have been seen in the photoemission from many other materials.

IV. BAND STRUCTURE OF ZnTe

We showed in Sec. III that the structure in the photoemission from ZnTe is due to direct transitions in which the electronic wave vector \mathbf{k} is conserved. However, it would not be possible to determine the regions of the Brillouin zone responsible for most of the structure without recourse to a theoretical band calculation. By comparing the photoemission data with the band calculations shown in Fig. 7, we are able to determine the energies of several levels in the electronic band structure. The dotted lines are the result of Cohen and Bergstresser's⁸ pseudopotential calculation and the solid points are the result of Herman's⁷ orthogonalized-plane-wave calculation. Neither calculation included spin-orbit splitting, which in ZnTe¹¹ has the effect of lifting the top of the valence band in the crystal (Γ_8) by 0.3 eV relative to Γ_{15g} , the valence-band maximum in the absence of spin-orbit coupling. Since the theoretical energy levels^{7,8} have been given relative to Γ_{15g} , we have subtracted 0.3 eV from all theoretical energies before comparing them with experimental values.

Let us first discuss the shoulders *S2* and *S3*. As shown in Sec. III, when they first appear they result from transitions from the top of the valence band at Γ_8 to higher-lying Γ conduction bands at 8.9 and 9.6 eV. On the basis of previous theoretical^{7,8} and experimental^{1-4,12} work on similar zincblende materials, one expects to

¹² R. C. Eden and W. E. Spicer (to be published); R. C. Eden, Ph.D. dissertation, Stanford University, 1967 (unpublished); Solid State Electronics Laboratories Technical Report No. 5221-1 (unpublished).

¹³ See, especially Fig. 1 of Ref. 3.

¹⁴ M. Cardona and D. L. Greenaway, Phys. Rev. **131**, 98 (1963).

¹⁵ W. E. Spicer, Phys. Rev. **154**, 385 (1967); **161**, 526(E) (1967).

¹⁶ B. Segall, in *Physics and Chemistry of II-VI Compounds*, edited by M. Aven and J. S. Prener (North-Holland Publishing Co., Amsterdam, 1967), p. 3, and references therein.

¹⁷ C. N. Berglund and W. E. Spicer, Phys. Rev. **136**, A1030 (1964); **136**, A1040 (1964).

¹⁸ L. Apker, E. A. Taft, and J. Dickey, J. Opt. Soc. Am. **43**, 78 (1953).

¹⁹ W. E. Spicer, J. Phys. Chem. Solids, **22**, 365 (1961).

find the conduction bands Γ_{1c} and Γ_{12c} about 9 or 10 eV above Γ_8 , with Γ_{1c} lying somewhat below Γ_{12c} . Hence, our photoemission data locate Γ_{1c} at 8.9 eV and Γ_{12c} at 9.6 eV in ZnTe.

We cannot adequately compare our value for Γ_{1c} with theoretical predictions, since the pseudopotential calculation was limited to energies below 8.2 eV and since Herman has used our result for Γ_{1c} as one of the experimental conditions for his adjusted OPW band model. However, since Γ_{1c} is the only photoemission result used by Herman in his adjustments,¹ we can compare our value for Γ_{12c} with the OPW result. Herman predicts that Γ_{12c} lies 10.2 eV above Γ_{8v} , whereas our experimental value is 9.6 eV. Although the discrepancy of 0.6 eV is only 6%, one would hope to find better agreement between theory and experiment. On the other hand, the photoemission structure $S3$ used to locate Γ_{12c} is the weakest feature of our data, and as such may be subject to error. Although we are able to locate $S2$ and $S3$ near $\mathbf{k}=0$ when they first appear, the theoretical calculations are insufficient at these high energies to trace the motion of $S2$ and $S3$ as $\hbar\omega$ is varied.

