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Quantum States and Optical Transitions of the F_2^+ Center Electron

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The quantum states of a system consisting of two negative vacancies and an electron in an ionic crystal (F_2^+ center) are considered. The thermal dissociation and ground state energies of the system are computed by quantum mechanical consideration of the motion of lattice ions. The parameters of the optical absorption band—its position and half–width—have been determined. For illustration the numerical values have been obtained using this theory for KCl and KBr crystals; the results are in a satisfactory agreement with the experimental results.

AN intensive experimental study of impurity absorption in ionic crystals now makes it possible at least for a part of the spectrum, to construct models of several absorbing centers (color centers). This discussion will concern the R and M absorption bands found at wavelengths longer than the F band.

Seitz[1] has attempted the foundation of a model of the absorbing centers which correspond to those bands with the aid of an analysis of the experimental data on the kinetics of their production, and both thermal and optical dissociation. In particular, one of the two R bands (R_2 band) was identified with the absorption band of F_2 centers, i.e., a system consisting of two negative vacancies and two electrons. The second band (R_1 band) was identified as an absorption band of F_2^+ centers. It is natural that, without a quantitative calculation of the parameters of these bands, it was not possible to carry out such identification at all reliably.

In the previous works [2,3] one of the authors calculated the quantum states and parameters of the F_2 center absorption band. These calculations showed that the parameters of an F_2 absorption band are indeed very close to the corresponding parameters of an R_2 band.

In recent works[4,5] the absorption of light by F_2 and F_2^+ centers was studied in two approximations: the approximation of crystal–continuum which was earlier employed by Pekar and Deigen[2,3], and the molecular orbital method.[5] In the molecular orbital approximation, the authors entirely ignored the interaction of the electron with the optical vibration modes of the crystal (the polarization of the crystal by the field of the electron), calculated incorrectly the overlap integrals, and took into account only the first sphere of ions surrounding the vacancy. Such rough approximation did not lead to an agreement of the theory with the experiment. In calculating the frequency of the optical transition by the first technique the following material errors were made: 1. the periodic crystal potential was igonored; 2. to determine the parameters of the ground and excited states in the Franck–Condon approximation the authors calculated the energy of the "self–consistent" state [6,2] instead of solving the Schrödinger equation for the system in a fixed potential well.

Calculations show that every one of the errors enumerated above can cause errors of the order of magnitude of the calculated quantity. Therefore the results of Nagamiya et al [4,5] are in sharp disagreement with experiment.

The quantum states and optical transitions of F_2^+ center are considered in the present work by a method developed previously[2,3] which does not contain the above errors.

1. THE GROUND STATE OF THE SYSTEM

In a complete analogy with the previously published results[6,2] the parameters of the ground state of an F_2^+ center are determined from the condition of an absolute minimum of the function

$$I_0[\psi] = \frac{\hbar^2}{2\mu}\int |\nabla\psi|^2 d\tau - \frac{e^2}{\varepsilon}\int |\psi|^2\left(\frac{1}{r_a} + \frac{1}{r_b}\right)d\tau - \frac{c}{8\pi}\int D^2[\psi, \mathbf{r}]\,d\tau \qquad (1)$$

with the additional normalization condition

$$\int |\psi|^2 d\tau = 1, \qquad (2)$$

where μ is the effective mass of an electron, ϵ

is the dielectric constant, $c = 1/n^2 - 1/\epsilon$, n is the index of refraction, ψ the electronic part of the system's wave function, D $[\psi, \mathbf{r}]$ the electrostatic field density produced by the electron cloud, r_a and r_b are the distances from the a and b vacancies. Direct variational method was used to find the minimum $I_0\ [\psi]$. The approximate wave function was chosen as

$$\psi(\mathbf{r}) = A(\psi_a + \gamma\psi_b); \tag{3}$$

in which ψ_a and ψ_b were given by the expressions

$$\psi_a = \alpha^{3/2} e^{-\alpha r_a} / \sqrt{\pi}; \quad \psi_b = \alpha^{3/2} e^{-\beta r_b} / \sqrt{\pi}; \tag{4}$$

Here A is the normalizing constant, α and γ the variational parameters.

