PHYSICAL REVIEW B VOLUME 42, NUMBER 18 15 DECEMBER 1990-II

Coulomb-interaction effects on the excited states of double acceptors in germanium and silicon

Andreas Giesekus and L. M. Falicov
Department of Physics, University of California, Berkeley, California 94720

(Received 6 August 1990)

Infrared spectra of double-acceptor impurities in Si and Ge show small splittings. This fine structure cannot be explained by a split ground-state manifold only. An analysis of the Coulomb hole-hole splitting of the excited states, an estimate of the parameters involved, and dipole transition probabilities between the various ground-state symmetries and the excited states are presented. The spacing of accessible states may be large enough to be experimentally resolved.

I. INTRODUCTION

Studies of the infrared-absorption spectra of double-acceptor (DA) impurities in germanium[1-4] and silicon[5-7] show that the main transitions are very similar to the spectra of single acceptors in the position of the lines and their oscillator strength. However, high-resolution spectra reveal a fine structure that is, so far, not completely understood.

Giesekus and Falicov[8] investigated the impact of the Coulomb hole-hole interaction on the ground-state manifold by means of a mean-field approach, based on effective-mass-like single-particle states. It is possible to distinguish experimentally between ground- and excited-state splittings by analyzing the temperature dependence of the spectra. Experiments on DA impurities show clearly that the infrared-absorption spectra cannot be understood by assuming a split ground-state manifold only. It is necessary to include splitting of the excited states as well. This splitting might be caused by the hole-hole Coulomb interaction, although the heliumlike envelope functions of the effective-mass two-particle states have very little overlap. The purpose of the present contribution is to investigate qualitatively the influence of a hole-hole Coulomb interaction on the excited-state manifold, the estimation of the parameters involved, and a calculation of the dipole transition probabilities for the resulting states.

II. SINGLE-PARTICLE STATES

For shallow single acceptors it is well established that the effective-mass approximation reproduces experimental results very accurately:

$$\Psi = F_{n,l,m_l}(\mathbf{r})\Phi_{j,m_j}(\mathbf{r}) \ , \tag{1}$$

where F_{n,l,m_l} denotes a hydrogenic envelope function with a typical length scale of an effective Bohr radius, ≈ 50 Å, and Φ_{j,m_j} are the Bloch functions at the top of the valence band (which is the Γ point, $\mathbf{k}=0$, in the Brillouin zone for Ge and Si). Their typical length scale is determined by the lattice constant for the host material (≈ 5 Å). The indices j and m_j are the quantum numbers of the spin-orbit-coupled angular momentum of the semiconductor p functions. Bloch states with $j=\frac{3}{2}$ ($j=\frac{1}{2}$) correspond to the four topmost valence states (the two split-off states).

The energy of the excited states is in excellent quantitative agreement between theory[9,10] and experiment (even for very deep centers), because the amplitudes of higher hydrogenlike envelope functions vanish at the origin and therefore they neither probe the central cell nor do they have a large overlap with the 1s orbital. Hence, their properties are determined by the properties of the host material only and the details of the central cell have little effect.

In this contribution, we restrict ourselves to envelope functions with $n=2$, $l=1$, $m_l=1,0,-1$ and to the four topmost Bloch states with $j=\frac{3}{2}$, $m_j=\frac{3}{2},\ldots,-\frac{3}{2}$. In order to reach good quantitative agreement, it may be necessary to also include f-like envelopes into a variational space. Here only the twelve states with p-like envelopes are considered.

The four s-like single-particle ground states are represented by the quantum numbers $n=1$, $l=0$, $m_l=0$, $j=\frac{3}{2}$, $m_j=\frac{3}{2},\ldots,-\frac{3}{2}$. Again, variational calculations should improve if a small d-like component is included.

III. COULOMB INTERACTION

The effect of the Coulomb interaction on the ground-state configuration with a $(1s)^2$ heliumlike envelope is discussed in detail in Ref. 8. There it was found that the parameters that determine the splitting size depend on the probabilities that the holes occupy the same and neighboring sites in the lattice. These probabilities might in fact become fairly large because of the large overlap between the two s envelope functions. The situation in the case of the excited states with a $(1s2p)$-like configuration is very different. The probability for the holes getting close is very small, and the effect of the Coulomb interactions on the Bloch functions $\Phi_{3/2,m_j}$ may be neglected, i.e., we treat the Coulomb interaction for the excited states similarly to the triplet-singlet splitting in helium and assume that the Coulomb interaction operates only on the envelope degrees of freedom.

