Effects of Uniaxial Strain on Band-Edge Optical Transitions in a Direct Zinc-Blende-Structure Semiconductor

P. T. Bailey
Monsanto Company, Saint Louis, Missouri 63166

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The electric-dipole selection rules have been obtained for the lines arising from weakly bound two- or three-carrier complexes in a uniaxially strained direct zinc-blende semiconductor. The optical transitions have been studied in the group of the k vector for a crystal strained along a high-symmetry axis, and certain effects of strain are found to change with the axis considered. The effects of exchange splittings and, in the three-carrier cases, of the Pauli exclusion principle, have been included. The relative strengths of each line in the polarizations parallel and perpendicular to the strain have been determined. Each line which becomes allowed only in the presence of strain has been called "weak," since its strength must decrease continuously to zero with decreasing strain. For each complex, the ordering of the lines has been determined. The predicted number of lines, their ordering, and their strengths differ with the type of complex. The predictions agree with experiment in the case of two-carrier complexes, and appear to agree with the limited data available for three-carrier complexes. There is thus a new means to identify the types of complexes involved in band-edge optical transitions in a direct zinc-blende-structure semiconductor. Further, because each line for a given complex can be identified from its relative strengths in the two polarizations, the exchange splittings in the strained material can also be determined from experiment. The approach may be extended to structures different from zinc blende, to a change in symmetry arising from an external field rather than a strain, and possibly to an indirect semiconductor.

I. INTRODUCTION

THE effects of uniaxial strain on band-edge optical transitions in a direct zinc-blende semiconductor have previously been investigated by means of the quasicubic model.[1,2] In that approach, the splitting of the free-carrier line when the symmetry is lowered from cubic is considered in terms of the splitting of a free-atom $s_{1/2} \rightarrow p_{3/2}$ transition when the symmetry is lowered from spherical to ellipsoidal. The relative strengths in π and σ polarizations (respectively, parallel and perpendicular to the strain) which the quasicubic model predicts were shown to be obeyed at least qualitatively by the $n=1$ free-exciton lines in uniaxially strained CdTe.[2] In our work the optical transitions are studied in the group of the k vector for a crystal strained along a high-symmetry axis, and certain effects of the strain are found to change with the axis considered. Further, we consider the effects of strain on transitions in complexes containing either two or three bound carriers, and we include the effects of exchange splittings.

This work was motivated by studies of the low-temperature band-edge photoluminescence in high-purity, unstrained GaAs.[3–5] The photoluminescence arises largely from the recombination of excitons (X) which are bound to isolated single donors (D) or acceptors (A). These impurity-exciton complexes exist both for charged impurities (D^+ or A^-) and for neutral impurities (D^0 or A^0), corresponding to two or three weakly bound carriers, respectively. Free-carrier and free-exciton recombinations have also been observed in photoluminescence[3] and may be regarded as limiting cases of recombination in the two-carrier complex D^+-X (or A^--X). The three-carrier complexes (two electrons and one hole, or vice versa) are of interest because the eigenstates of the two similarly charged particles are limited to those allowed by the Pauli exclusion principle.

The electric-dipole selection rules have been obtained for the lines arising from these complexes in a direct zinc-blende semiconductor which is strained along a high-symmetry axis. The effects of exchange splittings and of the Pauli principle have been included. The relative strengths of each line in π and σ polarizations have been determined. Each line which becomes allowed only in the presence of strain has been called "weak," since its strength must decrease continuously to zero with decreasing strain. For each complex the ordering of the lines has been determined. The predicted number of lines, their ordering, and their strengths are different for each of the cases X (or D^+-X, etc.), D^0-X, and A^0-X.

We present our approach and illustrate its application to a specific case by a detailed consideration of a three-carrier complex, whose states are limited by the Pauli principle. A complex A^0-X yields the greatest number of allowed states and lines, and best illustrates the approach. Such a complex is also of interest experimentally, because the strongest and hence best resolved lines observed in unstrained GaAs[3] arise from Zn^0-X, so we expect Zn^0-X uniaxial-strain data to provide the most stringent test of our predictions. For two-carrier complexes and for a D^0-X three-carrier complex, we present results only, since the details of the determinations can be reconstructed along the lines of the illustration given for A^0-X. Results for a [100] and a [111] strain are reported, since each strain splits the valence

