

Method of Calculating the Energy Levels of Trapped Electrons in Ionic Crystals*

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Starting from a many-body formulation that includes the phonon field, we develop a method of calculating the energy levels of an electron trapped at a defect in an ionic crystal. Detailed calculations for the F center are given. The higher excited states of the F center and the K absorption bands are also discussed.

I. INTRODUCTION

IN this paper we shall discuss a method of calculating the energy levels of the trapped electron in an ionic crystal. In particular, we shall consider in the formalism two types of traps, the F center and the interstitial ion.

The presently accepted picture of the F center is the lattice-defect model in which an electron is trapped by a negative-ion vacancy. The F absorption band arises from the $1s-2p$ transition of the trapped electron. The interstitial positive ion is also predicted to trap electrons, but there is little experimental work on this center.

The energy levels and properties of these two kinds of traps have been discussed by Simpson¹ and numerous other authors and are summarized in the works of Mott and Gurney,² Seitz,³ and Gourary and Adrian.⁴

We propose a new approach to the problem of the trapped electron which starts from the many-body point of view in the Hartree approximation and includes the electron-phonon interaction.

The higher excited states of the F center are also discussed; it is pointed out that the K absorption band may be associated with the transition from the ground state to its $3p$ state, and the highest true bound state may be viewed as a perturbation on the polaron problem.

II. FORMALISM

A point charge moving in a perfect ionic crystal with a rigid lattice would experience only the periodic electrostatic potential. This effect can be approximated by using an effective mass appearing in its kinetic energy. The remaining electrostatic forces are due to departure from perfect periodicity and rigidity of the lattice. Those include (a) the ion vacancies, impurities, and other type defects in the lattice; (b) temporary displacements of the ions in the crystal from their mean positions, due to thermal motion or the Coulomb field of the moving point charge.

In the present discussion we are concerned only with a negative-ion vacancy or an interstitial ion and the trapped electron. If we include the temporary displacements of the ions produced by the Coulomb field of the moving point charge, the Hamiltonian of such a system is

$$H = \frac{\mathbf{p}^2}{2m} + \sum_k \hbar\omega_k a_k^* a_k + H_{el} + H_{cl} + H_{ec}. \quad (1)$$

The first term of (1) represents the kinetic-energy operator of the system of trap and electron, i.e., the negative-ion vacancy and its electron, or the positive point charge ze and its electron. For convenience, we will call the negative-ion vacancy or the positive point charge the trap center to distinguish it from the system of trap and electron. m is the reduced effective mass of the system of trap center plus electron. The second term is the phonon part of the Hamiltonian in which the zero-point energy is omitted, as we shall be mainly concerned with energy differences of the problems. It is written in terms of creation and annihilation operators for phonons of momentum \mathbf{k} , a_k^* and a_k , respectively. H_{el} represents the interactions between the trapped electron and the phonons; H_{cl} , the interaction between the trap center and the phonons; and H_{ec} the interaction of the trapped electron with the trap center.

When the electron is moving in the region very close to the trap center, its electric field is almost completely shielded by the trap center. Furthermore, the electron moves relatively fast. Hence, the ions of the crystal do not follow the electronic motion and there is no temporary displacements of ions. Therefore, for this case the problem may be treated adiabatically and the second term in Eq. (1) vanishes since no phonons are created. On the other hand, when the electron is moving in the region far from the trap center, it moves relatively slowly compared to its motion when nearby. In this case, the ions can follow its movements and temporary displacements corresponding to acoustic and optical lattice vibration result. For this problem, only the long-wavelength longitudinal optical modes are important.

From considering these two cases, it seems reasonable to divide the crystal in two parts: one consisting of a spherical region of radius R with center at the trap center and the other consisting of the remaining part of the crystal. The radius R is taken as a parameter of the problem.

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¹ J. H. Simpson, Proc. Roy. Soc. (London) **A197**, 269 (1949); **A231**, 308 (1955).

² N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Clarendon Press, Oxford, 1948).

³ F. Seitz, Rev. Mod. Phys. **18**, 384 (1946); **26**, 7 (1954).

