

Electronic structure of a hole-attractive neutral Cu-related complex-defect bound exciton at 2.345 eV in ZnTe

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A detailed magneto-optical study of a Cu-related bound exciton (BE) at 2.345 eV (2K) in ZnTe is presented, for magnetic fields up to 10 T. The corresponding complex defect is found to be of trigonal symmetry, and also neutral, from the absence of thermalization in Zeeman transmission spectra of the BE line. It has been possible to fit the Zeeman data for the splitting of the BE line with a perturbation Hamiltonian considering both the electron-hole exchange interaction and the local strain field at the defect. It appears that the defect has a hole-attractive potential of a moderate strength, so that the angular momentum for the bound hole is not quenched. The local strain field has a compressive sign, which leaves a $|J, M_J\rangle = |\frac{3}{2}, \pm\frac{1}{2}\rangle$ hole state at lowest energy. The g factor of this hole state is evaluated as $K = 1.20 \pm 0.01$, $L = -0.31 \pm 0.01$, to our knowledge the first direct experimental data for such a $|\frac{3}{2}, \pm\frac{1}{2}\rangle$ hole state in a semiconductor. The electron of the BE is loosely bound, with a g value of $g_e = -0.40$ typical for shallow donors in ZnTe. The defect is tentatively identified as a trigonal $\text{Cu}_{\text{Zn}}\text{-Cu}_i$ pair.

I. INTRODUCTION

The electronic structure of complex defects in semiconductors is still not adequately understood, often due to a lack of sufficiently detailed experimental data, or inadequate evaluation of such data.¹ Some progress has recently been made in the study of neutral complex defects in both indirect²⁻⁷ and direct compound semiconductors.⁸⁻¹¹ A general model has recently been proposed to explain the variations in electronic structure of electronic excitations (bound excitons) at neutral complex defects in all semiconductors.¹² In particular, the magnetic properties of such defects in the direct-band-gap semiconductors ZnTe and CdTe are interesting in this respect, due to the large valence-band spin-orbit splitting in these materials. To get more experimental background for a comparison with theoretical developments of the electronic structure, we have undertaken a detailed magneto-optical study of a number of complex neutral defects in ZnTe.

In this paper we present a detailed account of one particular Cu-related defect with a bound exciton at 2.345 eV (2 K). Cu diffusion in ZnTe gives a large number of complex defects, each represented by a specific bound exciton in optical spectra, as reviewed separately.¹³ The defect associated with the 2.345-eV BE line, here called Z_1^0 , is one of the most commonly occurring complexes with Cu diffusion.¹³ Specific aspects of optical spectra for this defect, particularly in cases of very high doping levels, have been published separately.^{14,15} In this paper we concentrate on the electronic structure of the Z_1^0 BE at 2.345 eV. This is particularly interesting since it appears to be one of the rare cases where an approximately effective-mass-like description for the primary bound hole state in the local hole-attractive potential appears adequate. Also, it is the first case where the magnetic properties of a $|J, M_J\rangle = |\frac{3}{2}, \pm\frac{1}{2}\rangle$ hole state is demonstrated, since the

defect has a local axial strain field of a compressive sign. These data should therefore be very important for the comparison with theoretical calculations of g factors for bound holes in ZnTe.¹⁶ Also, in the general description of bound hole states at complex defects in semiconductors, detailed data for substitutional-interstitial pairs¹⁴ in II-VI compound materials have been missing.

This paper is organized in the following way. A brief description of samples and experimental conditions is given in Sec. II. Section III describes the experimental magneto-optical data, together with a detailed discussion of these data in terms of theoretically computed spectra for a proper spin Hamiltonian for the bound exciton in the magnetic field. Section IV contains a discussion of the electronic structure deduced from the magneto-optical data, consistent with the assumption of a $\text{Cu}_{\text{Zn}}\text{-Cu}_i$ trigonal pair as the defect identity. Section V finally briefly summarizes the most important conclusions from this work.

