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ELECTRONIC AND OPTICAL PROPERTIES OF SEMICONDUCTORS

Properties of Shallow-Level *D*⁻-Centers in Polar Semiconductors

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Abstract—The energy of the lowest singlet and triplet terms of shallow-level D^- centers (two electrons bound at a singly charged Coulomb center) in semiconductors with ion bonding is theoretically analyzed. Electron–phonon interaction is described by the Fröhlich Hamiltonian. The D^- -center energy is calculated by the Buĭmistrov–Pekar method of canonical transformations for an arbitrary coupling force with phonons. It was shown that, for the entire range of electron–phonon interaction parameters, the Buĭmistrov–Pekar method gives the lowest values of the ground-state energy of the D^- centers and the free bipolaron in comparison with the best available numerical computations of these values which were performed using direct variational methods. The calculations showed the absence of both bound metastable triplet states, corresponding to the lowest triplet term of a D^- center, and a bipolaron for the entire range of parameters of electron–phonon interaction. This is consistent with the Hill theorem concerning the absence of bound excited states of an H⁻ ion. © 2003 MAIK "Nauka/Interperiodica".

INTRODUCTION

Two-electron D^- centers in semiconductors (in Si and Ge [1–3], in GaAs, InP, and InSb [4]) are observed when studying the frequency dependence of photoconductivity in the far-infrared region. An atomic analogue of the considered system is the negative ion of hydrogen, H^- , with an ionization energy equal to 0.0555 Ry. If chemical shift and electron-phonon interaction are disregarded, the D^- centers in crystal have the same energy but measured in the effective rydberg $Ry^* =$ $m^*e^4/2\epsilon_0^2\hbar^2$ (where m^* is the effective electron mass, and ε_0 is the static dielectric constant of the crystal). Taking the interaction with phonons into account can result in a significant increase in the binding energy of a two-electron system in crystal in comparison with the value of 0.0555 Ry* [5, 6]. In alkali halide crystals, the F centers are similar to a D^- center [7].

Recently, studies of the energy structure of twoelectron systems have been also extended to lowdimensional systems, including quantum dots [8, 9] and clusters [10, 11]. The energy levels of quasi-twodimensional systems, which are similar to the D^- centers [14–17] and bipolarons [18] in isotropic crystals, have been studied both experimentally [12, 13] and theoretically. This partly is connected with the fact that interest in similar systems has considerably increased recently in relation to the development of nanotechnologies and the possibility of developing quantum computers based on electron spin resonance [19–21] particularly in Ge–Si structures [21]. In the first studies [7, 22] devoted to the calculation of the energy of two-electron formations, electronic correlations were disregarded that considerably lowered the binding energy of a D^- center. Nevertheless, the Buĭmistrov–Pekar method [22] for calculating the energy of one- and two-electron states in crystals with an arbitrary coupling force of the electron system with phonons is, in our opinion, one of the simplest and most effective methods for calculating the energy spectrum of electron systems in the solid state.

Due to its simplicity, this method is used quite frequently for calculating the energy of specific systems for both one- and two-electronic states in crystals with an arbitrary coupling force of electrons with phonons [5, 23–25]. Nevertheless, it is generally believed that the Buĭmistrov-Pekar method yields less precise values of the energy of two-electron systems in comparison with the method of optimized canonical transformation suggested by Adamowski [6], which presently gives the lowest values for the energy of two-electron systems (bipolarons and D^- centers or bound bipolarons). We will show that with a fairly flexible trial electron wave function (WF), which accounts for interelectronic correlations (direct dependence of the WF on the electronelectron distance), the method of the Buĭmistrov-Pekar yields lower values than the Adamowski method for the entire range of parameters of electron-phonon interaction. The latter circumstance makes it possible to use the above method combined with a tested set of wave functions, which are convenient for analytical calculations, for reliable computation of the energy of two-electron systems (D⁻ centers, bipolarons, and exchange-coupled

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pairs of paramagnetic centers) in anisotropic crystals and in low-dimensional systems with an arbitrary constant of electron–phonon coupling. Previously, this set of functions [26] was tested during the calculation of the bipolaron energy in crystals with an anisotropic effective mass and dielectric constant under conditions of strong electron–phonon coupling.