[1] J. J. Hopfield, J. Phys. Chem. Solids **15**, 97 (1960).
[2] D. G. Thomas, J. Appl. Phys. **32S**, 2298 (1961).
[3] M. A. Gilleo, P. T. Bailey, and D. E. Hill, Phys. Rev. **174**, 898 (1968).
[4] E. H. Bogardus and H. B. Bebb, Phys. Rev. **176**, 993 (1968).
[5] R. C. C. Leite, J. Shah, R. E. Nahory, and K. L. Lawley, Bull. Am. Phys. Soc. **13**, 1477 (1968).

band into two bands which belong to different double-valued irreducible representations of the new group of the k vector. There are resulting differences between the selection rules for transitions involving holes from one or other band. For a [110] strain the two valence bands belong to the same double-valued irreducible representation of the new group of the k vector (group C_{2v}). The selection rules are therefore the same for transitions involving holes from either band, and much less information is obtained. Results for this case are not reported but can be determined as indicated above. The strains considered are compressive, in order to conform with experiment.

II. APPROACH

The basis of this work is that the irreducible representations for a weakly bound direct-exciton complex are given by the direct product of the irreducible representations for the constituent carriers and for their envelope function.[1,6] Only the lowest-energy bound state of each complex is considered, so that every envelope function belongs to the identity representation Γ_1. The representations to which the individual carriers belong in the strained material follow from compatibility with the known representations for the unstrained case. For a three-carrier complex the antisymmetric (i.e., Pauli-allowed) representations for the two similarly charged particles are determined from the relation[7]

$$\{\chi\times\chi(R)\}=\tfrac{1}{2}\{[\chi(R)]^2-\chi(R^2)\}.$$

It is sufficient to consider throughout only the antisymmetric states of the three-carrier complexes, as there is no mixing of antisymmetric and symmetric states by strain, spin-orbit coupling, or any other mechanism. Each bound-exciton representation corresponds to the initial state of an exciton-recombination transition. The final state belongs to the representation of the remaining bound particle in the case of a neutral impurity, or to Γ_1 (the ground state of the crystal) if the impurity is charged.[1,6] The optical matrix element is nonzero for a given polarization only if the representation for the initial state is contained in the direct product of the representations for the final state and for the electric-dipole operator in that polarization. The relative strengths of an allowed line in π and σ polarizations are determined from the coupling coefficients.

The spin-orbit interactions for the carriers greatly exceed the Coulomb interactions between the carriers, so that we are determining impurity-exciton analogs of atomic states formed in a j-j coupling scheme.[6] The Coulomb interactions produce a small exchange splitting between the j-j coupled states. Thus small splittings are present even in unstrained material, and we have the additional interesting problem of identifying which lines in strained material arise from each exchange-split line in the unstrained case. For each complex we have made these identifications by determining the j-j coupled states in symmetries appropriate to a free atom, to unstrained material and to uniaxially strained material. The compatibility tables between the groups involved then provide the identifications sought, using the free-atom case as a bridge between the other two. The free-atom case is useful also because the ordering of its exchange-split levels is known,[8,9] and the ordering of the exchange-split levels in unstrained material can be deduced from it. By making the identifications described we find which transitions are allowed only in the presence of strain, and we call these lines "weak" since their strengths decrease to zero with decreasing strain.

The orderings in strained material we determine as follows. First, the ordering of the split valence bands is determined from experimental data for two-carrier complexes.[2,10] Second, we use the fact that the splitting of the valence band greatly exceeds the difference between the binding energies of the resultant split exciton states.[2] When the strain splitting is significantly larger than any exchange splitting, e.g., for stresses $\gtrsim 500$ kg cm^{-2} in GaAs, the above two considerations give the gross ordering due to strain. There remain fine orderings due to exchange, but these we have been unable to predict in the strained material.