In the case of helium, the spin and orbital degrees of freedom are almost uncoupled and may be symmetrized separately. The Pauli principle requires the total state to be antisymmetric under an exchange of particles. As a consequence, the triplet, which is symmetric under exchange, must have an antisymmetric ($1s2p$)-orbital wave function that is lower in energy by ($-J_{\mathrm{exch}}$), whereas the singlet with a symmetric combination of s and p orbitals is higher in energy by ($+J_{\mathrm{exch}}$). (The symbol J_{exch} denotes the exchange integral.) This parameter J_{exch} is well known for helium, its value is $\approx 0.5\times 10^{-2}$ hartrees (1 hartree $=27.2$ eV). In the semiconductors, the effective energy unit is reduced because of the (static) dielectric constant ϵ and the effective mass of the hole m_h^* (in units of the electron mass):

$$E_H=\frac{m_h^*}{\epsilon^2} \tag{2}$$

in hartrees. The order of magnitude of this effective Hartree energy is given in Table I. The first entry in the table gives the result based on the scaling factor in (2). All other entries are based on experimental first and second ionization energies, which are made equal, as in He, to $0.9E_h$ and $2E_H$. Experimental results for acceptor impurities in Ge are fairly consistent. However, Si-based systems show a strong dependence of the binding energies on the particular impurity (i.e., a "chemical shift"). They are considerably larger than the simple rescaled estimate. This trend may be explained by the fact that bound 1s holes probe the central cell. There the potential is deeper than the screened Coulomb potential and binds the s holes more tightly. The energy of the p states relative to the valence-band edge has no chemical shift (no matter how deep the impurity is) indicating that the spatial extent of the p states remains unchanged "chemistry." As a consequence, for deep impurities the parameter J_{exch} is *smaller* than for shallower DA's simply because the s states are more confined, whereas the p states are not. Hence, the influence of the Coulomb interaction on excited states exhibits the opposite trend to the Coulomb splitting of the ground-state manifold: for increasing central cell binding the Coulomb energy contribution decreases in the excited states but increases in the ground-state manifold.

TABLE I. Effective Hartree energy for double acceptors in Si and Ge. The first entry is based on the static dielectric constant and the hole effective mass. All others are based on first and second ionization energies and are therefore impurity dependent. This dependency measures the deviation of the impurity potential from a screened Coulomb potential at the central cell.

	E_h(Si)	E_h(Ge)
ϵ, m_h^*	67.6	20.9
Be^0	213	28
Be$^-$		29
Zn^0	356	37
Zn$^-$	310	44

IV. HAMILTONIAN MATRIX

A. Single-particle terms

The states that diagonalize the single-particle terms of the Hamiltonian may be obtained by symmetry considerations only. We write the 48 relevant two-hole states in terms of the antisymmetrized basis states,

$$\begin{aligned}|sm_j,m_lm_j'\rangle=\sqrt{\tfrac{1}{2}}[&F_{1,0,0}(\mathbf{r}_1)F_{2,1,m_l}(\mathbf{r}_2)\\ &\times\Phi_{3/2,m_j}(\mathbf{r}_1)\Phi_{3/2,m_j'}(\mathbf{r}_2)\\ &-F_{2,1,m_l}(\mathbf{r}_1)F_{1,0,0}(\mathbf{r}_2)\\ &\times\Phi_{3/2,m_j'}(\mathbf{r}_1)\Phi_{3/2,m_j}(\mathbf{r}_2)]\ .\end{aligned} \tag{3}$$

The unitary transformation U_1 that transforms the basis (3) into a basis with proper symmetry (tetrahedral group) one-particle quantum numbers is conveniently obtained as follows.

(a) The four single-particle s states are degenerate because they transform according to the representation Γ_8 of the point group $\overline{T}_d$.

(b) The twelve single-particle p states transform according to $\Gamma_5\otimes\Gamma_8$. This product decomposes into $\Gamma_6\oplus\Gamma_7\oplus 2\Gamma_8$. The various symmetry components may be projected out conveniently in two steps.