Consider now the photoemission peak $P3$. The structure plot in Fig. 6 shows that when the peak first appears it is due to initial states near -1.5 eV. As the photon energy is increased the initial state gradually sinks deeper into the valence band and remains near -2.0 eV over a moderately large range of photon energy. Figure 7 suggests that $P3$ results from transitions from the band-3 minimum along the Σ direction. There is a large density of states near this minimum²⁰ since the band is relatively flat over a large portion of the zone extending from the Σ axis out toward the W point. The $P3$ peak due to transitions from the band 3 minimum has been observed in photoemission studies of GaAs, GaP, and Si,¹² and CdTe.^{2,3,21} In ZnTe we find that this minimum lies -2.0 eV below Γ_{8v} . The pseudopotential⁸ value is -2.3 whereas Herman's¹ value is slightly higher. Since the photoemission peak is several tenths of an electron volt wide, the agreement between experiment and theory is really quite good. Furthermore, the energy from which the strongest transitions take place may or may not exactly equal the energy of the Σ minimum depending upon the detailed shapes of the bands away from this point.

The photoemission peak $P4$ first appears at 7.8 eV for $\hbar\omega=10$ eV. We see in Fig. 6 that as the photon energy increases, the initial states responsible for $P4$ gradually move deeper into the valence band. Figure 7 shows that these transitions originate in band 2, and that the energy of the initial states fall rapidly because of the large slope of this band. These transitions occur in the vicinity of the Σ and Δ directions; the Δ direction

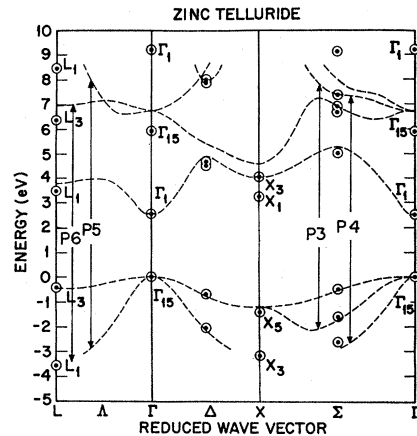


Fig. 7. Theoretical band structure for ZnTe. Herman's (Ref. 7) OPW result (points) is compared to Cohen and Bergstresser's (Ref. 8) pseudopotential result (dashed line). We indicate our suggestions for the transitions responsible for several photoemission peaks. In the text, valence and conduction band levels are often distinguished by v and c subscripts which do not appear in the above drawing. The bands are numbered in order of increasing energy. Shown above are the valence bands 2, 3, and 4, and the conduction bands beginning with band 5.

is excluded since we will soon show that the Δ transitions between bands 2 and 7 lead to a distinct peak $P5$. In our study of CdTe²⁻⁴ we were able to follow $P4$ almost to a photon energy such that the initial states lay at the X point. Hence, we were able to locate X_{3v} and X_{3c} in that material. In ZnTe, however, the LiF window sealing our vacuum chamber (see Sec. II) cuts off long before $P4$ approaches the X point. The best we can do therefore is place an upper bound on X_{3v} of -3.0 eV and a lower bound on X_{3c} of 8.6 eV.

Because of the large slope of band 2 the initial states for the peak $P5$ also fall rapidly as $\hbar\omega$ increases. We showed in Sec. III that $P5$ abruptly disappears from the NED for $\hbar\omega > 11.4$ eV. At this photon energy $P5$ lies at 8.0 eV and is due to initial states at -3.4 eV. We suggest that $P5$ is due to transitions between bands 2 and 7 along the Δ direction and that $P5$ disappears when it reaches the L point because L_{1c} is very nearly a relative maximum and L_{1v} a relative minimum. We therefore locate the upper L_{1c} at 8.0 eV and L_{1v} at -3.4 eV. We find that these values are in fair agreement with the OPW calculation (Table I), but pseudopotential results for these states have not been published.

The peak $P6$ abruptly disappears from the NED for $\hbar\omega > 10.9$ eV. For this photon energy, $P6$ lies at 7.2 eV and is due to initial states near -3.7 eV. Figure 7 suggests that $P6$ is most likely due to $L_{1v} \rightarrow L_{3c}$ transitions. However, this identification is tentative since it is inconsistent with some other results. In the first place, $P6$ locates L_{1v} at -3.7 eV whereas we previously located L_{1v} at -3.4 eV from our study of $P5$. It is conceivable, however, that the difference of 0.3 eV is due to a different lineshape for the $L_{1v} \rightarrow L_{1c}$ and the $L_{1v} \rightarrow L_{3c}$ transitions. Also the location of a photo-

²⁰ See, for example, Fig. 25 of Ref. 7.