III. TWO-CARRIER COMPLEXES

Free-Carrier (and Band-to-Impurity) Lines

In unstrained material (group T_d) the conduction- and valence-band states belong to[11] Γ_6 and Γ_8, respectively, and the electric-dipole operator to Γ_5. For a [100] strain (group D_{2d}) the dipole operator belongs to $\Gamma_4(\pi)$ and $\Gamma_5(\sigma)$, and for a [111] strain (group C_{3v}) to $\Gamma_1(\pi)$ and $\Gamma_3(\sigma)$. The ordering of the valence bands (Fig. 1) is determined by observations[2,10] that a two-carrier line T is strain-split into a lower-energy line T_1 of π and σ polarizations and a higher-energy line T_2 of σ polarization. For the free-carrier lines in either [100] or [111] strain we determine the strengths in the polarizations to be $F_1(\pi;\frac{1}{2}\sigma)$, $F_2(\frac{1}{2}\sigma)$. These results are exact. In Sec. I we mentioned the quasicubic model,[1,2] which merely approximates the lowering of the zinc-blende symmetry by the strain; this model gives

[6] D. G. Thomas and J. J. Hopfield, Phys. Rev. **128**, 2135 (1962).

[7] M. Hamermesh, *Group Theory and Its Application to Physical Problems* (Addison-Wesley Publishing Co., Reading, Mass., 1962), p. 134.

[8] Y. Yafet and D. G. Thomas, Phys. Rev. **131**, 2405 (1963).

[9] P. J. Dean, W. F. Flood, and G. Kaminsky, Phys. Rev. **163**, 721 (1967).

[10] For GaAs this ordering appears to have been determined first by F. H. Pollack, M. Cardona, and K. L. Shaklee, Phys. Rev. Letters **16**, 942 (1966).

[11] Notations are those given in G. F. Koster, J. O. Dimmock, R. G. Wheeler, and H. Statz, *Properties of the Thirty-Two Point Groups* (M. I. T. Press, Cambridge, Mass., 1963).

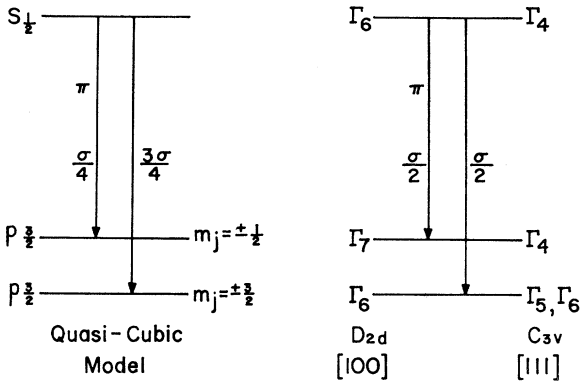

FIG. 1. Free-carrier optical transitions in a uniaxially compressed direct zinc-blende semiconductor, showing representations of the groups D_{2d} and C_{3v} to which the bands belong and the relative strengths, in π and σ polarizations, which pertain in quasicubic symmetry and in [100]- or [111]-strained material. In the group C_{3v}, Γ_5 and Γ_6 are a Kramers pair.

$F_1(\pi; \frac{1}{4}\sigma)$, $F_2(\frac{3}{4}\sigma)$.[12] The sum of the strengths of F_1 and F_2, in [100] or [111] strain and in the quasicubic model, remains equal to the strength of the isotropically polarized line F in unstrained material. The line F_2 may be unobservable because it moves with increasing strain into a region of rising optical absorption. This line is subject also to lifetime broadening,[2,6] as are all the similar higher-energy lines which are discussed in this paper.

