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Electric field effects on excitons and shallow donor impurities

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The first nonperturbative calculation of optical absorption by a hydrogenic impurity in an electric field including transition-matrix elements is presented. A nonperturbative method developed by Duke and Alferieff to treat the effects of an electric field on excitons is applied to shallow donor impurity states. The model potential consists of the Coulomb potential near the origin and a uniform electric field potential far from the origin. The potential switches from the Coulomb to the electric field potential at a crossover boundary. We show that the calculated absorption spectrum is strongly dependent on the choice of the crossover boundary and introduce a new choice for the crossover boundary. Using this new definition we calculate the absorption spectrum for both excitons and impurity states. The results closely resemble the experimental data. We also show that Elliot's approximation for optical absorption in excitons can be applied to shallow donor impurities at low electric fields.

INTRODUCTION

The effect of an electric field on the hydrogenic levels of shallow impurity states and excitons has received considerable attention.[1-24] Theoretical studies of the electric field effects are based on either the perturbation theory (first- and second-order Stark effect) or on numerical approximations.[1-5,11,12]

A nonperturbative theory of optical absorption by a hydrogenic system in a crystal in the presence of static electric field was first examined by Duke and Alferieff.[9] Duke and Alferieff (DA) calculated the optical absorption spectrum for excitons in a crystal subject to an electric field using an approximate potential. Comparison of the DA results with experiments[2,13-19] shows that their model overestimates the effect of the electric field on exciton excited states. It is also found that the predictions of the DA model are highly dependent on the choice of the crossover criteria.

In this paper we give another alternative for the choice of the crossover value. We recalculate the absorption spectrum for excitons in a crystal subject to an electric field by using this new definition. Compared to the experiments[13-19] the new definition of the crossover value describes the electric field energy shifts and broadening of exciton states more accurately.

We also apply the DA model which gives analytical solutions to the calculation of the impurity absorption spectrum. We calculate the impurity absorption by including the actual transition-matrix elements. Among all the theoretical papers in the literature concerning electric field effects none have previously reported such calculations. Finally, we show that Elliot's[25] approximation for exciton absorption can be applied to impurity absorption at low electric fields.

THEORY

The theoretical studies discussed in this paper are based on the nonperturbative theory of electric field effects on hydrogenic systems proposed by Duke and Alferieff[9] (DA). In describing the exciton potential, DA used an approximate potential defined in two regions [in parabolic coordinates with coordinates $\xi=r+z$, $\eta=r-z$, $\phi=\tan^{-1}(y/x)$]. In region I the potential is Coulomb and in region II it is the electric field potential. The boundary or crossover between the two potentials occurs at a value designated as $\rho_1^0=\rho_2^0=x_0/n$, where $\rho_1=\xi/n$ and $\rho_2=\eta/n$. The location of the crossover for the potential is determined by finding the points where the effective Coulomb and electric field potentials are equal when the principal quantum number $n=1$ and the parabolic quantum numbers $n_1=n_2=0$, where $n=n_1+n_2+1+|m|$ and $n=-1/(-2E)^{1/2}$.[9] For a hydrogen atom, n_1, n_2, and m determines the discrete spectrum in parabolic coordinates. In the presence of an electric field, the spectrum is continuous and consequently quantum numbers n, n_1, and n_2 are not confined to integer values.

DA solved the differential equation

$$(\tfrac{1}{2}\Delta+\varepsilon z+r^{-1}+E)\Phi_E(r)=0 \tag{1}$$

in parabolic coordinates, analytically, in each of the two regions. The effective electric field ε is given by

$$\varepsilon=\frac{|e|\mu a_B^3 F}{\hbar^2}\ ,$$

in which F is the external electric field. μ is the reduced mass of the hydrogenic system, a_B is the Bohr radius, and e is the charge on an electron.

Separation of Eq. (1) in parabolic coordinates gives

$$\frac{d^2X_i}{d\rho_i^2}+\left[-\frac{1}{4}-\frac{m^2-1}{4\rho_i^2}-(-1)^i\frac{\varepsilon n^3\rho_i}{4}-\frac{n_i+\frac{1}{2}(1+|m|)}{\rho_i}\right]X_i=0\ ,$$

with the following solutions: For $\rho_1<\rho_1^0$,

$$X_1(\rho_1)=c_1 n^{1/2}\rho_1^{(1+|m|)/2}\times\exp(-\rho_1/2)M(-n_1,1+|m|,\rho_1)\ ;$$

for $\rho_1>\rho_1^0$,

$$X_1(\rho_1)=b_1\mathrm{Ai}(z_1)\ ;$$

for $\rho_2<\rho_2^0$,

$$X_2(\rho_2)=c_2 n^{1/2}\rho_2^{(1+|m|)/2}\times\exp(-\rho_2/2)M(-n_2,1+|m|,\rho_2)\ ;$$