It will be shown below that the wave function of Eq. (3) approaches infinity as F_2^+ center changes into F center and isolated vacancy at R.

Substituting Eq. (3) into the functional (1), we obtain

$$I[\alpha, \gamma] = \frac{e^2}{\varepsilon} A^2 \left\{ \frac{\hbar^2 \varepsilon}{2\mu e^2} [(1 + \gamma\gamma^*) I_2 \right. \tag{5}$$

$$- (\gamma + \gamma^*) I_3] - [(I_4 + I_5)(1 + \gamma\gamma^*) + 2(\gamma + \gamma^*) I_6]$$

$$- \frac{\varepsilon c}{2} A^2 [(1 + \gamma^2\gamma^{*2}) u_1 + 2(\gamma + \gamma^*)(1 + \gamma\gamma^*) u_3$$

$$\left. + (\gamma + \gamma^*)^2 u_4 + 2\gamma\gamma^* u_2] \right\},$$

where I_k and u_k are integrals dependent on wave-functions ψ_a and ψ_b. The extremum condition

$$\partial I / \partial\gamma = 0, \quad \partial I / \partial\gamma^* = 0$$

leads to a system of equations for determination of γ. Two complex and four real roots are obtained as a final solution. The real roots are:

$$\gamma_1 = 1; \quad \gamma_2 = -1; \tag{6}$$

$$\gamma_{3,4} = (b_3 / 2b_4) \pm \sqrt{(b_3/2b_4)^2 - 1};$$

where b_k are complicated functions of the parameters of the crystal and of the integrals I_k and u_k. The roots $\gamma_{3,4}$ yield identical values for the functional (5). Following the usual procedure of the variation method, the solution of the problem is the root which yields the lowest value on substitution into the variation functional.

Minimizing over α was carried out graphically for each of the obtained values of γ. The quantities α and γ obtained in this manner determine the electronic part of the ground state wave–function of the system.

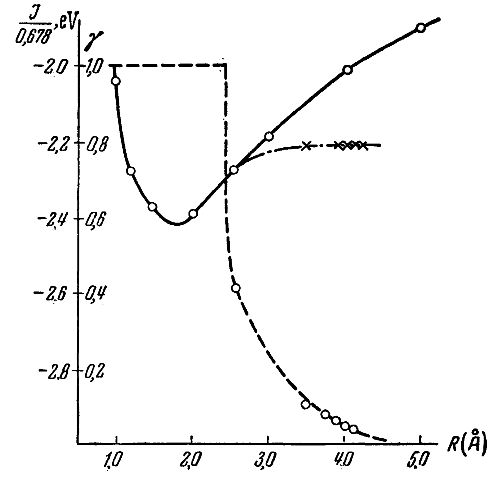

FIG. 1. Solid curve shows I for $\gamma = 1$, dotted curve for $\gamma = \gamma_{\text{ext.}}$ and dash–dotted curve for $\gamma = \gamma_{3,4}$.

Investigation of the dependence on R of the functional (5) showed that for sufficiently large R the lowest value of the functional $I\ [\alpha, \gamma]$ is obtained with the roots $\gamma = \gamma_{3,4}$. These roots approach zero as $R \to \infty$. It follows from Eq. (3) that the wave–function of the system then changes into a wave–function of the F center type and from Eq. (5) that the functional $I\ [\alpha, \gamma]$ goes into a functional for an F center. For small R, the extrema of the functional are given by the root $\gamma = 1$. It is interesting to note the existence of a continuous transition from the extreme value of the functional for $\gamma = \gamma_{3,4}$ to the extremum for $\gamma = 1$ as R decreases.

The results of a numerical calculation for the crystal KCl are shown in Fig. 1. It is evident from the Figure that the minimum $I_0 = I_0(R)$ corresponds to a separation R = 1.8 Å between vacancies. The difference in energies of the systems with F_2^+ and F centers turns out to be considerable. However, in real crystals with the NaCl type lattice, the minimum possible separation between two identical isolated vacancies is approximately $d\sqrt{2}$, where d is the separation between two nearest neighbors oppositely charged ions. At this separation, the derivative $\partial I_0 / \partial R$, which determines the attractive force between two vacancies in an F_2^+ center, is nearly zero and in any case is considerably smaller than other forces in the lattice. Therefore, separations approximately equal to $d\sqrt{2}$ should be considered as equilibrium separations rather than those that correspond to the minimum $I_0(R)$. For these same separations, it is evident from Fig. 1 that the energy gain due to creation of an F_2^+ center from an F center and vacancy is negligible for KCl crystal (as well as other alkali-halide crystals). Therefore the thermal equilibrium of an F_2^+ center is evidently connected mainly with large activation energy of diffusion of vacancies, rather than the energy gain in creation of F_2^+ centers.