Bound-Carrier Lines

A weakly bound two-carrier complex (X, D^+-X, or A^--X) should yield results identical to the free-carrier results when the exchange splitting is of negligible magnitude relative to the strain splitting. In unstrained material (group T_d) there are three exchange-split exciton lines: $\Gamma_5 \rightarrow \Gamma_1 (J=1 \rightarrow J=0$, allowed), and $\Gamma_4 \rightarrow \Gamma_1$ and $\Gamma_3 \rightarrow \Gamma_1 (J=2 \rightarrow J=0$, forbidden, and occurring at lower energy than the $J=1$ line). We call these lines X_a, X_b, and X_c, respectively. A [100] strain splits line X_a into a lower-energy line X_{1a} of π polarization and a higher-energy line X_{2a} of σ polarization (Fig. 2). The line X_b is split into an allowed, and hence "weak," σ line X_{1b} and a forbidden line X_{2b}. The line X_c is split into forbidden lines X_{1c} and X_{2c}. When the strain is large the weak and strong lines are of comparable strengths, and we determine their relative strengths in the polarizations to be $X_{1a}(\pi)$, $X_{2a}(\frac{1}{2}\sigma)$, $X_{1b}(\frac{1}{2}\sigma)$. As the strain decreases the σ line X_{1b} becomes weaker and the σ line X_{2a} correspondingly stronger. The exchange splitting between the Γ_5 and Γ_4 exciton levels can be determined from an extrapolation to zero strain of the position of X_{1b}. A [100] strain experiment involving the free-exciton lines in GaAs has confirmed these predictions.[13] For a [111] strain the strain pattern (number of lines, strain ordering, and relative strengths) is similar to that shown in Fig. 2 except that there are two exchange-split higher-energy lines, and in this case we find relative strengths $X_{1a}(\pi)$, $X_{2a}(\frac{1}{4}\sigma)$, $X_{2b}(\frac{1}{4}\sigma)$, $X_{1c}(\frac{1}{2}\sigma)$. The exchange splitting between the differently polarized lines X_{1a} and X_{1c} has been resolved in [111]-strained ZnSe and ZnS[14]; however, the exchange splitting between the similarly polarized lines X_{2a} and X_{2b} was not resolved. If the exchange splittings are ignored, the difference between the bound- and free-carrier cases is that the bound-carrier lines have strengths in σ polarization which vary with the strain: $X_1(\pi; 0 \rightarrow \frac{1}{2}\sigma)$, $X_2(\sigma \rightarrow \frac{1}{2}\sigma)$, where the arrow indicates the change with increasing strain. This result appears to agree with Thomas's reflectivity data for the exciton lines in CdTe, obtained at stresses from 0 to 1220 kg cm^{-2} (Fig. 5 of Ref. 2[15]).

IV. THREE-CARRIER COMPLEXES

Neutral Acceptor-Exciton Lines

A^0-X Complex in Cubic (Group T_d) Symmetry and in Spherical Symmetry

The representations for the two-hole ($2h$) states are given by (Fig. 3)

$$\Gamma_8(h)\times\Gamma_8(h)=\Gamma_1+\Gamma_3+\Gamma_5(+\Gamma_2+2\Gamma_4+\Gamma_5),$$

where the representations corresponding to states forbidden by the Pauli principle are in the parentheses. The A^0-X representations are

$$\Gamma_8(h)\times\Gamma_8(h)\times\Gamma_6(e)$$
$$=\Gamma_6+\Gamma_8+\Gamma_7+\Gamma_8(+\Gamma_7+2[\Gamma_6+\Gamma_8]+\Gamma_7+\Gamma_8).$$

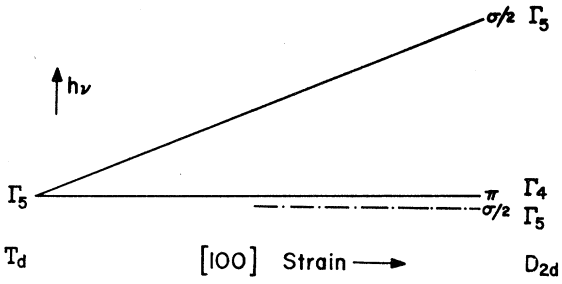

FIG. 2. Strain pattern (number of lines, strain ordering, and relative strengths) for a weakly bound two-carrier complex X, D^+-X, or A^--X, in [100]-strained material. A weak line is indicated by dots and dashes. The representations given correspond to initial states of exciton-recombination transitions. The [111]-strain pattern for a two-carrier complex is similar except that the higher-energy line is exchange-split into a strong and a weak line each of strength $\frac{1}{4}\sigma$.

[12] These relative strengths are for the free-atom transitions $s_{1/2} \rightarrow p_{3/2}$, $m_j=\pm\frac{1}{2}$ and $s_{1/2} \rightarrow p_{3/2}$, $m_j=\pm\frac{3}{2}$, respectively. The same relative strengths apply to the analogous group-T_d free-carrier transitions $\Gamma_6 \rightarrow \Gamma_8$, $m_j=\pm\frac{1}{2}$ and $\Gamma_6 \rightarrow \Gamma_8$, $m_j=\pm\frac{3}{2}$, respectively.

[13] M. A. Gilleo, P. T. Bailey, and D. E. Hill, J. Luminescence **1** (1969).

[14] K. Era, R. N. Euwema, and D. W. Langer, Bull. Am. Phys. Soc. **13**, 1476 (1968).