II. EXPERIMENTAL

The samples used in this work were bulk single crystalline originally undoped ZnTe, which were Cu doped by a single step diffusion procedure, typically at temperatures 300–500 °C for one hour. A detailed description of the sample preparation can be found in a previous paper.¹³

Photoluminescence and optical transmission spectra were obtained at 2 K with Ar^+ -laser excitation and a Spex 1404 double monochromator on the detection side. Photoluminescence excitation spectra were performed with the aid of a Coherent 590 cw tunable dye laser. Magneto-optical data reported here were obtained at the Max-Planck Hochfeld Magnetlabor in Grenoble, using a split-coil superconducting magnet at fields up to 10 T, together with a Jobin Yvon THR 1500 1.5-m high-resolution monochromator.

III. EXPERIMENTAL DATA

The defect discussed in this work is characterized via the properties of the bound exciton (electron-hole pair) which is localized to the defect at low temperatures. [Optical spectroscopy of bound excitons (BE's) is demonstrated to be a quite powerful technique to explore the electronic properties of complex defects in semiconductors,¹¹ particularly for neutral such defects which often appear in low concentrations and are difficult to study in electrical measurements.] The photoluminescence (PL) spectrum of this BE is shown in Fig. 1(a). A single electronic line appears at ≈ 2.3452 eV at 2 K, accompanied by a specific phonon wing within the one-phonon range towards lower energies (Fig. 1). Details of the phonon coupling for this

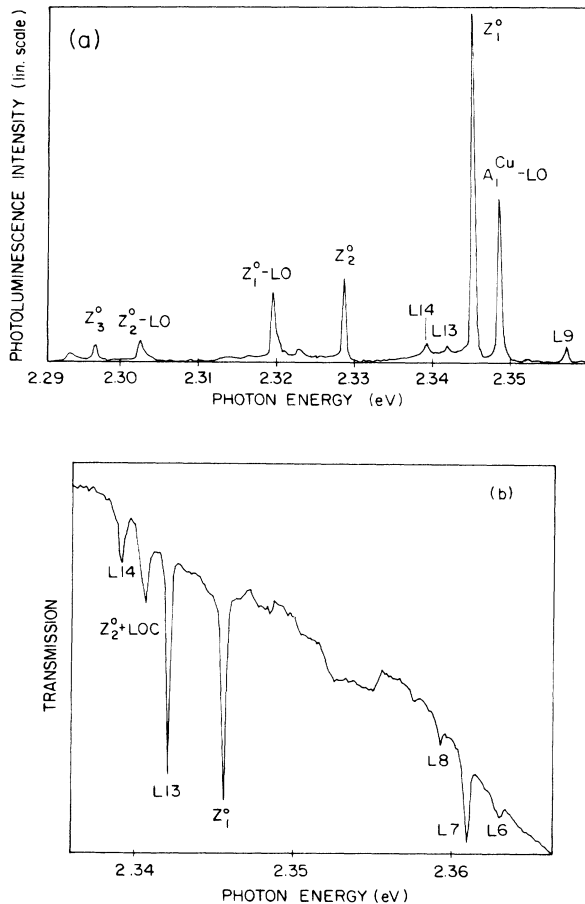


FIG. 1. (a) Photoluminescence spectrum of the 2.345-eV bound exciton in ZnTe, taken at 2 K with Ar^+ -laser excitation (5145 Å). A single electronic line Z_1^0 is seen at 2.345 eV, together with a weak one-phonon band at lower photon energies, terminated with the one-LO Γ -phonon replica close to 2.319 eV. Lines denoted L 9, A_1^{Cu} , L 13, L 14, Z_2^0 , and Z_3^0 do not belong to the Z_1^0 bound-exciton spectrum; they are electronic lines associated with other Cu-related complex defects. (b) Transmission spectrum of the Z_1^0 bound exciton at 2 K at zero field, showing no excited states within the band gap. Lines denoted by L 6–L 8, L 13, $Z_2^0 + \text{LOC}$, and L 14 belong to other Cu-related centers (Ref. 13).

defect have been discussed elsewhere.^{13,15} Also, in transmission only one electronic line is seen, as evident from Fig. 1(b). The phonon coupling is stronger in absorption,¹⁵ but obviously no excited electronic state for this BE is observed below the band gap. This is an important observation that will be discussed below in connection with the detailed model for the electronic structure of the defect.