⁴ B. S. Gourary and F. J. Adrian, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1960), Vol. 10, p. 128.

When the electron moves in the first region, the ions do not follow its motion. The Hamiltonian then consists of the kinetic energy plus the last three terms H_{el} , H_{cl} , and H_{ec} . For this case we shall denote the sum of these three terms as V_0 . On the other hand, when the electron moves in the second region the ions follow its motion. We shall treat this region as a dielectric medium. Then

$$H_{ec} = -ze^2/\epsilon_\infty r, \quad (2)$$

and the H_{el} and H_{cl} can be written in the Fröhlich forms, that is,

$$H_{el} = \sum_k (V_k a_k e^{i\mathbf{k}\cdot\mathbf{r}} + V_k^* a_k^* e^{-i\mathbf{k}\cdot\mathbf{r}}), \quad (3)$$

$$H_{cl} = -\sum_k (V_k a_k + V_k^* a_k^*), \quad (4)$$

$$V_k = -i(2\pi z e^2 \hbar \omega_k / k^2 V \epsilon^*)^{1/2}.$$

Here, we take the origin of position vectors at the trap center. \mathbf{r} is the position vector of the electron, V is the volume of crystal, ω_k is the frequency of the longitudinal optical modes, and $1/\epsilon^* = 1/\epsilon_\infty - 1/\epsilon_s$, where ϵ_∞ and ϵ_s are the optical and static dielectric constants for the crystal.

Then the Hamiltonian H may be written as follows:

$$H = \frac{\mathbf{p}^2}{2m} + \begin{cases} V_0 & \text{for } r < R \\ \sum_k \hbar \omega_k a_k^* a_k + \sum_k (V_k a_k e^{i\mathbf{k}\cdot\mathbf{r}} + V_k^* a_k^* e^{-i\mathbf{k}\cdot\mathbf{r}}) - \sum_k (V_k a_k + V_k^* a_k^*) & \text{for } r > R. \end{cases} \quad (5)$$

The wave function corresponding to Eq. (5) is given by

$$\Psi = \psi(\mathbf{r})\Phi, \quad (6)$$

where $\psi(\mathbf{r})$ and Φ are the wave functions of the trapped electron and the phonons, respectively. The phonon wave function Φ is written in the Hartree form

$$\Phi = \prod_k f(k), \quad (7)$$

where $f(k)$ is the wave function of phonons of momentum \mathbf{k} .

To determine the one-phonon wave function $f(k)$, we assume that the electron-phonon interaction is a small perturbation on the lattice energy. Furthermore, we average this interaction over the electronic motion to give a interaction energy which depends on the phonon coordinate alone. In this averaging, a fraction of the electronic charge is outside the spherical region of radius R . Thus, the electric field of the F center may not be completely shielded and ions in the crystal display displacements from their mean positions to interact with the F center, in other words, the ions follow the electronic motion. This will not give a large error to the final result, because the electronic part plays an important role in this problem. It is to be em-

phasized, however, that the approximation of averaging over electronic motion to find an average electron-phonon interaction is used only to find an approximation of $f(k)$, and is not used elsewhere.

Then the one-phonon wave function may be determined from perturbation theory. To first order this gives

$$f(k) = N_k \left[1 - \frac{V_k^*}{\hbar \omega_k} (\varphi_k^* - 1) a_k^* \right] |0\rangle. \quad (8)$$

Here $|0\rangle$ represents the phonons' vacuum state and φ_k is given by

$$\varphi_k = \int \psi^*(\mathbf{r}) \psi(\mathbf{r}) e^{i\mathbf{k}\cdot\mathbf{r}} d^3r. \quad (9)$$

N_k is a normalization factor of $f(k)$ and, since the crystal volume is large, we take N_k as approximately unity.