[15] The data presented in Fig. 5 of Ref. 2 are for a direction of strain near to, but not along, a [111] axis; however, Thomas notes that he obtained the same polarization properties also for [111] and [100] strains.

Experiments[9] have indicated that the exchange splittings between the two-hole states exceed those between the two-hole–one-electron states. We determine the ordering of these group T_d levels by identifying them with their analogs in a free atom containing two identical $p_{3/2}$ holes and an $s_{1/2}$ electron (Fig. 3). The ordering of the Pauli-allowed states is $\Gamma_8+\Gamma_7(J=\frac{5}{2}$, lowest), $\Gamma_8(J=\frac{3}{2})$, $\Gamma_6(J=\frac{1}{2}$, highest). Radiative exciton-recombination transitions occur between these A^0-X states and the impurity ground state $A^0(\Gamma_8)$. All four transitions are allowed, and from the coupling coefficients the relative strengths follow respectively as 2:1:2:1.[16] The analogous triplet in spherical symmetry[3,9] ($J=\frac{5}{2}, \frac{3}{2}, \frac{1}{2} \rightarrow j=\frac{3}{2}$) has relative strengths 3:2:1.

A^0-X Complex in [100]-Strained Material (Group D_{2d})

The two-hole representations are (Fig. 3)

$$\Gamma_7(h)\times\Gamma_7(h)=\Gamma_1(+\Gamma_2+\Gamma_5),$$
$$\Gamma_6(h')\times\Gamma_6(h')=\Gamma_1(+\Gamma_2+\Gamma_5),$$
$$\Gamma_7(h)\times\Gamma_6(h')=\Gamma_3+\Gamma_4+\Gamma_5,$$

where the Pauli principle has no effect on the product $\Gamma_7(h)\times\Gamma_6(h')$ because in this case the two holes have different spatial wave functions. The A^0-X representations are

$$\Gamma_7(h)\times\Gamma_7(h)\times\Gamma_6(e)=\Gamma_6(+\Gamma_6+\Gamma_6+\Gamma_7), \quad \text{states } \alpha$$
$$\Gamma_6(h')\times\Gamma_6(h')\times\Gamma_6(e)=\Gamma_6(+\Gamma_6+\Gamma_6+\Gamma_7), \quad \text{states } \beta$$
$$\Gamma_7(h)\times\Gamma_6(h')\times\Gamma_6(e)=\Gamma_7+\Gamma_7+\Gamma_6+\Gamma_7, \quad \text{states } \gamma.$$

For a radiative exciton-recombination transition there are two final states, corresponding to representations $\Gamma_7(h)$ and $\Gamma_6(h')$, but initial states of the types α and β can undergo transitions only to the final states belonging, respectively, to $\Gamma_7(h)$ and $\Gamma_6(h')$. Initial states of the type γ can undergo transitions to either final state.

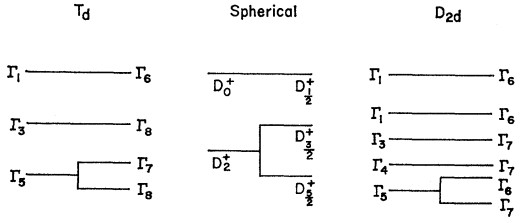

FIG. 3. Correspondence between A^0-X states in [100]-strained and in unstrained material, using free-atom states as a bridge. Within each column the two-identical-hole representations are at the left and the two-hole–one-electron representations are at the right. Compatible states are shown in horizontal alignment. The exchange ordering of the free-atom levels is known (Ref. 9), and the exchange ordering shown for the group T_d states follows from compatibility. The ordering shown in the group D_{2d} column approximates the exchange ordering as the [100] strain approaches zero but gives no information regarding strain ordering (see Fig. 4).

[16] We observe only a doublet for Zn^0-X in unstrained GaAs, and an A^0-X doublet in GaAs is mentioned also in Ref. 5. A polarization experiment on the triplet which we reported in Ref. 3 showed that residual strain was present in our sample (see also Ref. 21). If only a doublet is resolved, and if the hole-hole splitting exceeds the hole-electron splitting, then the relative strengths of the two lines should be 5:1.

