Intermediate excitonic state in interimpurity radiative recombination

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The influence of the Coulomb interaction between an electron and a hole on the process of interimpurity radiative recombination is investigated within the framework of the model of donor-acceptor pairs. The method of calculating the recombination probability is based on obtaining an asymptotically exact expression for the electron-hole wave function that describes the state of the pair. It is shown that the intermediate excitonic state produced upon recombination greatly increases the recombination probability of remote impurities and influences the shapes of the recombination-radiation spectral lines.

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Modern theory of interimpurity radiative recombination is based on the concept of donor-acceptor pairs, as developed in the papers by Williams.[1,2] The Heitler-London (HL) method was used[1-3] to calculate the dependence of the energy $\hbar\omega$ of the recombination quanta on the distance between the donor and the acceptor. The calculated positions of the peaks in the recombination-radiation spectra are in good agreement with the experiment.[2-4]

Calculations of the matrix elements of radiative interimpurity recombination[2,3] have shown that recombination takes place mainly in the region located between the donor and acceptor near the axis that joins their centers (interimpurity axis). In the present paper it is shown that in this region the wave functions of the donor and hole cannot be calculated in the HL approximation, but must be determined with account taken of the distortions due to the Coulomb attraction of the recombining electron and hole. Allowance for the correct asymptotic behavior of the electron-hole wave function alters the recombination probability and the form of the frequency dependence of the emission spectrum.

The wave function is calculated in two steps. The wave function of the donor-acceptor pair at sufficiently large distances between the electron and the hole is determined by the method proposed by Gor'kov and Pitaevskiĭ[5] and by Herring and Flicker[6] to calculate the H_2 molecule. This function takes into account the Coulomb attraction between the electron and the hole and their interaction with the "foreign" impurity centers.

At short distances between the electron and the hole, in constract to Refs. 5 and 6, their relative motion is determined by the Coulomb attraction, which leads to formation of an intermediate excitonic state. The parameters of the wave function of this intermediate excitonic state are determined by joining together the exciton solution and the donor-acceptor-pair wave function obtained during the first stage.

It has been shown that allowance for the intermediate excitonic state alters the magnitude and the analytic form of the recombination probability as functions of the interimpurity distance. Depending on the initial conditions, the recombination can have a resonant character when the energy of the electron and hole in the intermediate excitonic state coincides with the binding energy of the free exciton in the given semiconductor.

The article considers the simplest case of phononless recombination in a straight-band semiconductor under the assumption that the effective masses of the electron and hole are isotropic and are close in magnitude.

1. WAVE FUNCTION OF DONOR-ACCEPTOR PAIR AT LARGE DISTANCES BETWEEN THE ELECTRON AND HOLE

In the effective-mass approximation, the Hamiltonian for the determination of the envelope $\Psi(\mathbf{r}_e, \mathbf{r}_h)$ of the Bloch functions of the electron and hole in the donor-acceptor pair is of the form[1,2]

$$\hat{H} = -\frac{\hbar^2}{2m_e}\Delta_e - \frac{\hbar^2}{2m_h}\Delta_h - \frac{e^2}{\varepsilon|\mathbf{r}_e+\mathbf{a}/2|} - \frac{e^2}{\varepsilon|\mathbf{r}_h-\mathbf{a}/2|} + V(\mathbf{r}_e, \mathbf{r}_h), \tag{1}$$

where

$$V(\mathbf{r}_e, \mathbf{r}_h) = \frac{e^2}{\varepsilon|\mathbf{r}_e-\mathbf{a}/2|} + \frac{e^2}{\varepsilon|\mathbf{r}_h+\mathbf{a}/2|} - \frac{e^2}{\varepsilon a} - \frac{e^2}{\varepsilon|\mathbf{r}_h-\mathbf{r}_e|}. \tag{2}$$

In (1) and (2), $\mathbf{r}_{e,h}$ are the coordinates of the electron and hole reckoned from the midpoint of the interimpurity axis a; $m_{e,h}$ are the effective masses of the electron and hole; ε is the dielectric constant of the medium; a is the distance between the donor and acceptor.