(i) The envelope angular momentum ($l=1$) and the angular momentum of the Bloch state ($j=\frac{3}{2}$) are coupled to a total angular momentum of the p state with a resulting $I=\frac{5}{2},\frac{3}{2},\frac{1}{2}$. [This coupling is essential for the understanding of the excited states. Without the coupling of the envelope angular momentum (analog to the orbital angular momentum of helium) and the angular momentum of the Bloch states (analog to the spin in helium), no splitting of the single-particle excited states would occur. The splitting of the two-particle states would be irrelevant as well, because (as in helium, where spin-orbit coupling is extremely small) the ^3P triplet would not be accessible by a dipole transition under these circumstances.]

(iia) The states with angular momentum $I=\frac{3}{2}$ transform according to Γ_8 and are not split by the symmetry of the system. Similarly, the states with angular momentum $I=\frac{1}{2}$ transform according to Γ_6 are not split either.

(iib) The six states with $I=\frac{5}{2}$, $M_I=\frac{5}{2},\ldots,-\frac{5}{2}$ split into a Γ_7 and a Γ_8 component:

$$\Gamma_8:\quad\begin{cases}\sqrt{\frac{5}{6}}|M_I=\frac{5}{2}\rangle+\sqrt{\frac{1}{6}}|M_I=-\frac{3}{2}\rangle\ ,\\ |M_I=\frac{1}{2}\rangle\ ,\\ |M_I=-\frac{1}{2}\rangle\ ,\\ \sqrt{\frac{5}{6}}|M_I=-\frac{5}{2}\rangle+\sqrt{\frac{1}{6}}|M_I=\frac{3}{2}\rangle\ ,\end{cases} \tag{4a}$$

$$\Gamma_7:\quad\begin{cases}\sqrt{\frac{1}{6}}|M_I=\frac{5}{2}\rangle-\sqrt{\frac{5}{6}}|M_I=-\frac{3}{2}\rangle\ ,\\ \sqrt{\frac{1}{6}}|M_I=-\frac{5}{2}\rangle-\sqrt{\frac{5}{6}}|M_I=\frac{3}{2}\rangle\ .\end{cases} \tag{4b}$$

These unitary one-particle transformations provide orbitals with the proper symmetries. They are combined into a 48×48 direct-product unitary matrix that orders the excited two-particle states according to the symmetries of the s and the p hole $\Gamma_8\otimes\Gamma_6$, $\Gamma_8\otimes\Gamma_7$, $\Gamma_8\otimes\Gamma_8-I=\frac{3}{2}$, and $\Gamma_8\otimes\Gamma_8-I=\frac{5}{2}$.

B. Two-particle interaction terms

In order to diagonalize the Coulomb-interaction operator, we do not need to decompose the direct-product states given above into their fully irreducible representations. A unitary transformation U_2 that diagonalizes the two-particle interaction is conveniently obtained as follows. For the model introduced in Sec. III, it is sufficient to build linear combinations of the basis (3) with antisymmetrized (under particle exchange) envelopes and symmetrized Bloch functions or vice versa. We obtain this by combining the Bloch states into angular-momentum symmetrized products with angular momentum $J=3,2,1,0$. The states with $J=3$ and 1 are symmetric with respect to particle exchange, hence the two envelope functions must be antisymmetrically combined. The states with $J=2$ and 0 are antisymmetric. For these states the envelope must be symmetric.

C. Total Hamiltonian

In the case of helium, it is possible to diagonalize one- and two-particle terms of the Hamiltonian at once. This is because the $(1s2p)$ configuration consists of a single term. Here the coupling of the envelope and Bloch angular momenta destroys this convenience and we must transform the two parts of the Hamiltonian to a common basis before diagonalizing the total Hamiltonian,

$$H_{\text{tot}}=U_1D_1U_1^\dagger+U_2D_2U_2^\dagger\ , \tag{5}$$

which is now written in terms of the basis (3). The matrix D_1 (D_2) is the diagonalized one-particle (two-particle) Hamiltonian matrix. The matrix elements of D_1 (the one-particle energies) are taken from Binggeli and Baldereschi,[10] and the elements of D_2 are simply $(\pm J_{\text{exch}})$ with respect to the symmetry of the envelope functions. The final diagonalization of H_{tot} is performed numerically.

The symmetry of the DA center yields the following degeneracies. The manifold of states with one-particle-symmetry $\Gamma_8\otimes\Gamma_8$ splits into seven levels with symmetries $\Gamma_1\oplus\Gamma_2\oplus\Gamma_3\oplus 2\Gamma_4\oplus 2\Gamma_5$, whereas the $\Gamma_8\otimes\Gamma_7$ and $\Gamma_8\otimes\Gamma_6$ levels split into $\Gamma_3\oplus\Gamma_4\oplus\Gamma_5$. The Γ_1 and Γ_2 levels are nondegenerate, Γ_3 is a twofold-degenerate level, and the levels with Γ_4 and Γ_5 symmetries are threefold degenerate. There are two quadratic secular equations, corresponding to Γ_1 and Γ_2, a quartic secular equation corresponding to Γ_3, and two sixth-order secular equations for Γ_4 and Γ_5.