BE spectra for complex defects in ZnTe may look quite similar at zero magnetic field for neutral complex defects and for charged complexes, i.e., donors or acceptors.¹³ It is therefore necessary to arrive at a conclusion on the class of defect involved, before any detailed magneto-optical data can be interpreted. This can be successfully done in transmission data in a magnetic field.¹³ As shown in Fig. 2, the Zeeman splitting of this BE line in absorption at 10 T is symmetric, and shows no sign of thermalization. This is clear evidence that the ground state of the BE has no electronic particle, since otherwise a splitting of this ground state would occur in the magnetic field. Such a splitting would lead to thermalization of the spectrum in absorption,¹³ usually already at quite low magnetic fields. Therefore it is concluded that the 2.3452-eV BE derives from a neutral defect, with no spin in the ground state. The BE consequently contains just two particles, one electron and one hole. The more complicated case of donors or acceptors can be ruled out from the data in Fig. 2.

A detailed analysis of the splitting pattern of the 2.3452-eV BE line in a magnetic field has been undertaken. Figure 3 shows a typical splitting pattern with magnetic field, taken in an arbitrary direction. Also shown is an example of a Zeeman-split spectrum at 10 T. Due to a rather high total doping level of the samples (close to 10^{17} cm^{-3}) the natural linewidth of the BE lines is of the order 0.1 meV, which means that all components of the spectrum cannot always be resolved spectrally at 10 T. From the line shape of the different spectral lines it is possible

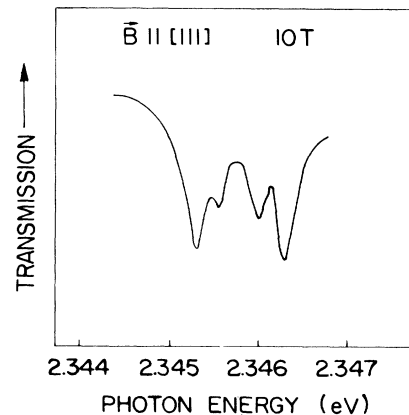


FIG. 2. Optical transmission spectrum of the Z_1^0 bound exciton line at 2.345 eV at a magnetic field of 10 T, with $\mathbf{B} \parallel [111]$. No thermalization occurs in this spectrum, which proves that no particle is bound to the defect in the ground state, i.e., the Z_1^0 defect is neutral ("isoelectronic").