The Schrödinger equation for $\Psi(\mathbf{r}_e, \mathbf{r}_h)$ is

$$\hat{H}\Psi(\mathbf{r}_e, \mathbf{r}_h) = (E_e+E_h)\Psi(\mathbf{r}_e, \mathbf{r}_h), \tag{3}$$

where $E_{e,h} = -m_{e,h}e^4/2\hbar^2\varepsilon^2$ are the ground-state energies of the isolated donor and acceptor. In the considered case $a \gg b_{e,h}$ ($b_{e,h} = \hbar^2\varepsilon/m_{e,h}e^2$ are the Bohr radii of the impurities) the correction term to the pair energy, which takes into account the Van der Waals attraction of the impurities, has been left out, since it is of sixth order of smallness in the parameter $(b_{e,h}/a) \ll 1$. We seek the solution of (3) in the form[5,6]

$$\Psi(\mathbf{r}_e, \mathbf{r}_h) = \chi(\mathbf{r}_e, \mathbf{r}_h) F_c(\mathbf{r}_e) F_v(\mathbf{r}_h), \tag{4}$$

$$F_c(\mathbf{r}_e) = (\pi b_e^3)^{-1/2} \exp\left[-\frac{|\mathbf{r}_e+\mathbf{a}/2|}{b_e}\right], \tag{5}$$

$$F_v(\mathbf{r}_h) = (\pi b_h^3)^{-1/2} \exp\left[-\frac{|\mathbf{r}_h-\mathbf{a}/2|}{b_h}\right]. \tag{6}$$

Here $F_c(\mathbf{r}_e)$ and $F_v(\mathbf{r}_h)$ are the wave functions of the isolated donor and acceptor impurities; χ is a smooth function that takes into account the distortion due to the interaction (2). We recall that $\chi(\mathbf{r}_e, \mathbf{r}_h) \equiv 1$ in the HL method.

Substituting (4)–(6) in (3), we obtain an equation for $\chi(\mathbf{r}_e, \mathbf{r}_h)$:

$$\left(-\frac{\hbar^2}{2m_e}\Delta_e - \frac{\hbar^2}{2m_h}\Delta_h\right)\chi + \frac{e^2}{\varepsilon}(\varkappa_e\nabla_e + \varkappa_h\nabla_h)\chi + V(\mathbf{r}_e, \mathbf{r}_h)\chi = 0, \tag{7}$$

where $\varkappa_{e,h}$ are unit vectors of the directions from the donor and acceptor centers to the electron and hole respectively.

Since the characteristic distances for χ are of the order of $a \gg b_{e,h}$,[5] we neglect in (7) the terms containing the electron and hole kinetic-energy operators. This is permissible so long as the distance between the electron and hole $r = |\mathbf{r}_h - \mathbf{r}_e|$ is large enough (as will be shown below, larger than the Bohr radius of the free exciton in the semiconductor). Since the coordinate region of importance in recombination is located near the interimpurity axis (the region of the effective overlap of F_c and F_v, Ref. 2), it follows that, by retaining in (7) the terms of lower order in $b_{e,h}/a \ll 1$, we obtain near the axis

$$\left(\frac{\partial}{\partial z_e} - \frac{\partial}{\partial z_h}\right)\chi + \left[\frac{1}{a/2 - z_e} + \frac{1}{a/2 + z_h} - \frac{1}{a} - \frac{1}{[(z_h - z_e)^2 + \rho^2]^{1/2}}\right]\chi = 0, \tag{8}$$

where $z_{e,h}$ is the projection of $\mathbf{r}_{e,h}$ on the $\mathbf{a}$ axis; $\rho = [r^2 - (z_h - z_e)^2]^{1/2}$ enters in (8) as a parameter.[5] The boundary conditions needed for the solution of (8) take the simple form

$$\chi(\mathbf{r}_e, \mathbf{r}_h) \to 1\,. \tag{9}$$

If $z_e \to -a/2$ or $z_h \to a/2$. This means that $\chi = 1$ when one of the carriers is located directly at its impurity center, and the distance between the electrons and hole is large enough.