Using these phonon functions, the effective Hamiltonian for the system of electron and trap center is

$$H_{\text{eff}} = \langle \Phi, H \Phi \rangle = \frac{\mathbf{p}^2}{2m} + \begin{cases} V_0 & \text{for } r < R \\ \langle \Phi, \sum_k \hbar \omega_k a_k^* a_k \Phi \rangle + \langle \Phi, \sum_k (V_k a_k e^{i\mathbf{k}\cdot\mathbf{r}} + \text{c.c.}) \Phi \rangle - \langle \Phi, \sum_k (V_k a_k + V_k^* a_k^*) \Phi \rangle - \frac{ze^2}{\epsilon_\infty r} & \text{for } r > R. \end{cases} \quad (10)$$

In evaluating this, the summation over k was replaced by an integration as is the usual procedure. In most cases, for the present problems the integrals are rapidly convergent as k increases and k_{max} , which equals π/a , can be replaced by infinity. The energy level of the trapped electron is

$$E = \langle \psi(\mathbf{r}), H_{\text{eff}} \psi(\mathbf{r}) \rangle. \quad (11)$$

III. THE F CENTER

For the F center we make the assumption that the negative-ion vacancy may be considered as having an effective charge of plus unity. From the previous work,⁵ the effective potential for the F center, that is, V_0 in Eq. (10) is

$$V_0 = -\frac{M e^2}{d} + \frac{e^2}{R_0} \left(1 - \frac{1}{\epsilon_\infty} \right). \quad (12)$$

M is the Madelung constant for the particular ionic crystal and d is the nearest-neighbor distance. ϵ_∞ is the optical dielectric constant and R_0 is the radius of a spherical region in which electronic polarizability is important, but ionic polarization is negligible. The

⁵ S. R. Tibbs, Trans. Faraday Soc. **35**, 1471 (1939).

value of R_0 is given approximately by Mott and Gurney⁶ as $R_0 \approx 0.9 d$.

The first term of Eq. (12) represents the interaction energy between the ions neighboring the negative-ion vacancy and the F -center electron at the center of the vacancy. The second term is the energy gained from the polarization of the medium surrounding the negative-ion vacancy when the F -center electron is removed.

To simplify the calculations we choose a value of R . A reasonable choice is to take $R=R_0$, since they are defined in essentially the same way.

Two methods have been used to form F -center wave functions. They are the vacancy-centered (VC⁴) method and the linear combination of the atomic orbitals (LCAO) method.⁴ In the VC method the wave functions are approximated by simple functions centered at the negative-ion vacancy, while in the LCAO method a linear combination of wave functions centered on the ions neighboring the vacancy is used. To illustrate the present technique we choose the simplest trial wave function used in the VC method.

We choose for the ground-state trial wave function a modified hydrogen-like function which is

$$\psi_1 = (\lambda^3/7\pi)^{1/2} (1 + \lambda r) e^{-\lambda r}, \quad (13)$$

where λ is a variational parameter. The trial wave function Eq. (13) yields an effective Hamiltonian for the ground state. It is

$${}_1H_{\text{eff}} = \frac{\mathbf{p}^2}{2m} + \begin{cases} \frac{-Me^2}{d} + \frac{e^2}{R_0} \left(1 - \frac{1}{\epsilon_\infty}\right) & \text{for } r < R \\ \frac{e^2}{\epsilon^*} \left(\frac{5373}{25088} \lambda - \frac{1}{a}\right) - \frac{e^2}{\epsilon_\infty r} - \frac{e^2}{3\epsilon^* r} \\ + \frac{e^2}{\epsilon^*} \frac{e^{-2\lambda r}}{r} \left(1 + \frac{17}{28} \lambda r + \frac{23}{42} \lambda^2 r^2 + \frac{1}{7} \lambda^3 r^3\right) \end{cases} \quad \text{for } r > R. \quad (14)$$

The quantity a is twice the nearest-neighbor distance. Therefore, the ground-state energy E_{1s} is given by

$$E_{1s} = \langle \psi_1, {}_1H_{\text{eff}} \psi_1 \rangle. \quad (15)$$

Minimizing E_{1s} by varying the parameter λ , we find the energy level E_{1s} .