V. DIPOLE TRANSITIONS

The dipole transition probability for absorption is proportional to the absolute square of the dipole matrix element between initial and final states. The final states are the eigenstates of H_{tot} [Eq. (5)]. They are obtained numerically.

We treat the $(1s)^2\rightarrow(1s2p)$ transition as a single-particle transition, where one of the holes remains in a $1s$ state and all Bloch-function quantum numbers are conserved. Therefore the only components of the initial states that connect to the excited states are $\Gamma_8\otimes\Gamma_8$ components, with two $j=\frac{3}{2}$ Bloch functions. They are written as

$$\begin{aligned}|sm_j,sm_j'\rangle=\sqrt{\tfrac{1}{2}}F_{1,0,0}(\mathbf{r}_1)F_{1,0,0}(\mathbf{r}_2)\\ \times[\Phi_{3/2,m_j}(\mathbf{r}_1)\Phi_{3/2,m_j'}(\mathbf{r}_2)\\ -\Phi_{3/2,m_j'}(\mathbf{r}_1)\Phi_{3/2,m_j}(\mathbf{r}_2)]\ .\end{aligned} \tag{6}$$

In a system with $\overline{T}_d$ point symmetry, spectroscopy with polarized photons yields identical results to unpolarized spectroscopy, because none of the cubic axes is preferred. Therefore, without loss of generality, we may restrict the calculation of transition probabilities to the case of polarization in the cubic z direction. Then the $1s$ ground states connect only to those excited states with a p_z envelope function, i.e., with the $F_{n=2,l=1,m_l=0}$ function. This fact eliminates the transformation from $l=1$, $m_l=1,0,-1$ to p_x,p_y,p_z orbitals.

In order to obtain the transition probabilities, the matrix elements of the electric dipole operator for z-polarized photons $\hat{d}_z$ in the present approximation are written in terms of the basis states (3) and (6):

$$\begin{aligned}&\langle sm_j,sm_j'|\hat{d}_z|sm_j'',m_lm_j'''\rangle\\ &\qquad=C\delta_{m_l,0}(\delta_{m_j,m_j''}\delta_{m_j',m_j'''}-\delta_{m_j,m_j'''}\delta_{m_j',m_j''})\ .\end{aligned} \tag{7}$$

The first Kronecker symbol reflects the fact that the z-polarized photons cause a transition from an s envelope to a p_z envelope only, and the Kronecker symbols within the parentheses enforce the conservation of the Bloch quantum numbers; C is a constant.

The basis (6) does not provide a complete space for the initial states,[8] because the Coulomb hole-hole interaction strongly mixes the states of $\Gamma_8\otimes\Gamma_8=\Gamma_1\oplus\Gamma_3\oplus\Gamma_5$ symmetries with other states of the same symmetry. In particular the Γ_3 and Γ_5 states are hybridized with those consisting of one $j=\frac{3}{2}$ and one $j=\frac{1}{2}$ Bloch orbitals $(\Gamma_8\otimes\Gamma_8=\Gamma_3\oplus\Gamma_4\oplus\Gamma_5)$, and the Γ_1 state mixes with that consisting of two $j=\frac{1}{2}$ Bloch orbitals. This hybridization, which has not been explicitly considered here, may lead to slightly different oscillator strengths for the lowest states, as well as to nonvanishing oscillator strengths from some of the higher states in the ground-state manifold. Inclusion of this hybridization effect is straightforward.

For the sake of completeness, we list the decomposition of the $\Gamma_8\otimes\Gamma_8$ initial-state components:

TABLE II. Excited-state splittings and oscillator strengths. All energies are given in meV, with $J_{\text{exch}}=0.25$ meV. The entries in the different columns are (from left to right) single-particle symmetry and single-particle energy, energy of the excited state, the same in first-order perturbation theory (in parentheses), two-particle symmetry and degeneracy, oscillator strength for the Γ_1 initial state, for the Γ_3 initial state, and for the Γ_5 initial state.