for $\rho_2>\rho_2^0$,

$$X_2(\rho_2)=b_2\mathrm{Ai}(z_2)+b_3\mathrm{Bi}(z_2)\ ;$$

where c_1, c_2, b_1, b_2, and b_3 are constants,

$$z_1=(n^3\varepsilon/4)^{1/3}[(n^3\varepsilon)^{-1}+\rho_1]\ ,$$

$$z_2=(n^3\varepsilon/4)^{1/3}[(n^3\varepsilon)^{-1}-\rho_2]\ ,$$

$M(a,b,x)$ is the Kummer function, and $\mathrm{Ai}(y)$ and $\mathrm{Bi}(y)$ are the Airy functions. The solution of Eq. (1) is

$$\Phi_E(\rho_1,\rho_2,\phi)=\left[\frac{2}{\pi}\right]^{1/2}\left[\frac{X_1(\rho_1)X_2(\rho_2)}{(n^2\rho_1\rho_2)^{1/2}}\right]e^{im\phi}\ .$$

DA used the continuity of the logarithmic derivative of $X_1(\rho_1)$ at $\rho_1=\rho_1^0$ to determine the quantum number n_1.[9] DA calculated the optical absorption coefficient for excitons as a function of their initial energies,

$$\alpha_0(\varepsilon,\omega)=\varepsilon^{-1/3}|\Phi_{0,n_\omega,\varepsilon}(0)|^2\ , \tag{2}$$

where energy $E=(\hbar\omega-E_g)/E_0=-1/2n_\omega^2$, $\hbar\omega$ is the energy of the absorbed photon, and E_g is the zero-field band gap.

In DA's result the $n=2$ levels are extremely broadened or smeared out for $\varepsilon=0.05$ and for almost any interesting value of the electric field. The failure of the model to produce the reasonably sharp peaks for the $n>2$ levels is principally due to the definition of the crossover for the potential.

If the strength function $|\Phi_E(0)|^2$ is calculated as a function of the crossover value of ρ_1^0 it is found that the $n=2$ peaks move to lower energies and increases as ρ_1^0 increases in magnitude. This is illustrated in Fig. 1 for $\varepsilon=0.05$. It appears that for the DA model the peak positions and the peak strengths are extremely sensitive to the crossover value.

Our principal aim is to modify and apply the DA model to the calculation of the optical absorption by shallow donor impurities. More specifically, we use the wave functions of the DA model to calculate the optical absorption coefficient as a function of photon energy.

Various combinations of wave functions are needed depending upon the values of ρ_1 and ρ_2. One has

$$\Phi_E(\rho_1<\rho_1^0,\rho_2<\rho_2^0,\phi)=C_1 n^{1/2}\rho_1^{(1+|m|)/2}\exp(-\rho_1/2)M(-n_1,1+|m|,\rho_1)C_2 n^{1/2}\rho_2^{(1+|m|)/2}\exp(-\rho_2/2)\times M(-n_2,1+|m|,\rho_2)\exp(im\phi)(2/\pi)^{1/2}(1/n^2\rho_1\rho_2)^{1/2}\ ,$$

$$\Phi_E(\rho_1>\rho_1^0,\rho_2>\rho_2^0,\phi)=b_i\mathrm{Ai}(z_1)|b_2\mathrm{Ai}(z_2)+b_3\mathrm{Bi}(z_2)|\exp(im\phi)(2/\pi)^{1/2}(1/n^2\rho_1\rho_2)^{1/2}\ ,$$

$$\Phi_E(\rho_1<\rho_1^0,\rho_2>\rho_2^0,\phi)=C_1 n^{1/2}\rho_1^{(1+|m|)/2}\exp(-\rho_1/2)M(-n_1,1+|m|,\rho_1)|b_2\mathrm{Ai}(z_2)+b_3\mathrm{Bi}(z_2)|\times\exp(im\phi)(2/\pi)^{1/2}(1/n^2\rho_1\rho_2)^{1/2}\ ,$$

and

$$\Phi_E(\rho_1>\rho_1^0,\rho_2>\rho_2^0,\Phi)=C_2 n^{1/2}\rho_2^{(1+|m|)/2}\exp(\rho_2/2)M(-n_2,1+|m|,\rho_2)b_1\mathrm{Ai}(z_1)\exp(im\phi)(2/\pi)^{1/2}(1/n^2\rho_1\rho_2)^{1/2}\ .$$

In order to obtain a more realistic behavior for the $n=2$ peak, we used a different criteria for the location of the crossover for the potential. For $\rho_2>\rho_2^0$ the wave function is given by

$$X_2(\rho_2)=b_2\mathrm{Ai}(z_2)+b_3\mathrm{Bi}(z_2) \tag{3}$$

where

$$z_2=\left[\frac{n^3\varepsilon}{4}\right]^{1/2}[(n^3\varepsilon)^{-1}-\rho_2]\ . \tag{4}$$