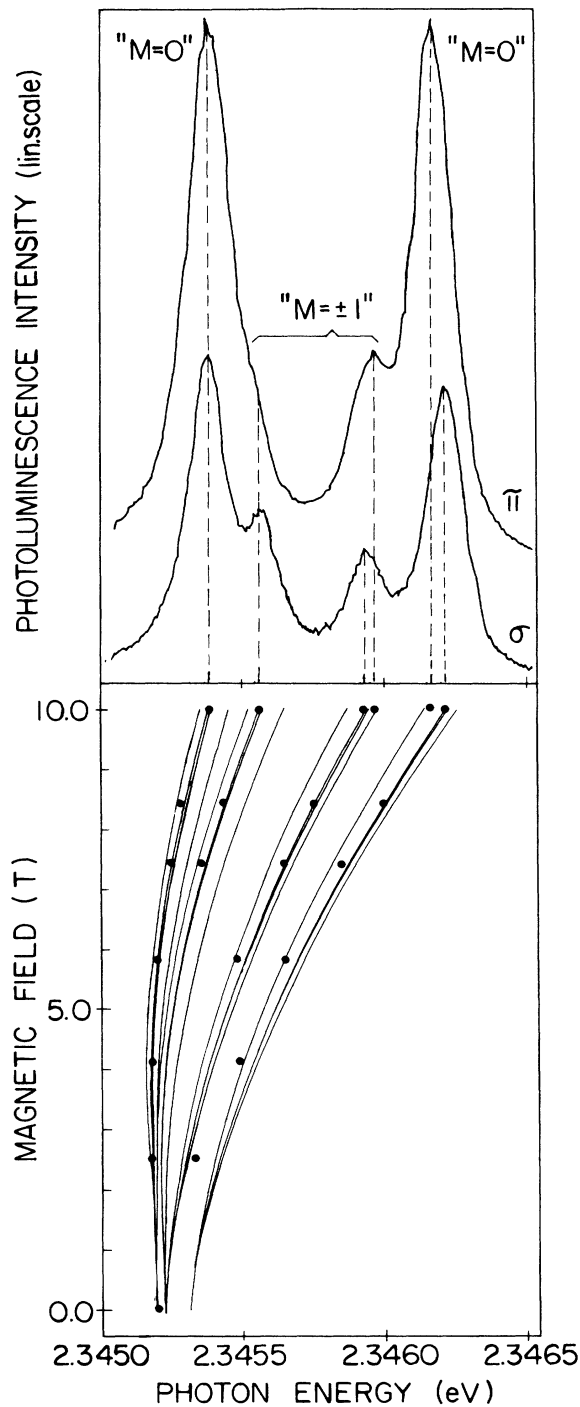


FIG. 3. Fan diagram for the splitting of the Z_1^0 bound-exciton line with magnetic field in the Voigt configuration at 2 K in a low-symmetry direction (\mathbf{B} at an angle of $\approx 17^\circ$ with $[001]$ in the $(1\bar{1}0)$ plane, with a misorientation of about 4°). The solid lines are computer simulated from the spin Hamiltonian given in the text, considering the 4° misorientation. A full spectrum of the Zeeman-split Z_1^0 line at 10 T is also shown, for the same sample orientation. The lines are labeled according to the right-hand side of Fig. 5.

to deduce the existence of weak spectral components, however, for comparison with a theoretical model. From such a careful analysis it has been concluded that at zero field there is actually a small residual splitting of the BE line, due to electron-hole ($e-h$) exchange interaction. The high-energy line is too weak to be separately resolved at 0 T, however, but can be seen separately at low fields as a shoulder. One important conclusion from Fig. 3 comes from the quadratic shift of all Zeeman-split lines with magnetic field, as will be commented on below.

The angular dependence of the Zeeman-split spectrum is the most important part of the experimental data presented in this work. It is shown in Fig. 4, for a rotation with \mathbf{B} in the $(1\bar{1}0)$ plane. The data in Fig. 4 are fitted by a theoretically calculated pattern (solid lines in Figs. 3 and 4), obtained from a perturbation Hamiltonian for the 2.3452-eV BE. The following Hamiltonian has been used:

$$H' = H_{\text{ex}} + H_{\text{LCF}} + H_{\text{LZ}} + H_{\text{QZ}}, \quad (1)$$

where

$$H_{\text{ex}} = -a\mathbf{J}\cdot\mathbf{S} - b(J_x^3S_x + J_y^3S_y + J_z^3S_z) \quad (2)$$

denotes the exchange interaction between the bound electron-hole pair, while

$$H_{\text{LCF}} = -D[J_\zeta^2 - \frac{1}{3}J(J+1)] \quad (3)$$

is the main local crystal-field term, acting on the bound hole states via the low-symmetry strain field.¹⁷ The linear Zeeman interacting term is written in the conventional way¹⁸

$$H_{\text{LZ}} = \mu_B [g_e\mathbf{S}\cdot\mathbf{H} + K\mathbf{J}\cdot\mathbf{H} + L(J_x^3H_x + J_y^3H_y + J_z^3H_z)], \quad (4)$$

while the quadratic Zeeman term H_{QZ} is taken as

$$H_{\text{QZ}} = C_1H^2 + C_2(\mathbf{J}\cdot\mathbf{H})^2 + C_3[\frac{1}{2}H_xH_y(J_xJ_y + J_yJ_x) + \text{c.p.}] \quad (5)$$

Here \mathbf{J} is the total angular momentum (including spin and orbital angular momentum) of the bound hole state, while \mathbf{S} is the corresponding quantity of the bound electron. μ_B is the Bohr magneton, x, y, z refer to the usual cubic axes of the lattice, while ζ is the defect axis. Cyclic permutations are denoted c.p. This Hamiltonian for an exciton bound to a neutral complex includes the major effects of both $e-h$ exchange and local strain field in addition to the Zeeman effect.