2. BASIC RELATIONS

In what follows, we use the Feynman system of units in which $\hbar = 1$, $\omega = 1$, and $2m^* = 1$. Hence, $\hbar\omega$ is a unit of energy and $L_0 = \sqrt{\hbar/2m^*\omega_0}$ is a unit of length. The Hamiltonian of the D^- center (or bound bipolaron in [6]) can be written as

$$H = \sum_{\mathbf{k}} a_k^+ a_k + \frac{\beta}{r_{12}}$$
$$+ \sum_{j=1}^2 \left[-\nabla_j^2 - \frac{\gamma}{r_j} - \sum_{\mathbf{k}} \sqrt{\frac{4\pi\alpha}{V} \frac{1}{k}} (a_k - a_{-k}^+) \exp(i\mathbf{k}\mathbf{r}_j) \right], \tag{1}$$
$$\alpha = \frac{e^2}{2\hbar\omega} \left(\frac{1}{\varepsilon_{\infty}} - \frac{1}{\varepsilon_0} \right) \frac{1}{L_0}, \quad L_0 = \sqrt{\hbar/2m^*\omega_0},$$
$$\beta = \frac{e^2}{\hbar\omega\varepsilon_{\infty}L_0} = \frac{2\alpha}{1-\eta}, \quad \gamma = \frac{e^2}{\hbar\omega\varepsilon_0L_0} = \frac{2\alpha\eta}{1-\eta},$$

where \mathbf{r}_j is a radius vector of the *j*th electron with an effective mass m^* and $r_{12}2$ is the electron–electron distance. We assume that the Coulomb charge is concentrated at the origin. The effect of polarization of a crystal on the Coulomb field of a static charge is taken into account by introducing the static dielectric constant ε_0 . The optical dielectric constant ε_{∞} is included in the operator of the electron–electron interaction, and $a_k^+(a_k)$ is the creation (annihilation) operator of a longitudinal optical phonon with a wave vector \mathbf{k} . We assume that the phonon frequency is independent of \mathbf{k} and is equal to ω ; α is the Fröhlich dimensionless coupling constant, and *V* is the crystal volume.

2.1. The Buĭmistrov–Pekar method Applied to Two-Electron Systems in Polar Crystals with Intermediate Electron–Phonon Coupling

In a present-day representation, the Buĭmistrov– Pekar method amounts to application of the canonical transformation $\exp(\alpha S)H\exp(-\alpha S)$ with the unitary operator

$$S = \sum_{\mathbf{k}} [F_{\mathbf{k}}^*(\mathbf{r}_1, \mathbf{r}_2)a_k - F_k(\mathbf{r}_1, \mathbf{r}_2)a_k^+],$$

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where $F_k(\mathbf{r}_1, \mathbf{r}_2)$ is a function of the coordinates of the electronic system, to the Hamiltonian (1). Then, the following expressions are valid:

$$\exp(S)a_k\exp(S) \longrightarrow a_k + F_k,$$
$$\exp(S)a_k^+\exp(S) \longrightarrow a_k^+ + F_k^*.$$

After averaging over the phonon variables, we obtain the following functional:

$$H = \frac{\beta}{r_{12}} + \sum_{j=1}^{2} \sum_{\mathbf{k}} \nabla_{j} F_{k}^{*} \nabla_{j} F_{k}$$

$$= \sum_{j=1}^{2} \sum_{\mathbf{k}} \left\{ -\nabla_{j}^{2} - \frac{\gamma}{r_{j}} + V_{k} [F_{k} \exp(i\mathbf{k}\mathbf{r}_{j}) + F_{k}^{*} \exp(i\mathbf{k}\mathbf{r}_{j})] \right\} + \sum_{\mathbf{k}} F_{k} F_{k}^{*}.$$
(2)

Expression (2) is initial for further calculations. Let us choose the function F_k as

$$F_k = C_k + \gamma_k f(\mathbf{r}_1, \mathbf{r}_2), \qquad (3)$$

where C_k and γ_k are the variational parameters.