The Airy functions[20] $\mathrm{Ai}(z_2)$ and $\mathrm{Bi}(z_2)$ are oscillatory functions when z_2 is negative. From the definition of z_2, it follows that when $\rho_2>(n^3\varepsilon)^{-1}$ the Airy functions will be oscillatory. This means that at the point where

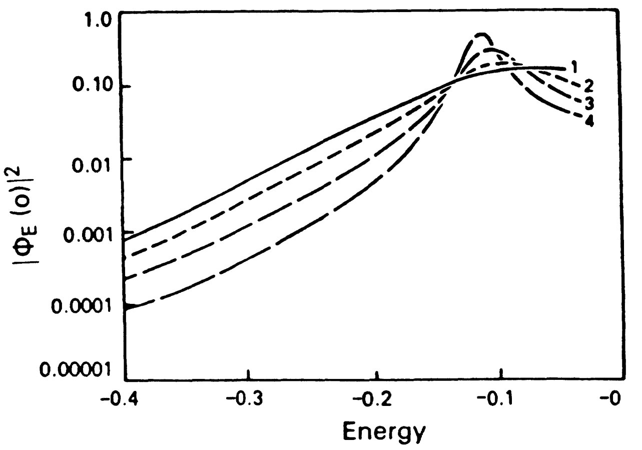

FIG. 1. Strength function square at the origin, $|\Phi_E(0)|^2$, at energies $E=(\hbar\omega-E_g)/E_0$, near the second exciton peak ($E=-0.125$, which corresponds to the principal quantum number $n=2$ when $\varepsilon=0$) for four different crossover values [1→6.5611, 2→10.5611 (DA value), 3→12.5611, 4→14.5611] with applied electric field $\varepsilon=0.05$.

$\rho_2=(n^3\varepsilon)^{-1}$ the effect of the electric field starts to produce oscillations. Therefore this point is selected as the crossover for the potential where $\rho_1=\rho_2$.

The one-electron Hamiltonian for shallow donor impurities includes the one-electron Hamiltonian for the perfect crystal plus the impurity potential. The impurity wave function consists of a Bloch function modulated by an envelope function. Since the Bloch functions for the initial and final states are the same, the value of the matrix element comes principally from the envelope functions. Consequently, selection rules and intensities can be calculated approximately from the envelope functions alone (effective-mass theory[26]).

Transition probabilities for shallow impurities is computed to first order by considering the matrix elements of the electron-photon interaction,

$$H'=\frac{e}{mc}\mathbf{A}\cdot\mathbf{P} ,$$

between the initial- and final-state wave functions. Here $\mathbf{A}$ is the electromagnetic vector potential and $\mathbf{P}$ is the momentum operator.

The transition probability amplitude between initial and final states a_{if} is given by

$$\mathbf{a}_{if}=\int \Phi^*_{E_i}(\mathbf{r})\mathbf{P}\Phi_{E_f}(\mathbf{r})d\mathbf{r} . \tag{5}$$

The transition probability per unit time is proportional to

$$\rho_{E_i\rightarrow E_f}\propto |\mathbf{e}\cdot\mathbf{a}_{if}|^2\delta(E_i-E_f-\hbar\omega) , \tag{6}$$

where $\mathbf{e}$ is the polarization vector in the direction of the electric field.

The optical absorption coefficient $\alpha(\omega)$ is proportional to the absolute square of the probability amplitude integrated over all initial and final states,

$$\alpha(\omega)\propto\int dE_i\int dE_f\,|\mathbf{e}\cdot\mathbf{a}_{if}|^2\delta(E_i-E_f-\hbar\omega) , \tag{7}$$

where $\hbar\omega$ is the energy of the photon causing the transition. We have carried out detailed calculations of $\alpha(\omega)$ using Eq. (7).

Before describing these results, we consider an approximate result which leads to a formula analogous to that used by DA for optical absorption by excitons. With the polarization in the z direction ($\mathbf{e}=\mathbf{e}_z$) one has

$$|\mathbf{e}\cdot\mathbf{a}_{if}|=\left|\int\Phi_{E_F}\nabla_z\Phi_{E_i}d\mathbf{r}\right|^2 . \tag{8}$$

When the electric field is small, the major contribution to $\nabla_z\Phi_{1s}$ is at the origin and the 1s band is highly peaked about the $\varepsilon=0$ energy E^0_{1s},

$$\nabla_z\Phi_{E_i}\simeq\nabla_z\Phi_{E_i}(0)\delta(r-0)\delta(E^0_{1s}-E_i)$$

and

$$\alpha(\omega)\propto[\,|\nabla_z\Phi_{E_{1s}}(0)|^2\,|\Phi_{E_f}(0)|^2] \text{ at } E_f=\hbar\omega+E^0_{1s}$$
$$=\text{const}\times|\Phi_{E_f}(0)|^2 . \tag{9}$$

Thus the absorption coefficient for transitions from the 1s ground state of a shallow-donor impurity in a low electric field can be taken as approximately proportional to the wave function square of the final state at the origin, provided that

$$|\Phi_{E_f}(0)|\neq 0$$

comparisons with numerical calculations of $\alpha(\omega)$ from Eq. (7) (following sections) show that this approximation is quite good.