Comparatively small radii of the electron cloud in the ground state of an F_2^+ center and large separations between vacancies allow a second approximation of an F_2^+ center as an F center in an approximately uniform field of a vacancy. To determine the energy of such a system one can use the results of Perlin,[7] according to which the energy gain in creation of an F_2^+ center from an F center and a vacancy is determined by the expression

$$\Delta I_0 = \eta E^2 / 2, \qquad (7)$$

where η is the polarizability of the F center, $E = e/\epsilon R^2$ is the field of a vacancy at the position of the F center. For KCl, for example,

$$\eta = 8.7\,(m/\mu)^3 \cdot 10^{-23}\ \text{cm}^3,$$

$$\varepsilon = 4.78,\ R = 4.44\ \text{Å}.$$

With these values of the parameters, we obtain $\Delta I_0 \sim 0.01$ ev. Therefore a calculation in this approximation leads to a negligible energy gain.

2. EXCITED STATE OF AN F_2^+ CENTER

As pointed out above, the parameters will be calculated for : a) the lowest excited state in a fixed polarization potential well, determined by the ground state wave–function; b) the lowest–lying self-consistent excited state.

a) The quantum states of the system are determined in this case by the solution of the wave equation:

$$\left(-\frac{\hbar^2}{2\mu}\Delta - \frac{e^2}{\varepsilon r_a} - \frac{e^2}{\varepsilon r_b} + V_p\right)\varphi(\mathbf{r}) = E_1\varphi(\mathbf{r}). \qquad (8)$$

Here V_p is the polarization potential well produced by the ground state electron cloud:

$$V_p = -e^2 c \int \frac{\psi^2(\mathbf{r}_1)}{|\mathbf{r} - \mathbf{r}_1|}\, d\tau_1. \qquad (9)$$

Equations (3) and (9) allow a determination of the explicit form of V_p . Substituting Eq. (9) into Eq. (8) and solving the latter, one obtains the parameters of the quantum states of the system.

The problem is simplified considerably for those crystals whose extreme value of γ is very small. In particular, such crystals include alkali-halides. For these crystals the polarization potential well practically coincides with the polarization well produced by an electron in the ground state of an isolated F center.

The Eq. (8) is solved in this case by perturbation theory, taking as a small perturbation $-e^2/\epsilon r_b$. The zero order equation represents the wave function of an F center with an electron localized at a vacancy a. As a zero order wave–function, a linear combination of functions corresponding to two unperturbed 2s and 2p wavefunctions of an F center is selected. The symmetry of the problem shows that 2p states with quantum number $m = \pm 1$ need not be taken into account. Therefore,

$$\varphi_0 = c_1\varphi_{a1} + c_2\varphi_{a2}, \qquad (10)$$

$$\varphi_{a1} = K_1 e^{-\beta r_a} r_a \cos\vartheta_a, \qquad (11)$$

$$\varphi_{a2} = K_2 e^{-\delta r_a}(1 - s r_a),$$

where K_1 and K_2 are normalization constants. The parameters of the non–self-consistent 2p state of an F–center were determined by Deigen,[8] the parameters of the 2s state were especially calculated.

The calculations could be carried out to a numerical result. In particular, for crystals KCl and KBr the energy of the excited state to the first order perturbation treatment turns out to be:

$$E_1(\text{KCl}) = -2.17\ \text{ev}\,;\quad E_1(\text{KBr}) = -1.93\ \text{ev}$$

Calculations to the higher order of perturbation theory show that the error in the above results does not exceed one percent.