Equation (8) differs from the equation for the two-electron function $\tilde{\chi}(\mathbf{r}_1, \mathbf{r}_2)$ ($\mathbf{r}_{1,2}$ are the coordinates of the electrons) in the H_2 molecule[5] in that the signs of all the terms of the perturbing potential (2) are different. Consequently, χ and $\tilde{\chi}$ are connected by the simple relation $\chi = 1/\tilde{\chi}$. Thus, the solution of (8) and (9) can be obtained by using the previously known solution at $\tilde{\chi}$ for H_2[5];

$$\chi = \frac{(a - 2z_e)(a + 2z_h)}{4a(a - z_e - z_h)}\left[\frac{((a - z_e - z_h)^2 + \rho^2)^{1/2} + a - z_e - z_h}{((z_h - z_e)^2 + \rho^2)^{1/2} + z_h - z_e}\right]^{1/2}\exp\left(\frac{1}{2} - \frac{z_h}{a}\right), \quad (z_h + z_e) \geqslant 0, \tag{10}$$

$$\chi = \frac{(a - 2z_e)(a + 2z_h)}{4a(a + z_e + z_h)}\left[\frac{((a + z_h + z_e)^2 + \rho^2)^{1/2} + a + z_e + z_h}{((z_h - z_e)^2 + \rho^2)^{1/2} + z_h - z_e}\right]^{1/2}\exp\left(\frac{1}{2} + \frac{z_e}{a}\right), \quad (z_h + z_e) \leqslant 0.$$

The function (10) does not contain as parameters the Bohr radii $b_{e,h}$ of the impurities (the effective masses $m_{e,h}$), since it takes into account only the distortion of the potential barriers by the interaction (2).

To investigate the recombination process it is necessary to know the behavior of the wave function $\Psi(\mathbf{r}_e, \mathbf{r}_h)$ at short distances between the electron and the hole. (By short are meant distances $\lesssim b_{e,h}$.) It follows from (10) that $\chi \to \infty$ as $r = |\mathbf{r}_h - \mathbf{r}_e| \to 0 (\chi \sim 1/r^{1/2})$. This divergence corresponds to the falling of the electrons on the force center (hole) and is due to discarding the kinetic energy of the relative motion of the electron and hole in (8). We note that for the H_2 molecule[5,6] no such divergence arises:

$$\chi = \tilde{\chi}^{-1}(\mathbf{r}_1, \mathbf{r}_2) \sim r_{21}^{1/2} \to 0 \tag{11}$$

as $r_{21} = |\mathbf{r}_2 - \mathbf{r}_1| \to 0$ because of the Coulomb repulsion between the electrons.

It follows from the foregoing that in the region of small relative distances between the electron and hole it is necessary to solve the exact exciton problem, with account taken of both Coulomb attraction between the electron and hole and the kinetic energy of their relative motion. This exciton solution in the limit of large r should go over into the previously obtained solution $\Psi(\mathbf{r}_e, \mathbf{r}_h)$ (4), (10).

2. INTERMEDIATE EXCITONIC STATE

We introduce the coordinates of the mass center $\mathbf{R}$ and of the relative motion $\mathbf{r}$ of the electron-hole system (the intermediate exciton). To determine the explicit form of the wave function of the intermediate excitonic state $\Psi(\mathbf{r}, \mathbf{R})$ at small $\mathbf{r}$ we take into account in the effective Hamiltonian (1), (2) only the potential of the Coulomb attraction between the electron and the hole. The largest of the discarded terms is here the potential of the interaction of the dipole moment of the intermediate exciton $e\mathbf{r}$ with the electric field $\mathbf{E}$ produced by the impurity centers: $U = -e\mathbf{r}\cdot\mathbf{E}$.

The order of magnitude of the discarded terms can be estimated at

$$\left|U\Big/\frac{e^2}{\varepsilon r}\right| \sim r^2\Big/\left|\mathbf{R} \pm \frac{\mathbf{a}}{2}\right|^2 \ll 1, \tag{12}$$

if the mass center $\mathbf{R}$ of the exciton is far enough from the impurities in the region characteristic of the recombination. The last statement is valid in the case of close effective masses:[1]

$$|\Delta m| \ll M (\Delta m = m_h - m_e,\ M = m_h + m_e).$$

In the approximation (12) the Schrödinger equation (3) expressed in terms of the variables $\mathbf{R}$ and $\mathbf{r}$ breaks up into equations for the wave functions of the mass center and of the relative motion:

$$\varkappa = -\frac{\hbar^2}{2M}\frac{\Delta_{\mathbf{R}}\varphi_\varkappa(\mathbf{R})}{\varphi_\varkappa(\mathbf{R})}, \tag{13}$$