Excited-state one-particle symmetry	Energy shift (exact)	Energy shift (perturbation)	Symmetry		Oscillator strength		
					Γ_1	Γ_3	Γ_5
$\Gamma_8\otimes\Gamma_8$	−0.250	(−0.250)	Γ_2	(1)			
$I=\frac{3}{2}$	−0.159	(−0.150)	Γ_1	(1)			0.018 34
$\epsilon_8=-15.8$	−0.158	(−0.150)	Γ_4	(3)		0.027 93	0.038 06
	−0.157	(−0.150)	Γ_5	(3)	0.000 03	0.051 27	0.023 94
	−0.061	(−0.050)	Γ_4	(3)		0.121 16	0.038 60
	−0.058	(−0.050)	Γ_3	(2)			0.082 23
	−0.179	(0.183)	Γ_5	(3)	0.318 49	0.108 83	0.103 81
$\Gamma_8\otimes\Gamma_8$	−0.250	(−0.250)	Γ_2	(1)			
$I=\frac{5}{2}$	−0.200	(−0.200)	Γ_3	(2)			0.022 34
$\epsilon_8=-11.7$	−0.196	(−0.194)	Γ_5	(3)	0.051 55	0.008 99	0.011 00
	−0.098	(−0.100)	Γ_4	(3)		0.138 50	0.010 73
	0.052	(0.050)	Γ_4	(3)		0.029 61	0.182 66
	0.078	(0.773)	Γ_5	(3)	0.285 70	0.148 06	0.024 94
	0.159	(0.150)	Γ_1	(1)			0.092 78
$\Gamma_8\otimes\Gamma_6$	−0.250	(−0.250)	Γ_3	(2)			0.000 00
$\Gamma_8\otimes\Gamma_7$	−0.161	(−0.167)	Γ_4	(3)		0.000 00	0.063 27
$\epsilon_6=\epsilon_7=-6.1$	−0.075	(−0.083)	Γ_4	(3)		0.182 81	0.000 01
	−0.075	(−0.083)	Γ_5	(3)	0.000 03	0.182 85	0.000 01
	0.007	(0.000)	Γ_3	(2)			0.117 65
	0.172	(0.167)	Γ_5	(3)	0.344 22	0.000 00	0.169 64

TABLE III. The same as Table II in first-order perturbation theory. All energies are in units of J_{exch}.

Excited-state one-particle symmetry	Energy shift	Symmetry		Oscillator strength		
				Γ_1	Γ_3	Γ_5
$\Gamma_8\otimes\Gamma_8$	−1.000	Γ_2	(1)			
$I=\frac{3}{2}$	−0.600	$\Gamma_1\oplus\Gamma_4\oplus\Gamma_5$	(7)		0.093 33	0.093 33
	−0.200	$\Gamma_3\oplus\Gamma_4$	(5)		0.133 33	0.133 33
	0.733	Γ_5	(3)	0.333 33	0.106 67	0.106 67
$\Gamma_8\otimes\Gamma_8$	−1.000	Γ_2	(1)			
$I=\frac{5}{2}$	−0.800	Γ_3	(2)			0.022 22
	−0.776	Γ_5	(3)	0.054 05	0.008 59	0.013 60
	−0.400	Γ_4	(3)		0.133 33	0.011 11
	0.200	Γ_4	(3)		0.033 33	0.177 78
	0.309	Γ_5	(3)	0.279 29	0.158 07	0.019 74
	0.600	Γ_1	(1)			0.088 89
$\Gamma_8\otimes\Gamma_6$	−1.000	Γ_3	(2)			0.000 00
$\Gamma_8\otimes\Gamma_7$	−0.667	Γ_4	(3)		0.000 00	0.055 56
	−0.333	$\Gamma_4\oplus\Gamma_5$	(6)		0.333 33	0.000 00
	0.000	Γ_3	(2)			0.111 11
	0.667	Γ_5	(3)	0.333 33	0.000 00	0.166 67

$$|\Gamma_1\rangle=\sqrt{\tfrac{1}{2}}(|s\tfrac{1}{2},s-\tfrac{1}{2}\rangle-|s\tfrac{3}{2},s-\tfrac{3}{2}\rangle) \tag{8a}$$