The above Hamiltonian has been used to calculate a theoretical family of spectral lines to be fitted to the data in Figs. 3 and 4. The theoretical lines have been computed by diagonalization of all the terms in Eq. (1) simultaneously. A basis set of spinlike electrons and p -like holes (derived from Γ_8 holes in T_d symmetry) was used in this procedure. It was evident, however, that a good fit could only be obtained if the D factor is assumed negative and quite large, i.e., the local crystal field is the dominating perturbation of the bound hole states and in addition of a compressive nature. The value of D is estimated to be large, so that the excited states of the hole is out of the

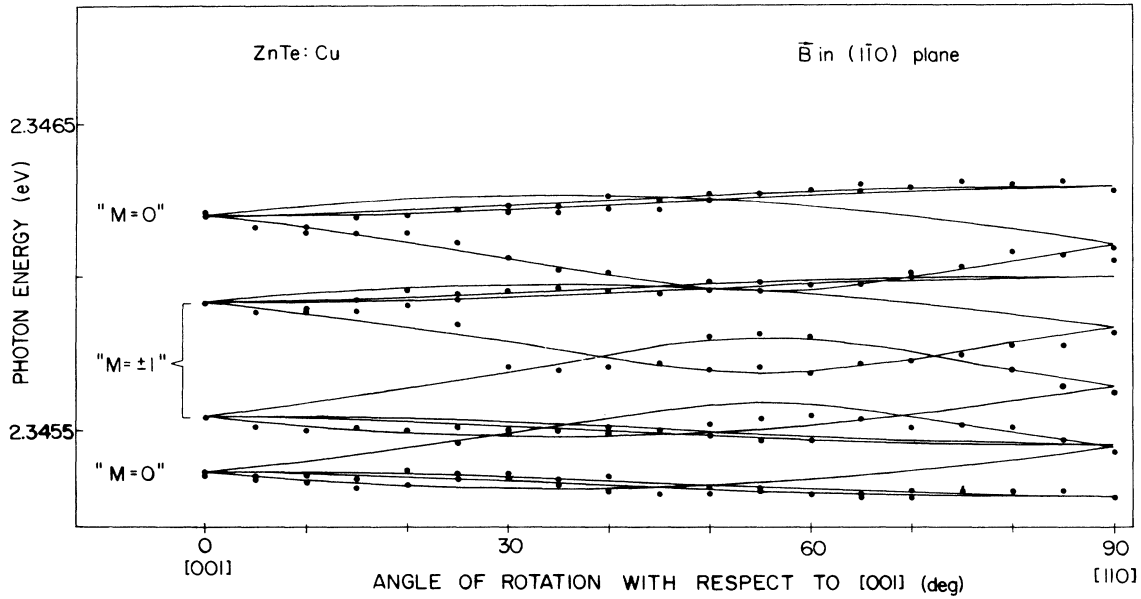


FIG. 4. Angular dependence of the Zeeman-split magnetic subcomponents of the Z_1^0 bound-exciton line in the Voigt configuration at 2 K, for the four sets of $\langle 111 \rangle$ -oriented defects. The sample was rotated with the magnetic field \mathbf{B} in a $(1\bar{1}0)$ plane, with a 4° misorientation. Solid lines are computer simulated from the spin Hamiltonian given in the text, considering the 4° misorientation. The labels are according to Fig. 5.

band gap, consistent with the experimental observation that only one (slightly broadened) electronic BE line associated with the Z_1 defect is seen below the band-gap energy at zero magnetic field. The exact value of D is not critical for the evaluation of additional parameters in Eqs. (1)–(5), as long as it is much higher than the e - h exchange interaction.