Replacement of Eq. (8) by an equivalent variation principle permits a calculation of the parameters of a non-self-consistent excited state by thevariation method. The appropriate variation function has the following form:

$$\bar{E}_1[\varphi] = \frac{\hbar^2}{2\mu}\int|\nabla\varphi|^2 d\tau \tag{12}$$

$$-\frac{e^2}{\varepsilon}\int|\varphi|^2\left(\frac{1}{r_a}+\frac{1}{r_b}\right)d\tau - e^2c\int\frac{\psi^2(1)\,\varphi^2(2)}{|\mathbf{r}_1-\mathbf{r}_2|}d\tau_1 d\tau_2.$$

The approximate wave function is chosen in the form

$$\varphi = N(\varphi_{a1} + k\varphi_{a2}), \tag{13}$$

where φ_{a1} and $\varphi_{a2}$are given by Eq. (11), N is the normalizing constant, β, δ, k the variation parameters. Such selection of approximate wavefunctions is motivated by the results of the perturbation theory. We note that such choice is good only in the case when γ is close to zero in Eq. (3), since only then is the orthogonality condition of Eqs. (13) and (3) fulfilled (with an appropriate choice of the parameter s). If γ is close to unity in Eq. (3), the approximate wave function should be chosen as a linear combination of the corresponding wave function of the centers a and b. Substituting Eq. (13) into Eq. (12) and performing the variation over k results in a relationship for k. After substituting the values for k into the variation functional and numerically minimizing it over β and δ one can carry out a calculation of the energy term of the excited state. In particular, for KCl and KBr crystals:

$$E(\text{KCl}) = -2.17 \text{ ev}; \quad E(\text{KBr}) = -1.94 \text{ ev}.$$

It should be noted that the results of the variation and perturbation methods agree very well.

b). The parameters of the self-consistent excited state of an F_2^+ center are obtained by seeking the minimum of the functional

$$I_1[\varphi] = \frac{\hbar^2}{2\mu}\int|\nabla\varphi|^2 d\tau \tag{14}$$

$$-\frac{e^2}{\varepsilon}\int|\varphi|^2\left(\frac{1}{r_a}+\frac{1}{r_b}\right)d\tau - \frac{e^2c}{2}\int\frac{\varphi^2(1)\,\varphi^2(2)}{|\mathbf{r}_1-\mathbf{r}_2|}d\tau_1 d\tau_2.$$

The wave function φmust satisfy the normalization conditions and must be orthogonal to the ground state wave function.

It is natural to try a wave function of the excited state similarly to Eq. (3) in the form:

$$\varphi = B(\varphi_a + \rho\varphi_b); \tag{15}$$

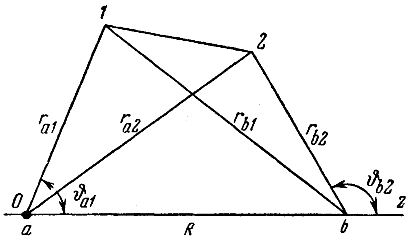

FIG. 2.

$$\varphi_a = \pi^{-1/2}\beta^{5/2}e^{-\beta r_a}r_a\cos\vartheta_a; \tag{16}$$

$$\varphi_b = \pi^{-1/2}\beta^{5/2}e^{-\beta r_b}r_b\cos\vartheta_b$$

Figure 2 shows the coordinates used; ρ and β are the variation parameters.

Substitution of Eq. (15) into (14) and subsequent variation over ρ leads to a relationship for ρ. Depending on the crystal parameters, the lowest value of the function of Eq. (14) is determined by one of the roots $\rho = -1$ or $\rho = \rho_{3,4}$ The quantities $\rho_{3,4}$ are determined in the same manner as $\gamma_{3,4}$ of Eq. (6), except that the integrals I_k and u_k depend on the functions φ_a and φ_b of Eq. (16).

The resulting solution will be good only if the orthogonality condition is fulfilled for the wavefunctions of Eq. (3) and (15). This condition is satisfied under two conditions: 1) if the extreme values of γ and ρ for a fixed R are equal to 1 and -1 respectively; 2) if the extreme values of γ and ρ for a fixed R are simultaneously close to zero.