RESULTS

We calculate the strength function $|\Phi_E(0)|^2$ with the new definition of the crossover x_0. The results are given in Fig. 2 for electric fields 0.05, 0.1, and 0.2. This figure shows that two improvements in the behavior of the strength function occur. First, the shift in the energy of the second peak is far less than with the DA definition of x_0. Second, the excitonic peaks are much less broadened. The second peak, shown in Fig. 2, remains observable for ε even as large as 0.1.

These results appear to be much more reasonable than those of DA (Ref. 9) in view of the experimental data on excitons and impurity states. For example, electroabsorption experiments indicate only modest broadening and small energy shifts of the exciton states of CdS (Ref. 19) for fields up to 10^5 V/cm, which corresponds to $\varepsilon=0.6$ for electrons and $\varepsilon=0.05$ for holes. Numerous excited state exciton peaks were observed by Grossman[21,22] for Cu_2O for fields up to 2×10^4 V/cm. Furthermore, electric field induced fine structure in the absorption at the indirect edge in Si (Ref. 23) also suggest that the exciton states do not broaden with increasing electric field as drastically as indicated by the DA crossover model.

In the case of impurities, Kopylov's[15] electroabsorption experiment on shallow donor states in GaP:S shows a $1s\rightarrow2p^{\mp}$ transition for fields on the order of 10^4 V/cm (corresponding to $\varepsilon=0.06$, $\mu^{-1}=m_\perp^{-1}+m_\parallel^{-1}$). Also, studies of the electric field dependence of the photoconductivity[24] show that excited states can be seen in Si at fields as large as 10^4 V/cm corresponding to $\varepsilon=0.1$.

We next discuss the results of our calculations of impurity absorption in electric field including the transition-matrix elements. Previous calculations in the literature do not include the actual transition-matrix elements or the effect of broadening.

In order to calculate the absorption coefficient as a function of photon energy, we first evaluate the transition probability per unit time from an initial energy state to a final energy state for a fixed photon energy $\hbar\omega$, by using Eq. (6). This transition probability is proportional to the absolute square of the transition-matrix elements. In the calculation of the transition-matrix elements an initial-state energy E_i is chosen and the final energy state is determined by the fixed photon energy, $E_f=E_i+\hbar\omega$. In the absence of an electric field, the transition probability from a 1s to a $2p^0$ state is a δ function in energy. Since the presence of an electric field broadens the 1s state ($E_{1s}=0.5$) into a band, the transition probabilities can not be neglected below or above (Fig. 3). Therefore the transition probabilities from every energy of the broadened 1s band to higher broadened bands must be calculated.

From our transition probability calculations (Figs. 4 and 5) we show how the bands broaden (the range of the initial energy) and how the peaks decrease in magnitude with increasing strength of the electric field.

In order to obtain the actual absorption spectrum, integrations are performed. The transition probabilities