$$\left(-\frac{\hbar^2}{2\mu}\Delta_r - \frac{e^2}{\varepsilon r}\right)\psi_{E-\varkappa}(\mathbf{r}) = (E - \varkappa)\psi_{E-\varkappa}(\mathbf{r}), \tag{14}$$

$$\Psi(\mathbf{r}, \mathbf{R}) = \varphi_\varkappa(\mathbf{R})\psi_{E-\varkappa}(\mathbf{r}), \tag{15}$$

where $\varphi_\varkappa(\mathbf{R})$ and $\varkappa$ are the wave function and energy of motion of the mass center; $\Psi_{E-\varkappa}(\mathbf{r})$ is the wave function of the relative motion of the electron and hole, and μ is the reduced mass; $E = E_e + E_h$ is the total energy of the intermediate excitonic state. In (14) it is necessary to consider only the s state of the relative motion (orbital angular momentum $l = 0$), since all the states with $l \neq 0$ have a node at $r = 0$ and therefore make no contribution to the recombination process.

The wave function satisfying (14) and bounded at $r = 0$ is of the form

$$\psi_{E-\varkappa}(\mathbf{r}) = \exp(-n_\varkappa r)\,{}_1F_1\left(1 - \frac{\mu e^2}{\varepsilon\hbar^2 n_\varkappa}, 2, 2n_\varkappa r\right)Y_{00}, \tag{16}$$

$$n_\varkappa=[-2\mu\hbar^{-2}(E-\varkappa)]^{1/2}, \tag{17}$$

where ${}_1F_1$ is the solution of the confluent hypergeometric equation that satisfies the required conditions,[7] $Y_{00}=(4\pi)^{-1/2}$ is the angular part of the wave function of the s state. By substituting (16) in (15) we obtain the wave function of the s state of the intermediate exciton:

$$\Psi_s(\mathbf{r},\mathbf{R})=\varphi_\varkappa(\mathbf{R})\exp(-n_\varkappa r)\,{}_1F_1\left(1-\frac{1}{n_\varkappa b},2,2n_\varkappa r\right)Y_{00}, \tag{18}$$

where $b=\hbar^2\varepsilon/\mu e^2$ is the Bohr radius of the free exciton.

The behavior of (18) in the region of the relative distances

$$n_\varkappa^{-1}\ll r\ll|R\pm a/2| \tag{19}$$

is obtained by using the asymptotic representation of ${}_1F_1$ at large orders of the argument $r\gg n^{-1}$ (Ref. 7),

$$\Psi_s(\mathbf{r},\mathbf{R})=\frac{\varphi_\varkappa(\mathbf{R})}{\pi^{1/2}\Gamma(1-1/n_\varkappa b)}\frac{\exp(n_\varkappa r)}{(2n_\varkappa r)^{1+1/n_\varkappa b}}. \tag{20}$$

In the region (19), the exciton solution $\Psi_s(\mathbf{r},\mathbf{R})$ (20) should coincide with the solution $\Psi(\mathbf{r}_e,\mathbf{r}_h)$ (4), (10) for the donor-acceptor pair, in which only the part corresponding to the s state of the relative motion of the electron and hole is considered. To separate in the wave function (4), (10) the s state it suffices to average $\Psi(\mathbf{r}_e,\mathbf{r}_h)$ over the solid angle of the space of the vectors $\mathbf{r}$:

$$\Psi_s(\mathbf{r},\mathbf{R})=\frac{1}{4\pi}\int_{4\pi}\chi(\mathbf{r}_e,\mathbf{r}_h)F_c(\mathbf{r}_e)F_v(\mathbf{r}_h)\sin\vartheta\, d\vartheta\, d\varphi, \tag{21}$$

where ϑ and φ are the polar and azimuthal angles of the vector r (the polar axis coincides with the **a** direction).