The other parameters deduced from the best fit to the data presented in Figs. 3 and 4 are as follows:

$$a = 0.060 \pm 0.005 \text{ meV},$$

$$g_e = -0.40 \pm 0.02,$$

$$K = 1.20 \pm 0.01,$$

$$L = -0.31 \pm 0.01,$$

$$C_1 = (5.5 \pm 0.2) \times 10^{-3} \text{ meV/T}^2,$$

while the parameters b , C_2 , and C_3 are close to zero, so that specific (very small) values for these constants cannot be evaluated given the present accuracy of experimental data.

The following important conclusions can immediately be drawn from the evaluation of the above parameters. From the general properties of the angular dependence in Fig. 4 it follows that the defect has trigonal symmetry, i.e., a $[111]$ -oriented axis. All four magnetic subcomponents of the BE show an orientational degeneracy with $\mathbf{B} \parallel [001]$, and a splitting pattern characteristic of a $\langle 111 \rangle$ -oriented defect (Fig. 4). Further, the evaluated negative sign of D means that the local strain field is compressive,¹⁹ which is expected for an identity of the defect as $\text{Cu}_{\text{Zn}}\text{-Cu}_i$, as discussed in more detail below in Sec. IV. While the local strain field is the dominating pertur-

bation, it leaves one specific hole state at lowest energy. Obviously the hole angular momentum is not quenched, since in that case the hole g factor (and also the total g factor for the BE) would be isotropic.^{2–7} In an effective-mass-like model the hole states in a compressive trigonal local strain field would be the $M_J = \pm \frac{1}{2}$ hole states of the valence-band top. The g factors for such hole states have not previously been studied in ZnTe, only the values for the total Γ_8 fourfold-degenerate shallow hole states,²⁰ as well as the $M_J = \pm \frac{3}{2}$ hole states in a tensional local strain field, have been previously reported.^{11,21} The K and L values reported here are therefore novel experimental information for hole states in ZnTe. It is noted that the K and L values derived here are considerably larger than typical values for $M_J = \pm \frac{3}{2}$ holes.^{11,21} A more-detailed discussion on the hole g factor in this case will be given below in Sec. IV.

The bound electron of the BE is deduced to be rather loosely bound in an effective-mass-like state, since the g factor g_e is -0.40 , exactly the same as for conduction-band electrons or shallow donors in ZnTe.²² This is also in agreement with the quadratic Zeeman shift C_1 , although this is reduced below the values of the shallow donors [these have C_1 values about $C_1 \approx 8.7 \times 10^{-3} \text{ meV/T}^2$ (Ref. 23)]. This is common also for other hole-attractive complex defect BE's in ZnTe.^{11,24} For a defect which is strongly electron attractive, on the other hand, C_1 is close to zero, and the g factor becomes close to $g_e = +2$.^{8,23} We consequently conclude that the Z_1 complex defect is basically hole attractive, and the electron is loosely bound as a secondary particle in the Coulomb field of the hole primarily bound to the defect potential. This is an example of the classical Hopfield-Thomas-Lynch

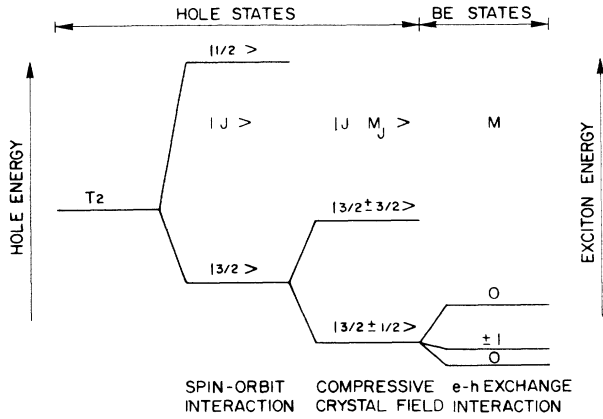


FIG. 5. Schematic picture of the electronic structure for the Z_1^0 bound exciton at 2.345 eV. The spin-orbit interaction is assumed to be the dominating perturbation on the bound hole states, but the effect of the local strain field is also large in this hole-attractive defect potential. The electron-hole exchange interaction, on the other hand, is rather small, and not well resolved at zero magnetic field.