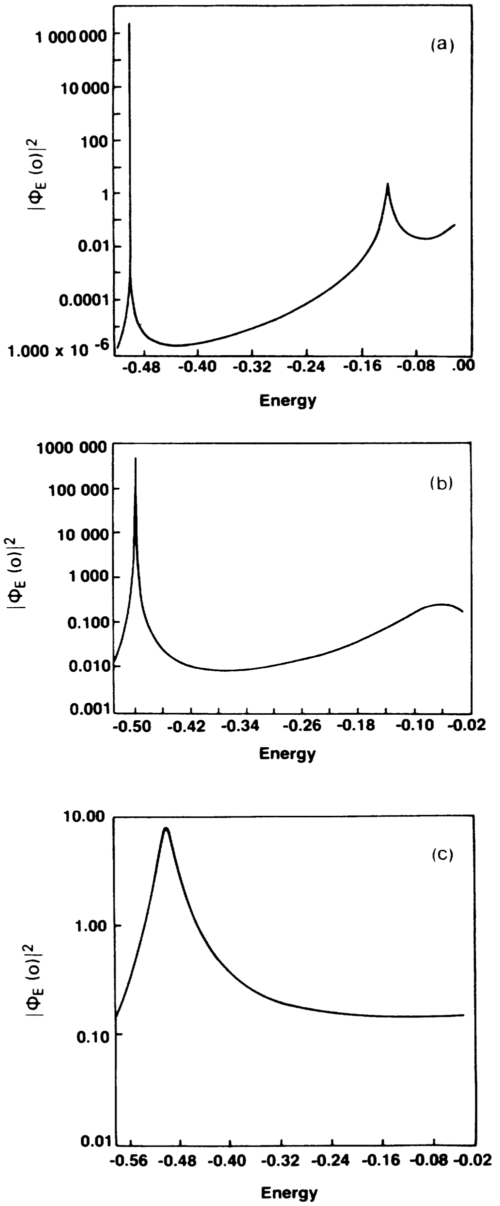

FIG. 2. DA strength function associated with allowed transitions at energies $E=-(E_g-\hbar\omega)/E_0$ near the first and second exciton peaks. The calculations are performed using the new crossover for the potential for electric fields: (a) $\varepsilon=0.05$, (b) $\varepsilon=0.1$, (c) $\varepsilon=0.2$.

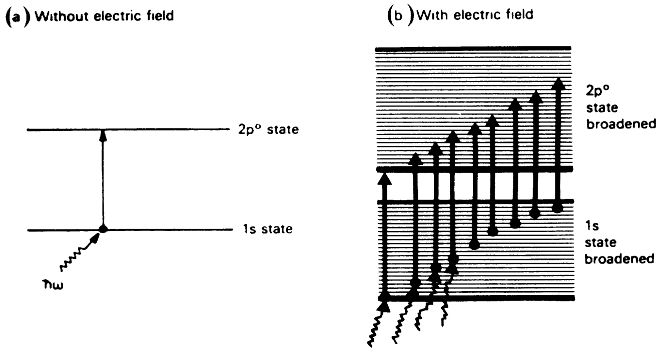

FIG. 3. 1s state and $2p^0$ state (a) without applied electric field and (b) with applied electric field. Excitation of an electron by a photon, with energy $\hbar\omega$, is shown schematically for both cases.

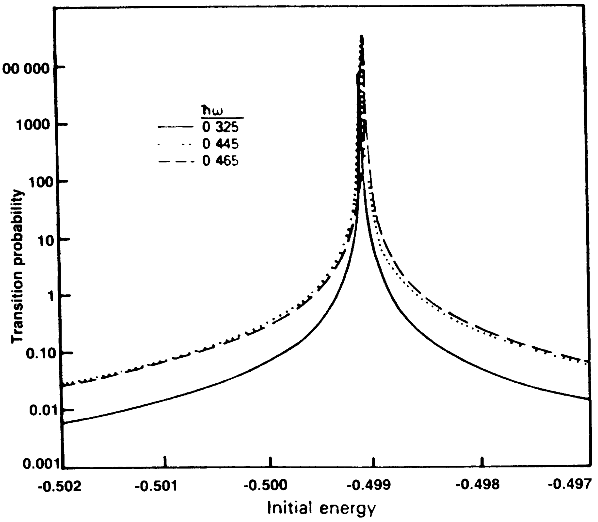

FIG. 4. Transition probability per unit time from 1s band to $2p^0$ band as a function of initial energies for three different photon energies for electric field $\varepsilon=0.05$ at energies $E=(\hbar\omega-E_g)/E_0$. Logarithmic scale is used for the transition probability axis. In these calculations the DA model is employed.

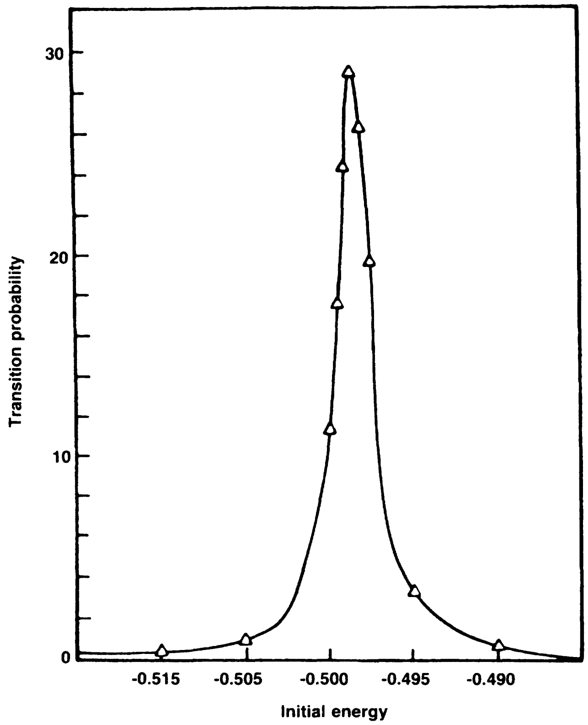

FIG. 5. Transition probability per unit time from 1s band to $2p^0$ band as a function of initial energies for photon energy $\hbar\omega=0.4$ and for electric field $\varepsilon=0.1$ at energies $E=(\hbar\omega-E_g)/E_0$. In these calculations the DA model is employed.