To integrate in (21), we write down the integrand expression in the coordinates R and r. Taking (12) into account, we obtain for the product of the envelopes

$$F_c(\mathbf{r}_e)F_v(\mathbf{r}_h)=F_c(\mathbf{R})F_v(\mathbf{R})\exp\left(\frac{2r}{b}\cos\vartheta\right)\left[1+O\left(\frac{r}{|\mathbf{R}\pm a/2|}\right)\right]. \tag{22}$$

We recall once more that $F_c(\mathrm{R})$ and $F_v(\mathrm{R})$ in (22) should be considered near the interimpurity axis, in analogy with Ref. 5. We will show below [see Eq. (27)] that $n_\varkappa^{-1}=b/2$. Consequently, in the entire interval (19) the argument of the exponential $2r/b\gg1$. The significant contribution to the integral in (21) is then made only by small angles $\vartheta\lesssim(b/r)^{1/2}$.

At small $\vartheta\to0$, the function $\chi(\mathbf{r}_e,\mathbf{r}_h)$ takes the form

$$\chi(\mathbf{r}_e,\mathbf{r}_h)=\chi_1(\mathbf{R})\chi_2(\mathbf{r})\left[1+O\left(\frac{r}{|\mathbf{R}\pm\mathbf{a}/2|}\right)\right], \tag{23}$$

where

$$\chi_1(\mathbf{R})=\left[1-\frac{2|Z|}{a}\right]^{1/2}\left[1+\frac{2|Z|}{a}\right]\exp\left[-\frac{|Z|}{a}\right], \tag{24}$$

$$\chi_2(\mathbf{r})={}^1/_4e^{1/2}(a/r)^{1/2}, \tag{25}$$

and Z is the projection of R on the interimpurity axis.

Substituting (22)–(25) in (21) and integrating over the angles, we obtain

$$\frac{\varphi_\varkappa(\mathbf{R})}{\pi^{1/2}\Gamma(1-1/n_\varkappa b)}\frac{\exp(n_\varkappa r)}{(2n_\varkappa r)^{1+1/n_\varkappa b}}=\frac{e^{1/2}}{2}\left(\frac{a}{b}\right)^{1/2}\chi_1(\mathbf{R})F_c(\mathbf{R})F_v(\mathbf{R})\frac{e^{2r/b}}{(4r/b)^{1/2}}. \tag{26}$$

Comparing in (26) the wave functions of the relative motion of the electron and hole, we obtain the exclusive form of $n_\varkappa$:

$$n_\varkappa=[-2\mu h^{-2}(E-\varkappa)]^{1/2}=2/b. \tag{27}$$

We now obtain the energy of the relative motion:

$$(E-\varkappa)=-\frac{h^2}{2\mu}n_\varkappa^2=-\frac{2\mu e^4}{h^2\varepsilon^2} \tag{28}$$

and the energy of the motion of the mass center of the intermediate exciton:

$$\varkappa=-\frac{(\Delta m)^2}{2M}\frac{e^4}{h^2\varepsilon^2}. \tag{29}$$

The wave function of the motion of the center of mass near the interimpurity axis is, according to (26),

$$\varphi_\varkappa(\mathbf{R})=\frac{\pi e^{1/2}}{2}\left(\frac{a}{b}\right)^{1/2}\chi_1(\mathbf{R})F_c(\mathbf{R})F_v(\mathbf{R}). \tag{30}$$

By direct substitution we can verify that (30) satisfies Eq. (13) with energy $\varkappa$ (29). Substituting (30) in (18), we obtain the final expression for the wave function of the s state of the intermediate exciton:

$$\Psi_s(\mathbf{r},\mathbf{R})=\frac{(\pi e)^{1/2}}{4}\left(\frac{a}{b}\right)^{1/2}\chi_1(\mathbf{R})F_c(\mathbf{R})F_v(\mathbf{R})e^{-2r/b}{}_1F_1\left(\frac{1}{2},2,\frac{4r}{b}\right). \tag{31}$$

It follows from (16) that the initial states of the donor-acceptor pair, at which $(n_\varkappa b)^{-1}=k$, $k=1,2,3,\ldots$, are at resonance with the k-th level of the free exciton. One of the consequences of this resonance may be the appreciable enhancement of the probability of the radiative recombination of impurities that are in excited states. On the other hand, if the impurities are in their ground states, then the energy of the relative motion $E-\varkappa$ from (28) in the intermediate exciton is four times larger in absolute magnitude than the self-energy of the ground state of the exciton and, naturally, there is no resonance.