The trial wave function for the excited $2p$ state is chosen as

$$\psi_2 = \left(\frac{\beta^5}{\pi}\right)^{1/2} r e^{-\beta r} \cos\theta, \quad (16)$$

where θ is the polar angle of the position vector of the electron \mathbf{r} . Using the wave function of Eq. (16), the

effective Hamiltonian for this state is

$${}_2H_{\text{eff}} = \frac{\mathbf{p}^2}{2m} + \begin{cases} \frac{-Me^2}{d} + \frac{e^2}{R_0} \left(1 - \frac{1}{\epsilon_\infty}\right) & \text{for } r < R \\ \frac{e^2}{\epsilon^*} \left(\frac{81}{512} \beta - \frac{1}{a}\right) - \frac{e^2}{\epsilon_\infty r} + \frac{e^2}{\epsilon^* r} \\ \times e^{-2\beta r} \left(1 + \frac{5}{4} \beta r + \frac{11}{6} \beta^2 r^2 + \beta^3 r^3\right) \end{cases} \quad \text{for } r > R. \quad (17)$$

Then the $2p$ state energy is

$$E_{2p} = \frac{\beta^2 \hbar^2}{2m} + \left(\frac{-Me^2}{d} + \frac{e^2}{R_0} \left(1 - \frac{1}{\epsilon_\infty}\right) \right) \times \left\{ 1 - \frac{1}{3} e^{-2\beta R_0} (2\beta^4 R_0^4 + 4\beta^3 R_0^3 + 6\beta^2 R_0^2 + 6\beta R_0 + 3) \right\} - \frac{e^2}{6\epsilon_\infty} \beta e^{-2\beta R_0} (4\beta^3 R_0^3 + 6\beta^2 R_0^2 + 6\beta R_0 + 3) + \frac{e^2}{3\epsilon^*} \left(\frac{81}{512} \beta - \frac{1}{a}\right) e^{-2\beta R_0} (2\beta^4 R_0^4 + 4\beta^3 R_0^3 + 6\beta^2 R_0^2 + 6\beta R_0 + 3) + \frac{e^2}{\epsilon^*} \beta e^{-4\beta R_0} (A\beta^6 R_0^6 + B\beta^5 R_0^5 + C\beta^4 R_0^4 + D\beta^3 R_0^3 + E\beta^2 R_0^2 + F\beta R_0 + G). \quad (18)$$

The coefficients A, B, C, \dots, G are real numbers.

In this problem, the F -center electron in its ground-state polarizes the lattice so that it experiences a self-consistent potential which includes a potential due to lattice. The term self-consistent here means that the potential is determined by the electronic wave function and the wave function by the potential. In addition, the frequency concerned in the optical transition is very much higher than that of the longitudinal optical mode. Therefore, during the optical transition (from the ground state to the higher state), the lattice does not change. In other words, the optical transitions occur according to the Franck-Condon principle. Once the optical transition is made, the electronic wave function is no longer a ground-state function so the lattice begins to relax. In view of these facts, for the actual optical transition case the effective Hamiltonian for the excited electron is the same as for the ground state, namely, Eq. (14). The "optical" $2p$ energy level to which the $1s \rightarrow 2p$ transition is just made is, therefore, different from that calculated by Eq. (18) in which lattice relaxation is involved. We denote the energy of the "optical" $2p$ state as ${}_oE_{2p}$. It is given by

$${}_oE_{2p} = \langle \psi_2, {}_1H_{\text{eff}} \psi_2 \rangle. \quad (19)$$

In the expression for the energy level of the ${}_oE_{2p}$, the

⁶ See Ref. 2, p. 58.

TABLE I. Energy levels and constants for an F center.

	m/m_e	$d(\text{\AA})$	M	ϵ_∞	ϵ_s	E'_{1s} (eV)	E'_{2p} (eV)	${}_oE'_{2p}$ (eV)	$-e^2/\epsilon^*a$ (eV)	${}_o[\Delta E_{1s-2p}]_{\text{th}}$ (eV)	$[\Delta E_{1s-2p}]_{\text{obs}}$ (eV)
NaCl	1	2.81	1.74756	2.25	5.62	-2.60	-0.49	-0.35	-0.69	2.25	2.70
KCl	1	3.14	1.74756	2.13	4.68	-2.31	-0.52	-0.39	-0.52	1.92	2.30

factor corresponding to $[(81/512)\beta - 1/a]$ in Eq. (18) is $[(5373/25088)\lambda - 1/a]$.