Having substituted (3) in (2) and variating over C_k and γ_k (in contrast to the Buĭmistrov–Pekar method [6], the parameters C_k and γ_k are selected in a specified analytical form), we obtain the following expression for the functional of the ground state of a two-electron system:

$$E_{Bp} + J_S + J_i, \tag{4}$$

$$J_i = -\sum_{\mathbf{k}} V_k^2 \frac{\tilde{U}_k^2}{2k^2 + U_k \omega_k},\tag{5}$$

$$U_{k} = \langle \Psi_{12} | f_{k}(\mathbf{r}_{1}, \mathbf{r}_{2}) L_{k}^{*}(\mathbf{r}_{1}, \mathbf{r}_{2}) | \Psi_{12} \rangle - \langle \Psi_{12} | f_{k}(\mathbf{r}_{1}, \mathbf{r}_{2}) | \Psi_{12} \rangle \langle \Psi_{12} | L_{k}^{*}(\mathbf{r}_{1}, \mathbf{r}_{2}) | \Psi_{12} \rangle,$$
(6)

$$L_k^*(\mathbf{r}_1, \mathbf{r}_2) = \exp(-i\mathbf{k}\mathbf{r}_1) + \exp(-i\mathbf{k}\mathbf{r}_2), \qquad (7)$$

$$U_{k} = \langle \Psi_{12} | f_{k}(\mathbf{r}_{1}, \mathbf{r}_{2}) f_{k}^{*}(\mathbf{r}_{1}, \mathbf{r}_{2}) | \Psi_{12} \rangle$$

- $\langle \Psi_{12} | f_{k}(\mathbf{r}_{1}, \mathbf{r}_{2}) | \Psi_{12} \rangle \langle \Psi_{12} | f_{k}^{*}(\mathbf{r}_{1}, \mathbf{r}_{2}) | \Psi_{12} \rangle.$ (8)

Here, J_i is an addition appropriate for intermediate coupling, and J_s corresponds to the D^- -center functional in the limit of strong electron–phonon coupling.

$$J_{s} = \overline{T}_{12} + \beta \overline{V}_{ee} - \gamma \overline{V}_{e}$$

$$+ \sum_{\mathbf{k}} V_{k}^{2} |\langle \exp(-i\mathbf{k}\mathbf{r}_{1}) + \exp(-i\mathbf{k}\mathbf{r}_{2})\rangle|^{2}, \qquad (9)$$

$$\overline{T}_{12} = -\left[\Psi_{12}(\Delta_{1} + \Delta_{2})\Psi_{12}d\tau, \qquad (10)\right]$$

$$\bar{V}_{ee} = \int \frac{|\Psi_{12}|^2}{r_{12}} d\tau,$$
(11)

$$\overline{V}_{e} = \int \frac{|\Psi_{12}|^{2}}{r_{1}} d\tau + \int \frac{|\Psi_{12}|^{2}}{r_{2}} d\tau.$$
(12)

2.2 The Functional of the Ground State of a Two-Electron System

Choosing $f_k(\mathbf{r}_1, \mathbf{r}_2) = L_k^*(\mathbf{r}_1, \mathbf{r}_2)$ and, hence, $\tilde{U}_k = U_k$, we obtain

$$J_{i} = -\sum_{k} V_{k}^{2} \frac{U_{k}^{2}}{2k^{2} + U_{k}\omega_{k}}.$$
 (13)

In the specific case of the absence of electronic correlations, when the WF of an electronic system can be written as the product of one-electron WFs $\Psi(\mathbf{r}_1, \mathbf{r}_2) = \psi(\mathbf{r}_1)\psi(\mathbf{r}_2)$, relation (13) yields

$$J_{i} = -\sum_{j=1}^{1} \sum_{\mathbf{k}} V_{k}^{2} \frac{\left[1 - F_{k}^{2}(\mathbf{r}_{j})\right]^{2}}{k^{2} + \left[1 - F_{k}^{2}(\mathbf{r}_{j})\right]\omega_{k}},$$
 (14)

where

$$F_k(\mathbf{r}_j) = \int \Psi(\mathbf{r}_j) \exp(i\mathbf{k}\mathbf{r}_j) \Psi^*(\mathbf{r}_j) d\tau_j.$$

Having substituted (14) in (4), we obtain a functional coinciding with formula (18) from the Buĭmistrov and Pekar paper [22].

With a more general choice of the WF (a nonmultiplicative form corresponding to a consideration of electronic correlations), it is necessary to use a more general functional in which the addition, which is appropriate for intermediate coupling J_i , is defined by expression (13).