(HTL) model for the electronic structure of neutral (“isoelectronic”) defects.²⁵

A loosely bound electron is also consistent with the very small value $a = 0.06$ meV found for the e - h exchange interaction for the Z_1^0 BE. As soon as the electron is more strongly localized, this factor immediately grows larger, to be typically of the order a few meV in ZnTe.²³

The above discussion on the electronic structure of the Z_1^0 BE is schematically summarized in Fig. 5. The small e - h exchange interaction will split the Z_1^0 BE line at zero field. These details cannot be sufficiently resolved spectroscopically, since the linewidth is of the same order as the separation of the lines.

IV. DISCUSSION

A. Electronic structure of the Z_1^0 BE

The perturbation order given in Fig. 5 already describes the main electronic properties of the Z_1 defect. As discussed above, the center has a dominantly hole-attractive neutral potential, which is plausible if Cu_{Zn} is part of the defect. The pseudopotential of Cu_{Zn} is not very strong [Cu_{Zn} is an acceptor with a binding energy of ≈ 150 meV in ZnTe (Ref. 22)], but the compressive strain field also contributes to the potential. The electron, on the other hand, can consistently be described as a loosely bound secondary particle, with properties similar to a shallow donor electron.

An interesting problem to be discussed is the general validity of the description of bound hole states presented above, which is virtually identical to effective-mass-like hole states in a compressive trigonal field. Yet it is clear that a hole bound in a very localized neutral potential should, in general, not at all be described as an effective-mass-like state. The fact that the BE energy is close to the band gap of the material is not important in this case,

since the valence-band edge is not a relevant reference continuum for bound hole states in a very localized neutral potential.

As described in more detail separately,¹² the condition for quenching of hole angular momentum for a hole bound in a hole-attractive potential depends critically on the strength of the spin-orbit interaction experienced by the bound hole. This perturbation is, in fact, described as the main perturbation in Fig. 5, stronger than the local strain field. This is reasonable in view of the large spin-orbit splitting of the valence-band top in ZnTe, about 1 eV. This strong influence of the spin-orbit interaction has profound effects on the magnetic properties of bound hole states. In the absence of such strong spin-orbit effects the hole angular momentum would, in general, be expected to be quenched, since a strongly hole attractive potential of a symmetry lower than tetrahedral would, in general, leave the bound hole state nondegenerate. A nondegenerate hole state has a quenched angular momentum,¹² as has been known to be true for localized electronic states in ESR.²⁶

With the perturbation scheme in Fig. 5 a description of the bound hole states is most naturally done with the spin-orbit Hamiltonian term $H_{\text{s.o.}}$ included in the unperturbed Hamiltonian, while the low-symmetry crystal field is treated as the main perturbation. In this case the perturbed bound hole states are twofold degenerate according to Kramer’s theorem.¹² Since these hole states are orbitally degenerate, there is no reason why quenching of orbital angular momentum should occur. Also, our experimental data for the Z_1 defect presented in this work confirm that the angular momentum is not quenched. Whether the K and L values obtained here would be different in the case of an effective-mass-like acceptor in a similar compressive trigonal field cannot be concluded from this work, since no example of the latter case seems to have been published as yet.

With the above assumption about the dominating strength of the spin-orbit interaction for the bound hole state, and the conclusion that the bound hole state would be twofold degenerate in this case even in a low symmetry, the description of these bound hole states formally becomes similar to the effective-mass picture.¹⁹ Therefore the formalism used above with a $|J, M_J\rangle = |\frac{3}{2}, \pm\frac{1}{2}\rangle$ twofold degenerate hole state at lowest energy is valid as long as the spin-orbit interaction dominates. (For the present problem with the Z_1^0 BE in ZnTe, the Cu_{Zn} pseudopotential is rather weak, and the deviation from an effective-mass-like picture might not be expected to be very large.) In fact, a quite similar scheme has been useful in the analysis of the 2.261-eV BE in Cu-doped ZnTe, although in this case the local field is tensional so that the $|J, M_J\rangle = |\frac{3}{2}, \pm\frac{3}{2}\rangle$ twofold degenerate hole state is at lowest energy.¹¹