Under these conditions, one can obtain a defnite energy value of the self-consistent excited state by substituting the extreme values of ρ into the functional andminimizing it with respect to β. In all other cases the wave function must be selected in a different form. In particular, if γ is close to zero in Eq. (3), the wave function of the excited state should be taken in the form

$$\varphi = M(\varphi_a + \nu\varphi_b + \lambda\psi_a), \tag{17}$$

where ψ_a is given by Eq. (4) and M is a normalization constant.

The parameter λ is determined from the condition of orthogonality of the ψ wave functions of the ground and excited states, β and ν are the variation parameters.

The selection of the wave functions in the form of Eqs. (15) and (17) is also justified by the fact that these wave functions go into the ψ wavefunction of an isolated F center as $R \to \infty$, in accordance with the physical formulation of the problem. Substitution of Eq. (17) into Eq. (14) and minimizing over ν and β also allows a calculation of the energy of the excited self-consistent state.

In numerical calculations, we have limited ourselves to approximate methods, setting β approximately equal to the corresponding parameter of an isolated F center and carrying out the variation procedure only for ν. Therefore the values I_1 are somewhat higher than they should be. The numerical calculations carried out for two crystals yielded the following values

$$I_1(\mathrm{KCl}) = -0.78\,\mathrm{ev}\,;\quad I_1(\mathrm{KBr}) = -0.77\,\mathrm{ev}\,.$$

3. PARAMETERS OF THE ABSORPTION BAND OF F_2^+ CENTERS

Illumination of a crystal containing F_2^+ centers by light can lead to a transition of an electron from ground state into an excited state or to a photodissociation of an F_2^+ center with simultaneous absorption of light. The energy of the photodissociation of an F_2^+ center is determined by the expression

$$E_0 = I_0[\psi_{\mathrm{ext}}] - \frac{e^2c}{2}\int\frac{\psi^2_{\mathrm{ext}}(1)\,\psi^2_{\mathrm{ext}}(2)}{|\mathbf{r}_1-\mathbf{r}_2|}\,d\tau_1 d\tau_2, \tag{18}$$

where ψ_{ext} is the value of ψ wave function in the ground state corresponding to the minimum I_0 The frequency of the band absorption peak by an F_2^+ center is determined by the expression

$$\hbar\Omega_{\max} = |E_0 - E_1|. \tag{19}$$

Numerical calculations yielded the following values for crystals KCl and KBr (in ev):

$$E_0(\mathrm{KCl}) = -4.07;\quad E_0(\mathrm{KBr}) = -3.62;$$

$$\hbar\Omega(\mathrm{KCl}) = 1.90;\quad \hbar\Omega(\mathrm{KBr}) = 1.68.$$

Using the results of Pekar[9] in a way similar to the theory of F centers,[6] it is possible to obtain the following expressions for the frequency of the band absorption peak of an F_2^+ center and the half-width δ_0 of the absorption band at low temperatures:

$$\hbar\Omega_{\max} = |I_0 - I_1| + a\hbar\omega/2, \tag{20}$$

$$\delta_0 = 2\hbar\omega\sqrt{a\ln 2},$$

$$a = \frac{c}{4\pi\hbar\omega}\int(\mathbf{D}_0 - \mathbf{D}_1)^2\,d\tau. \tag{21}$$

Here ω is the limiting vibration frequency of the ions in the lattice, $\mathbf{D}_0$ and $\mathbf{D}_1$ are the electrostatic field densities produced by the ψ electron cloud in the ground and excited states.

The expression for a can be written in a somewhat different form:

$$a = e^2c\left\{\int\frac{\psi^2(1)\,\psi^2(2)}{|\mathbf{r}_1-\mathbf{r}_2|}\,d\tau_1 d\tau_2 \right. \tag{22}$$

$$\left. + \int\frac{\varphi^2(1)\,\varphi^2(2)}{|\mathbf{r}_1-\mathbf{r}_2|}\,d\tau_1 d\tau_2 - 2\int\frac{\psi^2(1)\,\varphi^2(2)}{|\mathbf{r}_1-\mathbf{r}_2|}\,d\tau_1 d\tau_2\right\}.$$