(which are functions of initial energies for a fixed photon energy) are integrated over all possible initial energies to obtain the absorption coefficient for that photon energy. This process is repeated for all photon energy values to obtain the absorption spectrum.

The results for the absorption spectrum for the $1s\rightarrow 2p^0$ band transitions are shown in Fig. 6. For these results, the definition given by DA for the crossover of the Coulomb and electric field potentials has been employed. Figure 6 shows the results for the electric fields equal to 0.04, 0.05, and 0.1. It is of some interest to compare these curves with the available experimental data to determine whether the DA model is applicable to shallow donor impurities or not.

In light of the experimental papers, Stark effect calculations, and numerical calculations on excitons and impurity levels, we expect to see peaks that are slightly broadened and shifted for low electric fields, i.e., $\varepsilon=0.03$ (552 V/cm for GaAs). At moderate electric fields, an increase in the broadening and shifting should occur with increasing electric field. At large electric fields, field ionization occurs and the absorption peak should disappear. All these features can be seen in Fig. 6. However, the question of whether the quantitative effects are in agreement with experiment needs further examination.

Comparison of these absorption curves in Fig. 6 with the available experimental data (Vrehen,[16] White,[13] Carter,[14] and Kopylov[15]) indicate that Duke's method, when applied to shallow donor impurities, greatly overestimates the effect of the electric field just as it does for excitons.

In order to determine if our definition of the crossover has improved the situation, we have repeated the calculations using the new crossover and including the transition-matrix elements as well. Our crossover for the potential makes the excited states less sensitive to the electric field; therefore, the broadening and shifts are reduced in the absorption spectrum. The absorption coefficient versus photon energy for the 1s-band–to–$2p^0$-band transitions are shown in Fig. 7. The effect of changing the crossover can be seen by comparing Figs. 6 (DA model) and 7 (our model). In both cases the $2p^0$ band shifted to higher-energy values; however, the shift in DA's case for the broadened $2p^0$ band far too large. The level is at $E=-0.07$ for $\varepsilon=0.05$ compared with $E=-0.125$ for $\varepsilon=0$. In our case, the level is at $E=-0.123$ for $\varepsilon=0.05$. The large $2p^0$-band shift in DA's case is directly related to the location of the crossover.

The absorption coefficient is also calculated by assuming that it is proportional to a constant times the square of the final-state wave function at the origin [Eq. (9)], just as in the case of excitons. The result is shown in Fig. 8. We have compared this figure with Fig. 6(b) (which is obtained by including the transition-matrix elements for the same electric field, $\varepsilon=0.05$). The shape of the curve and the energy of the excited state are essentially the same in both figures. This similar behavior shows that for low electric fields, the absorption coefficient formula for excitons can be used also for donor impurities. This approximation can not be applied to $m\neq 0$ final states because these wave functions vanish at the origin. The approximation which gives the absorption coefficient as a function of final energy can be used as a very good check for the complicated calculations of the absorption coefficient.

DISCUSSION AND CONCLUSION

In this paper we have examined the effects of an electric field on the energy levels and the absorption spectrum of both excitons and impurities using a modified version of the model proposed by Duke and Alferieff.[9] We have calculated the absorption spectrum including the actual transition-matrix elements. Such calculations have not been previously reported in the literature.

It is shown that with the DA model[9] the peak positions and the peak strengths are very sensitive to the way in which the crossover for the potential is defined. By changing the location of the crossover for the potential, the peak shifts and widths can be changed substantially. Consequently, the location of the shape of the peaks can not be reliably predicted unless some physically meaningful criterion can be found for selecting the location of the crossover for the potential in a precise fashion. Another problem with the DA model is the restriction placed on the quantum numbers employed in calculating the crossover boundary. The principal quantum number n is set to 1 and the parabolic quantum numbers n_1 and n_2 are set to zero. This choice leads to a model which gives reasonable results only for the 1s ground state.

We have generalized the DA model by introducing a different definition of the crossover boundary. Using this new definition we have calculated the absorption spectrum for both excitons and impurity states. The results (Figs. 2 and 7) more closely resembled the experimental data.[13–19] In particular, we have found much sharper peaks in the absorption spectrum, and the shift of the peak energy with increasing electric field was much smaller than that produced by the DA model.