²¹ In CdTe (Ref. 2), however, it was shown that most of the electrons in the peak $P3$ resulted from nondirect transitions from a peak in the valence band density of states at -1.9 eV. This high density of states most likely results from the Σ minimum in band 3.

TABLE I. Comparison of photoemission results and theoretical energy levels for ZnTe.

Band structure feature located	Photoemission data		Band calculations ^b	
	Label of structure	Experimental ^a energy of initial or final state	OPW ^c	Pseudo-potential ^d
Γ_{1c}	S2	8.9	8.9	...
Γ_{12c}	S3	9.6	10.2	...
Band-3 minimum (Σ)	P3	-2.0	-2.2	-2.3
X_{3c}	P4	>8.6 ^e
X_{3v}	P4	<-3.0 ^e	-3.5	...
L_{1c}	P5	8.0	8.2	...
L_{1v}	P5	-3.4	-3.9	...
L_{3c}	P6	7.2	6.1	6.7
L_{1v}	P6	-3.7	-3.9	...

^a Energy is measured in electron volts above the valence-band maximum in the crystal. The estimated experimental uncertainty is ± 0.2 eV.

^b Since the valence band maximum is spin-orbit-split by 0.9 eV, the top of the valence band lies 0.3 eV above the location of Γ_{12v} in the absence of spin-orbit splitting. Hence, we have subtracted 0.3 eV from all theoretical levels before entering them into Table I.

^c Reference 7.

^d Reference 8. The published pseudopotential band structure was limited to conduction band states below 8.2 eV and valence band states above -3.3 eV.

^e As discussed in the text, this is only an upper bound on X_{3v} and a lower bound on X_{3c} .

emission peak is only accurate to within a few tenths of an electron volt. A more serious problem is the experimental value for L_{3c} which we find at 7.2 eV. The pseudopotential and OPW values are 6.7 and 6.1 eV, respectively. These lie 0.5 and 1.1 eV below the experimental value. It is surprising to find this large disagreement at L_{3c} since experiment and theory agree within a few tenths of an electron volt for most of the other states discussed earlier. Nonetheless, we are unable to find an alternate identification for $P6$ any-

where in the band structure in Fig. 7. It is possible that the transition occurs in regions of the zone not shown in Fig. 7.

There is one piece of evidence which tends to support our identification for $P6$ in ZnTe. Although $P6$ was never identified in CdTe, further inspection of the data²² shows that there is a peak which appears only for photon energies near 9.7 eV. For $\hbar\omega=9.7$ eV, this peak is located at 6.4 eV and is due to initial states at -3.3 eV. If this peak is identified as $P6$ then we find L_{1v} at -3.3 eV and L_{3c} at 6.4 eV in CdTe. This value for L_{1v} is in excellent agreement with our value of -3.3 eV deduced from $P5$ in CdTe²⁻⁴ and also agrees with the OPW calculation¹ (-3.3) and the pseudopotential calculation⁸ (-3.2). Our value of 6.4 eV for L_{3c} agrees favorably with the OPW value of 6.1 eV and the pseudopotential value of 6.3 eV. Hence, the experimental values deduced from $P6$ in CdTe are in excellent agreement with theory, whereas the values deduced from $P6$ in ZnTe are not.

In Table I we summarize the features of the electronic structure of ZnTe deduced from our photoemission data and compare these results with the OPW and pseudopotential band calculations.

ACKNOWLEDGMENTS

One of us (J.L.S.) would like to express his appreciation to the National Aeronautics and Space Administration for supporting the postdoctoral under which this work was performed. We are also grateful to the Bell Telephone Laboratories where the final version of this manuscript was prepared.

²² Reference 2, Fig. 14.