Substitution of Eqs. (3) and (17) into Eq. (22) and performance of the calculations for KCl and KBr crystals results in the values0.63 and 0.57 ev, respectively, for $a\hbar\omega/2$. Using the above values of I_0, I_1 and $a\hbar\omega/2$, we obtain (in ev):

$$\hbar\Omega_{\max}(\mathrm{KCl}) = 2.0;\quad \hbar\Omega_{\max}(\mathrm{KBr}) = 1.8; \tag{23}$$

$$\delta_0(\mathrm{KCl}) = 0.30;\quad \delta_0(\mathrm{KBr}) = 0.25.$$

The discrepancy of the numerical values of $\hbar\Omega_{\max}$ in Eq. (23) from the ones obtained earlier can be explained by the fact that the values of Eq. (23) were obtained by an approximate calculation in which the absolute magnitude of I_1 was somewhat lower. The latter circumstance has only a small effect on the numerical value of the half-width of the absorption band because δ_0 is proportional to $\sqrt{a}$. Furthermore, any error in the evaluation of a caused by some inaccuracy in the calculation of β was found to be very small in more detailed calculations.

A comparison of the above values of $\hbar\Omega_{\max}$ and δ_0 with the corresponding parameters of the R_1 band is shown in the following table:

	$\hbar\Omega_{\mathrm{exp}}$	$\hbar\Omega_{\mathrm{th.}}$	$\delta_{0\,\mathrm{exp}}$	$\delta_{0\,\mathrm{th.}}$
KCl	1.85[10]	1.91	~0.25[10]	0.30
KBr	1.7[11]	1.68	—	0.25

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Investigation of the Energy Levels of the F^{20} Nucleus by Magnetic Analysis

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The products of the reaction F^{19} (*d,p*) F^{20} were studied by applying the magnetic analysis method. In the investigated excitation energy range (up to 6.75 mev) 24 proton groups were recorded which determine the ground state and 23 excitation levels of the F^{20} nucleus. Six of these levels, with excitation energies 4.55, 4.86, 5.41, 5.54, 6.07 and 6.36 mev, have not been observed previously. The $E^* = 6.74$ mev level in the (*d,p*) reaction has likewise been observed previously, but the relatively close F^{20} level has been detected in investigations on resonance scattering of neutrons.

ONE of the most effective means of investigating the levels of the nucleus F^{20} is analysis of the products of the reaction $F^{19}(d,p)$ $F.^{20}$ This reaction has been investigated in a series of experiments.[1–6] The results of the most recent of these can be summarized as follows.

Burrows, Powell and Rotblat investigated the energy spectrum of protons from the indicated reaction by measuring their range in photo plates. Notwithstanding the comparatively high energy of the bombarding deuterons (8 mev), the authors studied only a relatively narrow band of excitation energies of the F^{20} nucleus, up to 4.73 mev. In this interval the authors observed ten levels of the F^{20} nucleus. 8 proton groups with excitation energy less than 4.01 mev were reported by Allen and Rall[4] In their work, the proton energy was determined by range in aluminum, that is, under circumstances giving poor accuracy and resolution.

In a subsequent short communication of Shull,[5] the proton energy was studied by a more exact method, with the aid of magnetic analysis. However, notwithstanding the bombardment by deuterons of energy greater than 10 mev in this work, the author investigated an energy range of excitation only to 4.4 mev and observed only seven levels of the F^{20} nucleus.

In 1952 Watson and Buchner[6] gave more exact and complete information about the energy levels of the F^{20} nucleus in the excitation interval up to 4.3 mev. The target was bombarded by deuterons of comparatively small energy (up to 2.1 mev) and the authors were able to observe, with the aid of a magnetic spectrometer, 19 proton groups characterizing the ground state of the F^{20} nucleus and 18 excited levels.

At the beginning of the present work (May, 1955) a collection of the data on the levels of the F^{20} nucleus given in the survey of Ajzenberg and Lauritsen[7] gave an incomplete picture. There was an excitation energy region up to 4.3 mev which had been sufficiently thoroughly studied by analysis of the reaction F^{19} (*d, p*) F^{20}. There were also data in the region of higher excitation energy (6.6 to 8.5 mev) obtained by study of the resonance scattering of neutrons by F^{19}. The interval between these two regions remained practically uninvestigated.

The present work had the aim of investigating the levels of F^{20} in the region of higher excitation energy and a filling of the uninvestigated gap between two series of data which existed till then.