3. INFLUENCE OF INTERMEDIATE EXCITON ON THE PROCESS OF RADIATIVE RECOMBINATION

The probability of radiative recombination of a donor-acceptor pair in which the impurities are seperate by a distance a is equal to

$$W(a)=\frac{4e^2\omega(a)n}{3\hbar c^3m^2}|\langle c|\mathbf{p}|v\rangle|^2J^2(a), \tag{32}$$

where m is the electron mass, c is the speed of light,

$$\omega(a)=\frac{1}{\hbar}\left[E_g-|E_e+E_h|+\frac{e^2}{\varepsilon a}\right]$$

is the frequency of the interimpurity recombination radiation, $\langle c|\mathbf{p}|v\rangle$ is the matrix element of the transitions from the valence to the conduction band, and n is the refractive index (n is approximately constant for the interband-recombination frequency band);

$$J(a)=\int\Psi_s(\mathbf{r},\mathbf{R})\delta(\mathbf{r})d^3rd^3R=\int\Psi_s(0,\mathbf{R})d^3R. \tag{33}$$

Substituting $r=0$ in (31) we get

$$\Psi_s(0,\ \mathbf{R})={}^1/_4(\pi e)^{1/2}(a/b)^{1/2}\chi_1(\mathbf{R})F_c(\mathbf{R})F_v(\mathbf{R}), \tag{34}$$

where $F_{e,v}(\mathrm{R})$ are the envelopes for the isolated donor and acceptor, and $\chi_1(\mathrm{R})$ is given by (24). Consequently

$$J(a)=\frac{(\pi e)^{1/2}}{4}\left(\frac{a}{b}\right)^{1/2}\int\chi_1(\mathbf{R})F_c(\mathbf{R})F_v(\mathbf{R})d^3R. \tag{35}$$

Calculating (35) for the case $m_e=m_h$, we obtain the following expression for the recombination probability:

$$W(a)\approx 2{,}8\cdot 10^{-2}\frac{e^2\omega(a)n|\langle c|\mathbf{p}|v\rangle|^2}{\hbar c^3 m^2}\left(\frac{a}{b_c}\right)^5\exp\left[-\frac{2a}{b_e}\right]. \quad (36)$$

In the calculation of the probability of the radiative recombination by the HL method it is necessary to replace $J(a)$ in (32) by the overlap integrals F_c and F_v (Refs. 2, 3):

$$J_{HL}(a)=\int F_c(\mathbf{R})F_v(\mathbf{R})\,d^3R. \quad (37)$$

Since the HL method does not take into account the intermediate excitonic state, a comparison of the results obtained in the present article with the calculations based on the HL method reveals the following recombination-process singlarities due to formation of the intermediate exciton.

1. An analysis of the ratio of the recombination probabilities

$$\frac{W(a)}{W_{HL}(a)}=\frac{J^2(a)}{J_{HL}^2(a)}\sim\left(\frac{a}{b}\right)\gg 1 \quad (38)$$

shows that the intermediate exciton state increases the probability of the recombination of remote pairs ($a\gg b$).

2. The form of the spectral intensity band of the recombination radiation (more accurately, of its low-frequency part) changes when account is taken of the intermediate excitonic state, since the analytic form of the dependence of the recombination probability on a changes.

For the case of weakly doped semiconductors and sufficiently high rates of generation of neutral donor-acceptor pairs, the low-frequency edge of the recombination-radiation spectrum, obtained from (32) and (35) with the aid of Refs. 2 and 10 takes the form

$$I(\omega)\sim(\omega-\omega_0)^{-9}\exp\left[-\frac{2|E_e+E_h|}{\hbar(\omega-\omega_0)}\right], \quad (39)$$

where $\omega_0=\hbar^{-1}[E_g-|E_e+E_h|]$ is the lower limit of the frequency band of the interimpurity recombination radiation. Calculation by the HL method yields a different value of the pre-exponential factor in (39), namely $-(\omega-\omega_0)^{-8}$.

3. A possibility of a considerable increase of the probability of recombination of the excited impurities as a result of resonance with the exciton.

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[1)]For greatly differing effective masses ($|\Delta m|\sim M$) it follows from an analysis of the overlap of F_c and F_v that recombination occurs mainly near the deeper impurity, at distances $\lesssim\hbar^2\varepsilon/|\Delta m|e^2$ from its center. It is then necessary to take into account in the Hamiltonian of the excition problem the interaction of the electron and of the hole with the given impurity.

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