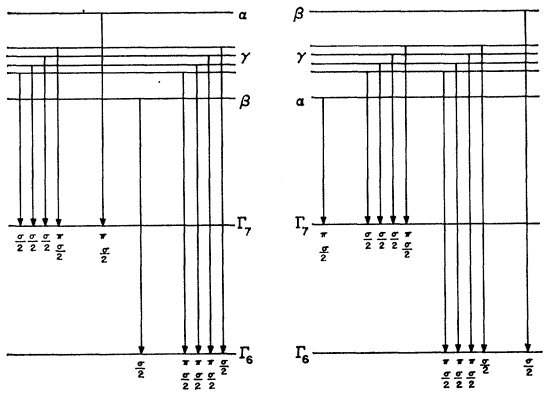

FIG. 4. Possible strain orderings of the levels and lines of an A^0-X complex in [100]-strained material. The separations between the initial states α, β, and γ are exaggerated relative to the separation between the final states Γ_7 and Γ_6. The exchange ordering shown for the γ lines is arbitrary. Experiment (Ref. 18) indicates that in GaAs the strain ordering on the left applies.

The strain ordering of the $\alpha \rightarrow \Gamma_7$ lines relative to the $\beta \rightarrow \Gamma_6$ lines is known (Figs. 4 and 5), because the separation between the initial states is greatly exceeded by the separation between the final states.[2] It remains to determine the strain ordering of the $\gamma \rightarrow \Gamma_7$ and $\gamma \rightarrow \Gamma_6$ lines relative to the $\alpha \rightarrow \Gamma_7$ and $\beta \rightarrow \Gamma_6$ lines. The γ levels correspond to a complex containing a hole from each band, so these levels lie between the α and β levels. But we cannot predict the ordering of levels α and β.[17] If β is above α then the $\gamma \rightarrow \Gamma_7$ and $\gamma \rightarrow \Gamma_6$ lines lie between the $\alpha \rightarrow \Gamma_7$ and $\beta \rightarrow \Gamma_6$ lines, while if α is above β the γ lines lie outside the α and β lines (a third possibility is that the levels α, β, and γ coincide, but

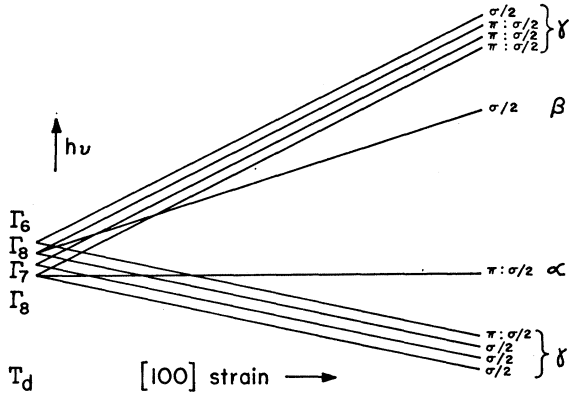

FIG. 5. Strain pattern for A^0-X in [100]-strained material. The group-T_d representations at the left are for A^0-X states in unstrained material.

[17] Even if the $\Gamma_7(m_j=\pm\frac{1}{2})$ and $\Gamma_6(m_j=\pm\frac{3}{2})$ holes are regarded as light and heavy, respectively [E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957)], the theory for the binding energy of an A^0-X complex still does not predict the ordering of the levels α and β (see Refs. 12–14 cited in Ref. 3).

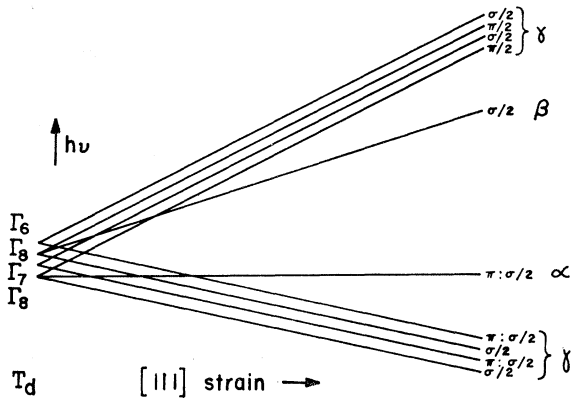

FIG. 6. Strain pattern for A^0-X in [111]-strained material. The group-T_d representations at the left are for A^0-X states in unstrained material.

this is improbable). Experiment[18] indicates that in GaAs the second alternative occurs, i.e., that the α level lies above the β level.[19] When the relative strengths in π and σ polarizations are determined for each line, the resulting strain pattern for A^0-X (Fig. 5) is markedly different from the two-carrier patterns. Data for Zn^0-X in [100]-strained GaAs appear to conform to this pattern.[18]

A^0-X Complex in [111]-Strained Material (Group C_{3v})

The strain pattern in this case (Fig. 6) differs from the [100] pattern (Fig. 5) in the polarizations of some of the γ lines. No data are available for this case.