In Sec. IV we shall show that $-e^2/\epsilon^*a$ is the energy of the lowest state of the conduction band. Thus, the energy level with respect to the bottom of the conduction band is $E' = E + e^2/\epsilon^*a$. The theoretical results for E'_{1s} , E'_{2p} , and ${}_oE'_{2p}$, as calculated for NaCl and KCl, are summarized in Table I. The values of the constants used are also given. The calculated energy difference between the optical $2p$ state and the ground state, ${}_o[\Delta E_{1s-2p}]_{\text{th}}$, is given and compared with the experimentally observed value $[\Delta E_{1s-2p}]_{\text{obs}}$.

IV. HIGHER STATES OF AN F CENTER

The $3p$ state of an F center is one of the interesting higher states. The trial wave function ψ_3 for this state is chosen as

$$\psi_3 = (2\lambda^5/3\pi)^{1/2}(2 - \lambda r)re^{-\lambda r} \cos\theta. \quad (20)$$

Using the effective Hamiltonian given by Eq. (14) and the wave function of Eq. (20) we get the optical energy level ${}_oE_{3p}$, corresponding to the $3p$ state. The theoretical results of ${}_oE_{3p}$ and E_{1s} for KCl and NaCl are given in Table II. The theoretical transition energies for the $1s-3p$ transition of the F center are also given and compared with the experimental frequency (expressed as transition energy) of the K band in KCl. In view of this calculation, the K band may be associated with a transition from the ground state of an F center to its $3p$ state.

The energy level of the ground state or E_{2p} of the F center has the asymptotic value $-e^2/\epsilon^*a$, if the variational parameter approaches zero. A similar behavior is to be expected for the higher energy states. Now in the highest state we expect diffuse wave function corresponding to a small value of the variational parameter in the wave function. Hence, the energy of the highest true bound state (i.e., the highest state after lattice relaxation) approaches the asymptotic value $-e^2/\epsilon^*a$ as a limit of the energy-level series for the true bound-state case.

TABLE II. Energy levels for $1s$ and $3p$ states of the F center.

	E_{1s} (eV)	${}_oE_{3p}$ (eV)	${}_o[\Delta E_{1s-3p}]_{\text{th}}$ (eV)	$\Delta E(K \text{ band})$ (eV)
KCl	-2.83	-0.47	2.36	2.71
NaCl	-3.29	-0.37	2.92	...

For KCl and NaCl the values of these limits are, respectively, 0.52 and 0.69 eV. These are the energies of the lowest state of the conduction band. This is reasonable since when the variational parameter λ in the trial wave function is very small, the factor $e^{-\lambda r}$ approaches unity which is the same as the exponential factor for a plane-wave electron of zero momentum. In fact, for the highest true bound state in which the electron is far away from the negative-ion vacancy, the Coulomb interaction in Eq. (5) is small compared with the electron-phonon interaction; the term V_0 may not be considered important. Hence, the effective Hamiltonian becomes the polaron Hamiltonian plus a small perturbation $-e^2/\epsilon r$, that is,

$$H = \frac{\mathbf{p}^2}{2m} + \sum_k \hbar\omega_k a_k^* a_k + \sum_k (V_k a_k e^{i\mathbf{k}\cdot\mathbf{r}} + V_k^* a_k^* e^{-i\mathbf{k}\cdot\mathbf{r}}) - \frac{e^2}{\epsilon r}. \quad (21)$$

Here the last two terms, i.e., the interactions H_{ci} and H_{ec} in Eq. (5) are replaced by $-e^2/\epsilon r$. The effective dielectric constant ϵ is larger than ϵ_∞ due to H_{ci} contributing a small positive energy. Thus, we picture the highest true bound state of an F center as lying very close to the bottom of the conduction band, but never above it.

On the other hand, for the actual optical transition case, the energy of the highest state approaches the asymptotic value $(e^2/\epsilon^*)[(5373/25088)\lambda - 1/a]$, where λ is a variational parameter corresponding to the ground state. For KCl and NaCl, these asymptotic values are almost the same. It is approximately +0.1 eV with respect to the zero energy, i.e., the optical highest states for KCl and NaCl lie 0.62 and 0.79 eV above the bottom of the conduction band, respectively.