We have shown that Elliot's[25] approximation for exci-

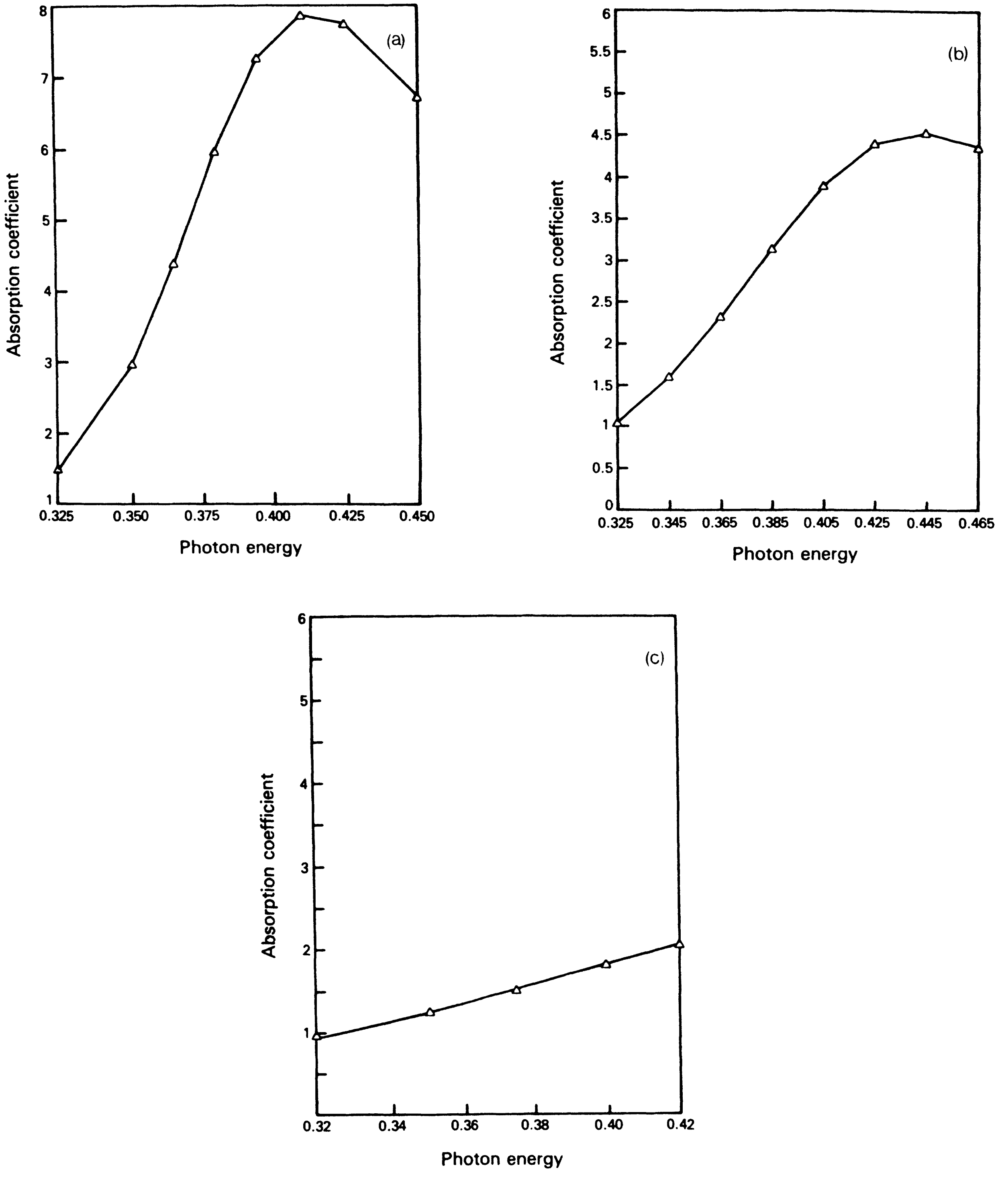

FIG. 6. Calculated absorption coefficient vs photon energy (in atomic units) from 1*s* to $2p^0$ band for electric fields (a) $\varepsilon=0.04$, (b) $\varepsilon=0.05$, (c) $\varepsilon=0.1$. In these calculations the transition-matrix elements are included and DA's definition for the crossover for the potential is used.

tons (absorption coefficient is proportional to the absolute square of the wave function at the origin) can also be applied to shallow donor impurities at low electric fields (Fig. 8). It is shown that this approximation agrees well with the full calculation of the absorption coefficient including transitions from the ground states to states with $m=0$.

The transition probability calculations reveal how the bands are broadened and shifted with increasing electric field. The transition probability curves also indicate at what electric field values the electrons in shallow donor states are field ionized. The absorption-coefficient calculations show how the absorption edge changes with electric field, and the amount of photon energy needed to excite an electron from the broadened 1s band to higher bands or the continuum.