B. Defect identity

The Z_1 defect is one of the most common defects observed (via the 2.3452-eV BE) in Cu-diffused ZnTe, and should therefore be one of the simplest complexes formed by such diffusion. It is known that Cu diffuses very rap-

idly by an interstitial mechanism in ZnTe,^{13,21} and it is natural to assume that interstitial Cu (Cu_i) may be frozen in as paired with another defect during cool down after the diffusion. At the diffusion temperature it may be assumed that Cu_i acts as a positively charged shallow donor, while Cu_{Zn} would act as a negatively charged acceptor. Therefore a mechanism driving the formation of $\text{Cu}_{\text{Zn}}\text{-Cu}_i$ complexes is simple Coulomb attraction at the diffusion temperature. A tetrahedral interstitial position for Cu_i is the most natural one, leading to the observed trigonal axial symmetry for the Z_1 complex if it is identified as $\text{Cu}_{\text{Zn}}\text{-Cu}_i$. Certainly a compressive local strain field is expected, since two atoms of similar size as the host Zn atom are to be accommodated locally within one primitive cell of the material. We conclude that a tentative identification of the Z_1 complex defect in ZnTe as $\text{Cu}_{\text{Zn}}\text{-Cu}_i$ is in agreement with the physical properties observed in this work.

V. SUMMARY AND CONCLUSIONS

This work is of a significant general importance in two aspects. (1) The electronic structure of a hole-attractive complex neutral defect in a compressive axial strain field is studied in detail, and the primary bound hole is found to be adequately described in a rather simple way as a $|J, M_J\rangle = |\frac{3}{2}, \pm\frac{1}{2}\rangle$ Kramer's doublet. (2) The defect studied is believed to be a typical example of a specific class of simple complex defects involving a substitutional-interstitial pair in axial symmetry, ($\text{Cu}_{\text{Zn}}\text{-Cu}_i$), expected to create a compressive local strain field at the defect.

The detailed analysis of Zeeman data for the 2.345-eV BE was done with a sufficiently complete spin Hamiltonian, by taking account of both the low-symmetry local strain field, the electron-hole exchange interaction, and the Zeeman splitting caused by the external magnetic field. The corresponding terms were diagonalized simultaneously in calculating expected line positions in the Zeeman spectrum, for various angles of the magnetic field versus sample orientation. The satisfactory fit obtained in

this work supports the general conclusion that the bound hole may be described as an orbitally degenerate $|\frac{3}{2}, \pm\frac{1}{2}\rangle$ state. This is consistent with the assumption for the impurity potentials of the constituents of the defect (Cu_{Zn} and Cu_i). The derived value of the g factor for the bound hole is to our knowledge the first direct experimental data for a $|\frac{3}{2}, \pm\frac{1}{2}\rangle$ hole state in ZnTe, and are found to deviate considerably from the previously established data for $|\frac{3}{2}, \pm\frac{3}{2}\rangle$ hole states in ZnTe.

ZnTe has a large spin-orbit splitting of the valence band, which means that the magnetic properties of a bound hole remain essentially similar to those for shallow acceptor bound holes even for a quite localized defect potential (as appropriate for the neutral complex studied in this work), if this potential is not too strong. It is expected that in ZnTe either a quite strong defect potential or a very strong local strain field is necessary to significantly quench the orbital angular momentum of a bound hole state, an effect that is common for GaP and Si, where the valence-band spin-orbit splitting is much smaller. The case studied here fits well into the weak potential picture; other cases of complex defects in ZnTe which deviate from this behavior will be discussed separately.

The proposed identification of the 2.345-eV defect as a $\text{Cu}_{\text{Zn}}\text{-Cu}_i$ pair is still to be regarded as tentative, although it is certainly consistent with available experimental data. Improvements in the techniques for identification of such complex defects are therefore of high priority in future work on the understanding of such defects, and of defect reactions in general.

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¹For a recent detailed review of electronic properties of bound excitons in semiconductors, see P. J. Dean and D. C. Herbert, in *Excitons*, Vol. 14 of *Topics in Current Physics*, edited by K. Cho (Springer-Verlag, Berlin, 1979), p. 55.

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