Neutral Donor-Exciton Lines

For a D^0-X complex the [100] and [111] strain patterns are the same (Fig. 7), and they are identical with the free-carrier [100] and [111] strain patterns.[20] A [100] strain experiment has shown that a line previously identified with an Se^+-X complex in GaAs[3] actually arises from an Se^0-X complex.[21]

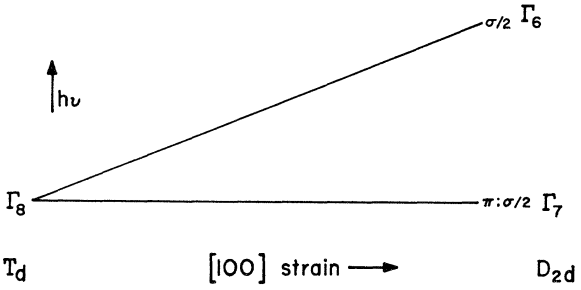

FIG. 7. Strain pattern for D^0-X in [100]- or [111]-strained material (identical with the [100] and [111] strain pattern for free carriers). The group-T_d representation at the left is for the D^0-X state in unstrained material, the group-D_{2d} representations at the right for D^0-X in [100]-strained material. For the [111] case (group C_{3v}) the higher-energy D^0-X level belongs to the Kramers pair of representations Γ_5, Γ_6, and the lower-energy level belongs to the representation Γ_4.

[18] M. A. Gilleo (private communication).

[19] Of the several strain orderings presented in this paper, the ordering under discussion is the only one which does not follow from our approach (Sec. II).

V. DISCUSSION

The uniaxial-strain patterns (number of lines, ordering, and strengths) which we have determined are different for the three cases X, A^0-X, and D^0-X. The predictions agree with experiment in the case of two-carrier complexes, and appear to agree with the limited available data for three-carrier complexes. There is thus a new means to identify the types of complexes involved in band-edge optical transitions in a direct zinc-blende-structure semiconductor. Further, because each line for a given complex can be identified from its relative strengths in π and σ polarizations, the exchange splittings in the strained material can also be obtained from experiment. Our approach can be applied to a direct semiconductor of structure different from zinc blende, and also to a case where there is a change of symmetry arising from an external field rather than a strain. Extension to indirect semiconductors is a possibility, using the work of Lax and Hopfield[22] as a guide.

Note added in proof. The four-carrier complex X_2, or excitonic molecule, has been observed, bound to a nitrogen trap, in the indirect zinc-blende semiconductor gallium phosphide.[23] The excitonic molecule in a direct zinc-blende semiconductor may be identified from the strain pattern which our approach predicts for it (initial states X_2, final states X).

ACKNOWLEDGMENTS

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[20] Bogardus and Bebb (Ref. 4) have observed a weak shoulder 0.5 meV above the line which they identify with D^0-X in unstrained GaAs. This, they state, might be attributed to $j-j$ splitting of the energy levels of the D^0-X system. Their statement differs from the conclusion (Refs. 6, 8, 9) that the Pauli principle allows only a singlet for D^0-X.

[21] M. A. Gilleo, P. T. Bailey, and D. E. Hill (to be published). In Ref. 3 an "Se^+-X doublet" was reported: Our strain experiments and a polarization experiment at zero strain have shown that the doublet was due to residual strain in the sample (see also Ref. 16).

[22] M. Lax and J. J. Hopfield, Phys. Rev. **124**, 115 (1961); M. Lax, in *Proceedings of the International Conference on the Physics of Semiconductors, Exeter, 1962* (The Institute of Physics and The Physical Society, London, 1962), pp. 395–402.

[23] R. A. Faulkner, J. L. Merz, and P. J. Dean, Solid State Commun. **7**, 831 (1969); J. L. Merz, R. A. Faulkner, and P. J. Dean (to be published).