In a recent publication [27], which was concerned with the two-center bipolaron of intermediate coupling, the electron WF was selected in the form of a symmetrized product of the polaron wave functions centered at different points, i.e., in a nonmultiplicative form (the Heitler–London method applied to a bipolaron). Thus, the functional with a J_i addition defined by Eq. (14) was used for variation. This is correct only for the specific case when the WFs of a two-electron system are chosen as the product of one-electron WFs. Notably, Mukhomorov [27] varied the incorrect functional and performed erroneous numerical calculations. The correct formulas for calculating the bipolaron energy using the Buĭmistrov-Pekar method were given in [24]; however, the trial function chosen in [24] for the entire range of electron-phonon interaction parameters yielded a higher bipolaron energy in comparison with the results in [28].

3. NUMERICAL CALCULATIONS

Let us choose a two-electron WF as a linear combination of Gaussian orbitals:

$$\Psi(\mathbf{r}_{1}, \mathbf{r}_{2}) = \frac{1}{\sqrt{N_{12}}} \sum_{i=1}^{N} C_{i} [1 + (-1)^{S} P_{12}]$$

$$\times \exp(-a_{1i}r_{1}^{2} - 2a_{2i}\mathbf{r}_{1}\mathbf{r}_{2} - a_{3i}r_{2}^{2}),$$
(15)

where P_{12} is an interchange operator for the electron coordinates; and the exponent is equal to S = 0 and 1 for singlet and triplet states of a two-electron system, respectively. The WF of a polaron and *F* center is chosen as

$$\Psi_{p}(\mathbf{r}) = \frac{1}{N_{1}} \sum_{i=1}^{n} C_{i} \exp(-a_{i}r^{2}), \qquad (16)$$

where C_i , a_i , a_{1i} , a_{2i} , and a_{3i} are variational parameters; **r** are the electron coordinates in a polaron; **r**₁ and **r**₂ are coordinates of the first and second electron in a bipolaron, respectively; and N_{12} and N_1 are normalization factors.

In the limit $\eta \longrightarrow 0$ ($\eta = \varepsilon_{\infty}/\varepsilon_0$, where ε_{∞} and ε_0 are optical and static dielectric constants, respectively) the coupling of electrons to the Coulomb core weakens and the D^- center becomes equivalent to a one-center bipolaron or to a Pekar bipolaron. The one-center configuration of a bipolaron can be considered as an elementary two-electron system in a crystal. At the same time, the functional of this system includes the most complex component describing nonlocal interaction of the two-electron system with the phonon field. From this point of view, the addition of interaction with the field of static charge only insignificantly complicates the numerical calculations of the energy spectrum of a bound polaron or D^- center.

Bipolaron energies E_{Bp} , calculated using the system of Gaussian functions (15) (S = 0), are summarized in Table 1. For the entire domain of existence of a bipolaron, the Buĭmistrov–Pekar method yields the lowest ground-state energies for a free bipolaron and the widest region of existence for a bound bipolaron in comparison with the best numerical calculations of these parameters performed using direct variational methods

[28]. For comparison, numerical calculations of E_{Bp}^{A} , performed in [28], are also listed in Table 1. The number of terms N = 5 in the WF (15) used for calculating the bipolaron energy.

When considering a bound bipolaron in a system, an additional parameter describing the interaction of electrons with the field of static charge appears. Let us express the energy of the D^- center as a function of two dimensionless parameters: the Fröhlich electron–phonon coupling constant $\alpha = e^2/(2r_0\hbar\omega)\tilde{\epsilon}$ (where $1/\tilde{\epsilon} = 1/\epsilon_{\infty} - 1/\epsilon_0$, $r_0 = \sqrt{\hbar/2m^*\omega}$, $\hbar\omega$ is the frequency of long-

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	α										
η	6		7		9		20				
	E_{Bp}	E^{A}_{Bp}	E_{Bp}	$E^{ m A}_{Bp}$	E_{Bp}	E^{A}_{Bp}	E_{Bp}	$E^{ m A}_{Bp}$			
0	-12.703	-12.601	-16.234	-16.067	-24.927	-24.652	-111.928	-110.504			
0.01	-12.595	-12.487	-16.053	-15.91	-24.650	-24.354	-110.497	-109.064			
0.1			-14.598	-14.500	-22.068	-21.756	-96.878	-95.335			

Table 1. The ground-state energy of free bipolaron for various parameters α

Note: The parameters E_{Bp}^{A} are taken from [28].

Table 2. The ground-state energy of the D^- -center (bound bipolaron) in $\hbar\omega$ units.