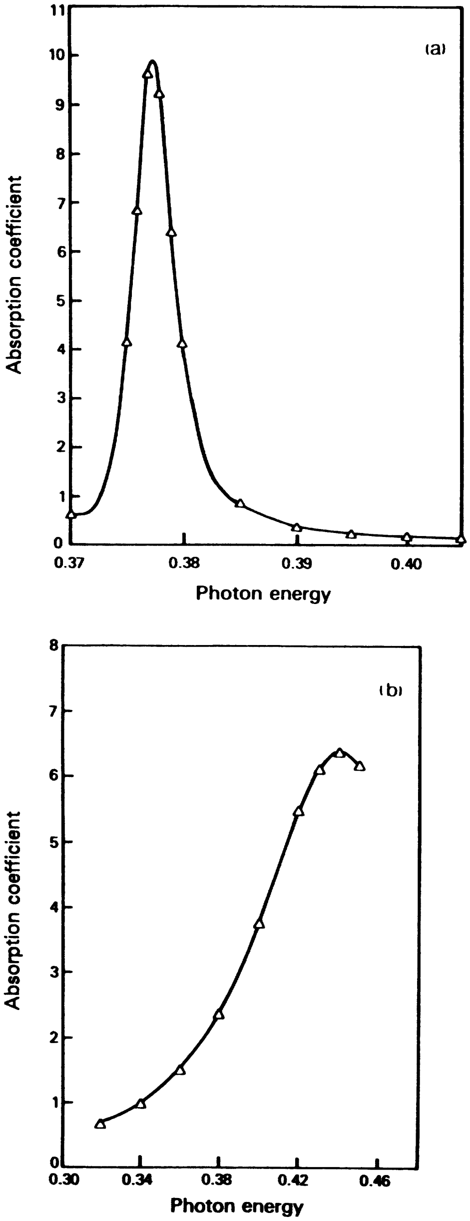

FIG. 7. Calculated absorption coefficient vs photon energy (in atomic units) from 1s to $2p^0$ band for electric fields (a) $\varepsilon=0.05$ and (b) $\varepsilon=0.1$. In these calculations these transition-matrix elements are included and the new choice of the crossover for the potential is used.

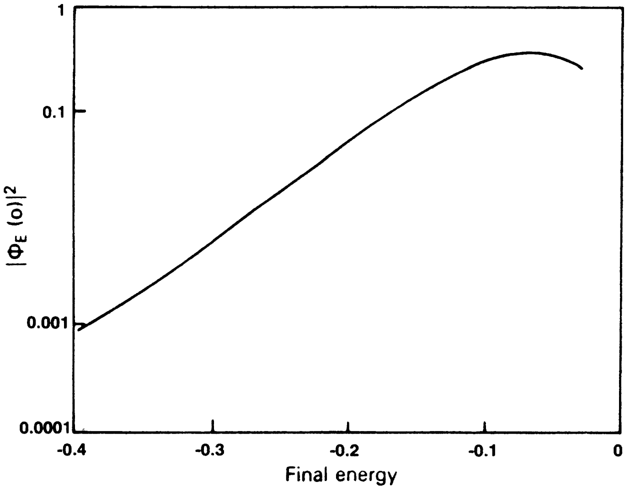

FIG. 8. Calculated absorption coefficient [by using the $|\Phi_E(0)|^2$ approximation for impurities, Eq. (9)] vs final energy from 1s to $2p^0$ band for electric field $\varepsilon=0.05$ at energies $E=(\hbar\omega-E_g)/E_0$. DA's definition for the crossover is used.

Due to the lack of experimental data, we were not able to compare our results directly with experimental absorption data on shallow donor impurities. The only experimental data for comparison with our results are those due to Carter[14] on the photoconductivity of shallow donor impurities, White's[13] results on absorption by impurity acceptors in an electric field, and Kopylov's[15] electroabsorption results on GaP:S.

The photoconductivity experiments measure the conductivity by exciting electrons from impurity levels into the conduction band. These experiments show that as the electric field increases the energy of the conductivity peaks moves towards lower photon energies. This shift towards lower energies means that the excited states move closer to the conduction band under the influence of the electric field. In our calculations, we observed the same type of shift. The excited states move towards higher energies with increasing electric field. The ground state is not shifted as much as the excited states. Therefore, as the electric field increases, the energy difference between the broadened 1s band and the broadened $2p^0$ band increases as well. Consequently, the $1s\rightarrow 2p^0$ transition energy increases with increasing electric field.

Comparisons with related experiments show clearly that with our choice of the crossover boundary, the model predicts the correct electric field effects. We have presented results for $1s\rightarrow 2p^0$ transitions. The model can be employed for transitions from 1s or excited levels to higher excited levels depending on the strength of the electric field. The question of whether there is a more "physically meaningful way" to select a criterion for the crossover is not answered. However, one might use experimental data to find a reasonable choice for the crossover boundary. Therefore we conclude by saying that what is needed are careful absorption measurements on donor impurity states at high electric fields.

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