V. DISCUSSION

Our predictions for the $1s$ and $2p$ energy level underestimate the F -band transition energy. This is the same as previous works in the variational method, for example, Simpson's work.¹ He also uses a self-consistent method to consider the lattice interaction with an F -center electron in different way.

Probably the underestimation of the F -band transition energy arises mainly from the ground-state wave function used in this calculation. Since the potential energy of an F -center electron is Coulomb-like only for the electron at distances greater than R_0 , the modified hydrogen-like trial wave function is not a good approxi-

mation for the ground state. In view of the previous studies,^{5,7} the use of a Bessel-like wave function for $r < R_0$ and a modified hydrogen-like function for $r > R_0$ may lead to a ground-state energy lying below the one predicted by using just a hydrogen-like function.

For the excited states, the modification of the Bessel-like wave function leads to a negligible change in the energy levels, since the F -center electron spends most of its time in the Coulomb-like potential region. Hence, the choice of a hydrogen-like function for the $2p$ or the other higher state is expected to be a good approximation.

Consequently, the approach to the problem of the trapped electron, which includes phonons and phonon-electron interactions does raise the predicted position

⁷ J. A. Krumhansl and N. Schwartz, Phys. Rev. **89**, 1154 (1953).

of the energy level of a trapped electron relative to previous calculations and can bring the position of the energy level into line with recent experiment,⁸ due to a small quantity of positive energy contributed by the phonons.

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⁸ R. K. Swank and F. C. Brown, Phys. Rev. Letters **8**, 10 (1962).

Field and Angular Dependence of Critical Currents in Nb₃Sn*

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Measurements have been made on the critical current of Nb₃Sn vapor-deposited strips on ceramic material. The Nb₃Sn was obtained from vapor deposition and is single phase without any preferred orientation in the plane of the strip. Critical currents were obtained at 4.2°K as a function of field up to 20 000 G, and as a function of the angle between the field and the current axis. Below 10 000 G a sharp rise in critical current by about a factor of two was observed for longitudinal fields, and a gradual decrease in critical current for transverse fields. Above 10 000 G the field and angular dependence is in good agreement with the predictions of the Lorentz force model of Kim *et al.* Below 10 000 G, the field dependence can be associated with a transition from an inhomogeneous to homogeneous current distribution with consequent lower local critical current densities. Experimental results are also reported for field shielding measurements on similar material in cylindrical form. Excellent agreement is obtained in both magnitude and field dependence with the strip data.

THAT the magnetic behavior of hard superconducting tubes in a longitudinal field H , can be described in terms of a single model depending on the concept of a critical state,¹ and a field-dependent critical current density, j_c , where

$$j_c = \alpha / (H + B_0), \quad (1)$$

has been shown by Kim, Hempstead, and Strnad.² In (1), α and B_0 are constants depending on the tempera-

ture and the properties of the medium. Anderson³ has shown that (1) can be obtained in terms of flux penetration through the specimen^{4,5} and the Lorentz force exerted on a flux bundle by the current. When a pinning force on a flux bundle is exceeded, either the phenomena of flux creep or quenching can occur.^{1,6}

In the magnetization experiments of Kim *et al.*² the current and field are perpendicular to each other, and the current is internally generated. An important question concerns the extension of the above ideas to linear geometries where current is externally supplied. Apart from the practical significance of these geometries there

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¹ C. P. Bean, Phys. Rev. Letters **8**, 250 (1962).

² Y. B. Kim, C. F. Hempstead, and A. R. Strnad, Phys. Rev. **129**, 528 (1963).

³ P. W. Anderson, Phys. Rev. Letters **9**, 309 (1962).

⁴ A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [translation: Soviet Phys.—JETP **5**, 1174 (1957)].

⁵ B. B. Goodman, IBM J. Res. Develop. **6**, 63 (1962).

⁶ C. J. Gorter, Nuovo Cimento **6**, Suppl. 3, 1168 (1957).