$$|\Gamma_3\rangle=\begin{cases}\sqrt{\tfrac{1}{2}}(|s\tfrac{3}{2},s\tfrac{1}{2}\rangle+|s-\tfrac{1}{2},s-\tfrac{3}{2}\rangle)\ ,\\ \sqrt{\tfrac{1}{2}}(|s\tfrac{1}{2},s-\tfrac{1}{2}\rangle+|s\tfrac{3}{2},s-\tfrac{3}{2}\rangle)\ ,\end{cases} \tag{8b}$$

$$|\Gamma_5\rangle=\begin{cases}\sqrt{\tfrac{1}{2}}(|s\tfrac{3}{2},s\tfrac{1}{2}\rangle-|s-\tfrac{1}{2},s-\tfrac{3}{2}\rangle)\ ,\\ |s\tfrac{3}{2},s-\tfrac{1}{2}\rangle\ ,\\ |s\tfrac{1}{2},s-\tfrac{3}{2}\rangle\ .\end{cases} \tag{8c}$$

VI. RESULTS AND DISCUSSION

Tables II and III present numerical results. For the one-particle energies we choose the values obtained by Binggeli and Baldereschi.[10] They are listed together with the one-particle symmetries in the first column. The states with Γ_6 and Γ_7 symmetries are, following also Binggeli and Baldereschi, assumed to be accidentally degenerate.[10] The second column provides the energies of the Coulomb-split excited states relative to their one-particle energy. The values in parentheses are the results in first-order perturbation theory. This approximation is, as expected, in very good agreement with the full diagonalization for small values of J_{exch}. The approximation has the advantages that it may be performed analytically and that the oscillator strengths (the last three columns) do *not* depend[11] on the actual value of J_{exch}. The oscillator strength and energy spacings in units of J_{exch} obtained by first-order perturbation theory are listed in Table III. The quoted oscillator strengths contain a summation over all degenerate final states and an average over degenerate initial states.

An analysis of Tables II and III yields the following conclusions.

(1) If the DA ground state has Γ_1 symmetry, the DA spectrum is very similar to the single-acceptor spectrum. There are essentially three accessible excited states—a fourth one has only a very small oscillator strength—that are spaced by (very nearly) the single-particle energy differences.

(2) This situation is changed if the ground state has either Γ_3 or Γ_5 symmetry. Then strong transitions to more excited states become possible. In these cases, typical spacing between accessible levels in a given excited-state configuration is between $0.4J_{\mathrm{exch}}$ and J_{exch}.

(3) These results clearly show that a more quantitative theory on DA's must necessarily include hole-hole Coulomb interaction. The fine structure of the spectra cannot be explained otherwise.

ACKNOWLEDGMENTS

The authors are grateful to E. E. Haller, J. Heyman, and E. Merk for many stimulating discussions. One of us (A.G.) gratefully acknowledges the support from the Deutsche Forschungsgemeinschaft (DFG). This work was supported by the National Science Foundation through Grant No. DMR-88-06756.

---

[1] M. L. W. Thewalt, D. Labrie, I. J. Booth, B. P. Clayman, E. C. Lightowlers, and E. E. Haller, Physica B+C **146B**, 47 (1987); M. L. W. Thewalt, B. P. Clayman, and D. Labrie, Phys. Rev. B **32**, 2663 (1985).

[2] W. J. Moore, J. Chem. Phys. Solids **32**, 93 (1971); Phys. Rev. B **29**, 7062 (1984).

[3] T. A. Callot and J. W. MacKay, Phys. Rev. **161**, 698 (1967).

[4] R. K. Crouch, J. B. Robertson, and T. E. Cilimer, Phys. Rev. B **5**, 3111 (1972).

[5] E. Merk, J. Heyman, and E. E. Haller, Solid State Commun. **72**, 851 (1989), and private communication.

[6] J. Heyman, A. Giesekus, L. M. Falicov, and E. E. Haller, Bull. Am. Phys. Soc. **35**, 279 (1990), and private communication.

[7] A. Dörnen, R. Kienle, K. Thonke, P. Stolz, G. Pensl, D. Grünebaum, and N. A. Stolwijk, Phys. Rev. **40**, 12 005 (1989).

[8] A. Giesekus and L. M. Falicov, Phys. Rev. B **42**, 8975 (1990).

[9] N. O. Lipari and A. Baldereschi, Solid State Commun. **25**, 655 (1978).

[10] N. Binggeli and A. Baldereschi, Solid State Commun. **66**, 323 (1988).

[11] It should be noted that the calculation, when treated in first-order perturbation theory, contains for each degenerate manifold a single parameter, J_{exch}; the results for the oscillator strength are therefore parameter independent.