Crystal	α	R	ħω, meV	E^-	E_0	E_B^-	E_B^0	E_B^-/E_B^0
CdTe	0.272	0.657	21.08	-1.266	-0.965	0.029	0.693	0.042
CdS	0.529	0.783	38.0	-1.931	-1.363	0.039	0.834	0.047
ZnSe	0.45	0.924	31.4	-1.926	-1.428	0.048	0.978	0.049
AgBr	1.64	1.68	15.4	-5.656	-3.818	0.198	2.178	0.091
				-5.637^{L}	-3.817 ^L	0.180 ^L	2.177 ^L	0.083^{L}
						0.132 ^A	2.166 ^A	0.061 ^A
AgCl	1.9	1.90	24.4	-6.668	-4.483	0.285	2.583	0.110
				-6.643^{L}	-4.482^{L}	0.261 ^L	2.582 ^L	0.101^{L}
				-6.662^{A}		0.202 ^A	2.560 ^A	0.078^{A}
CdF^2	2.53	1.274	50.0	-7.357	-4.510	0.317	1.98	0.016

Note: Superscripts L and A indicate the results of calculations in [5] and [6], respectively.

wavelength longitudinal optical phonons, and m^* is the effective electron mass) and the ratio of the effective rydberg to $\hbar\omega$ given by

$$R = m^* e^4 / 2\varepsilon_0^2 \hbar^3 \omega = (e^2 / 2r_0 \hbar \omega \varepsilon_0)^2$$

As an example, Table 2 lists the energies of the ground state of the $D^-(S=0)$ and D^0 centers: E^- and E^0 , respectively, for a number of crystals. The binding energy is designated as E_B . All energies are expressed in $\hbar\omega$. The superscripts L and A define the parameters obtained in [5] and [6], respectively. The number of terms in expressions (15) and (16) for the WF, which were used to calculate the energy of D^- and D^0 centers, equals 12.

In order to assess the flexibility of the WFs used for calculating the energy of two-electron systems, we used these WFs to calculate the ground-state energy of the negative hydrogen ion H⁻ (this atomic system is the closest analogue of a bipolaron in atomic physics). This energy is equal to -1.055470 [N = 28 in (15)], whereas the precise value is equal to -1.055502 [29].

The possibility of forming metastable triplet states of D^- centers should be considered separately. This matter has assumed great importance because the

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absence of bound triplet states of D^- centers (similar to the absence of bound excited states of the H⁻ ion [30]) is one of the basic conditions for realizing the detection of the spin state of a two-electron system in quantum computers operating on the basis of electron spin resonance [21]. The scheme proposed in [21] can be briefly described as follows: the application of an electric field along the line connecting a singlet-state exchange-coupled pair of shallow-level paramagnetic centers, can cause the controlled charge transfer to one of the Coulomb centers and to the formation of a D^- center. In the triplet state, similar combination is impossible. Donors remain neutral, and there is no charge transfer between centers. Due to this circumstance, the spin state of the two-electron system can be observed.

Variatonal calculations with the use of a system of functions (15) for S = 1 showed that, for the entire range of parameters of electron-phonon interaction, including the region of extremely strong coupling ($\alpha > 20$), the relation $E_p + E_D \le E_{Dt}$ is valid, where E_p , E_D , E_{Dt} are the energies of a polaron, a neutral donor and a D^- center in the triplet state, respectively. Thus, electron-phonon interaction in the continuous approximation does not lead to the formation of a metastable triplet state of the D^- center. This is a close analogy to the the-

orem concerning the absence of bound excited states of a negative hydrogen ion H⁻ [30]. Our calculations showed that the bound state of the triplet bipolaron that formed as a result of coupling to optical phonons is also energetically unfavorable. Our test calculations of the orthohelium energy, with the use of the system of functions (15) for S = 1, show that there is good agreement with the experimental values of the orthohelium energy.

It should be noted that an external magnetic field gives rise to upper excited states of the D^- center [31]. Calculations of the triplet states of the D^- center in spherical quantum dots were performed in [8].

4.CONCLUSIONS

The chosen system of Gaussian functions in combination with the Buĭmistrov–Pekar method of calculations yields lower D^- center energies and enables one to perform calculations of the energy spectrum of more complex two-electron systems, such as exchange-coupled pairs of paramagnetic centers, and also obtain reliable values of the ground-state energy of paramagnetic centers and their complexes in anisotropic crystals and low-dimensional systems.

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