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Treatment of the relaxed F center in KCl with the recursion method

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Ham's vibronic model for the relaxed excited state of the F center in KCl is investigated with a new approach based on the recursion method. We have calculated the energy and the wave function of the ground vibronic state and the lowest excited state, the dichroism in light emission in the presence of external fields (magnetic field, electric field, uniaxial stress), and the temperature dependence of the polarization induced by the electric field and by the uniaxial stress. Ham's vibronic model is found to account satisfactorily for the above properties. The perspectives of the recursion procedure for studying coupled electron-phonon systems are discussed.

I. INTRODUCTION

In the last few years there has been considerable progress in the theoretical treatment of systems constituted by electrons coupled to boson fields.[1–3] This progress is related to the development of techniques for the calculation of correlation functions and spectra, such as the recursion method[4,5] and the closely related memory-function methods.[1] Although quite promising, these methods have not yet been applied to the vibronic model of relaxed defects. In this paper we show the convenience and the advantages of studying the relaxed excited states of color centers by the recursion procedure with appropriate implementations.

As a specific example we consider explicitly the relaxed F center in KCl. We study the relaxed excited state (RES) and we calculate the energy and the wave function of the ground vibronic state and the lowest excited state. We can thus discuss the dichroism in the light emission induced by external fields and the influence of the temperature, and compare our results with those based on a perturbation approach[6,7] or symmetry treatment[8] of angular momenta.

In our calculation we consider the simplest vibronic model of the RES. Basically this model consists of an electronic $2s$ orbital interacting via a vibrational mode of symmetry Γ_4^- with threefold $2p$ orbitals of different energy. The model was proposed originally by Bogan,[9] and its extreme consequences were investigated by Ham and Grevsmühl.[7]

Later, Kayanuma and co-workers[10,11,13] and Iwahana, Iida, and Ohkura[12] proposed more complicated models which also included the Jahn-Teller interaction between $2p$ levels[10–12] or vibronic coupling among higher excited states[13] ($3s$, $3p$, $3d$). Our approach based on the recursion method could also allow in this case a nonperturbative treatment of the problem. However, at this stage of theoretical and experimental development, we consider only the model of Bogan; more complicated models should be corroborated by detailed band-structure calculations and a quantitative analysis of the deep defects corresponding to an anion vacancy.

II. THE MODEL HAMILTONIAN AND THE RECURSION METHOD

The most general situation of a relaxed excited state can be described by a Hamiltonian of the type

$$\mathcal{H}=\mathcal{H}_e+\mathcal{H}_L+\mathcal{H}_{e\text{-}L}+\mathcal{H}_{\text{ext}} , \tag{1}$$

where the terms are, respectively, the electronic Hamiltonian, the lattice Hamiltonian, the electron-lattice coupling, and the interaction with external fields.

In the vibronic model of interest here, the expression for the various terms appearing in Eq. (1) is simplified as follows. The electron Hamiltonian is taken to be

$$\mathcal{H}_e=E_s\,|\,s\,\rangle\langle\,s\,|+E_p(\,|\,p_x\,\rangle\langle\,p_x\,| + |\,p_y\,\rangle\langle\,p_y\,|+|\,p_z\,\rangle\langle\,p_z\,|\,) , \tag{2}$$

where E_s and E_p denote the orbital energy of the $2s$ and $2p$ wave functions of the F center in the relaxed configuration. We notice, however, that with a trivial extension it is possible to modify Eq. (2) in order to account for other wave functions and spin-orbit coupling, whenever necessary.

We turn now to the lattice Hamiltonian and to the electron-lattice coupling. We assume that only one lattice mode, namely the longitudinal-optical mode of symmetry Γ_4^-, is relevant and couples linearly the $2s$ and $2p$ states. We can thus write for $\mathcal{H}_L$

$$\mathcal{H}_L=\tfrac{3}{2}\hbar\omega_{\text{LO}}+\hbar\omega_{\text{LO}}(a_x^\dagger a_x+a_y^\dagger a_y+a_z^\dagger a_z) , \tag{3}$$

where $\hbar\omega_{\text{LO}}$ is the energy of the longitudinal-optical mode and a_i and $a_i^\dagger$ ($i=x,y,z$) are the creation and annihilation operators of symmetry x,y,z of the longitudinal mode.

The electron-phonon coupling interaction $\mathcal{H}_{e\text{-}L}$ is in the linear approximation

$$\mathcal{H}_{e\text{-}L}=\sum_{i=x,y,z}\gamma(a_i+a_i^\dagger)(\,|\,p_i\,\rangle\langle\,s\,|+|\,s\,\rangle\langle\,p_i\,|\,) , \tag{4}$$

where γ is a coupling constant related to the coupling energy E_G encountered in the literature via the relation

$$\gamma=(E_G\hbar\omega_{\rm LO})^{1/2}=S^{1/2}\hbar\omega_{\rm LO}\ .$$

It is convenient to represent the Hamiltonian (3) and (4) on the basis set

$$|\phi_i;lmn\rangle\ , \tag{5}$$

where $i=s,x,y,z$ and l,m,n are integer numbers (positive or zero), giving the phonon occupation numbers. We have

$$\mathscr{H}_L=\tfrac{3}{2}\hbar\omega_{\rm LO}+\sum_{\substack{l,m,n\\ i=s,x,y,z}}\hbar\omega_{\rm LO}(l+m+n)\,|\phi_i;lmn\rangle\langle\phi_i;lmn|\ , \tag{6}$$

$$\begin{aligned}\mathscr{H}_{e\text{-}L}=\gamma\sum_{l,m,n}&\sqrt{l}\,(|p_x;l-1\,m\,n\rangle\langle\phi_s;lmn|+|\phi_s;l-1\,m\,n\rangle\langle p_x;lmn|)\\&+\sqrt{l+1}\,(|p_x;l+1\,m\,n\rangle\langle\phi_s;lmn|+|\phi_s;l+1\,m\,n\rangle\langle p_x;lmn|)\\&+\sqrt{m}\,(|p_y;l\,m-1\,n\rangle\langle\phi_s;lmn|+|\phi_s;l\,m-1\,n\rangle\langle p_y;lmn|)\\&+\sqrt{m+1}\,(|p_y;l\,m+1\,n\rangle\langle\phi_s;lmn|+|\phi_s;l\,m+1\,n\rangle\langle p_y;lmn|)\\&+\sqrt{n}\,(|p_z;l\,m\,n-1\rangle\langle\phi_s;lmn|+|\phi_s;l\,m\,n-1\rangle\langle p_z\,|\,lmn|)\\&+\sqrt{n+1}\,(|p_z;l\,m\,n+1\rangle\langle\phi_s;lmn|+|\phi_s;l\,m\,n+1\rangle\langle p_z;lmn|)\ .\end{aligned} \tag{7}$$

The representation of the operator $\mathscr{H}$ in the basis set (5) is very simple and of sparse form, and combined with the recursion method it allows us to include a large number of phonons. For instance, a basis set of about 5000 states allows inclusion of at least ten phonons for each direction. There is, however, no need to diagonalize explicitly such large matrices. In fact, since the matrix is sparse we can use the recursion method, basically in the form given by Lanczos,[14] and we can transform the initial and large sparse matrix into a tridiagonal one, whose dimension is the number of iterations performed. It is well known that the eigenvalues of the small tridiagonal matrix converge rapidly toward the eigenvalues of the large matrix, whose eigenstates have relevant overlap with the starting state of interest. Typically in the present problem, a tridiagonal matrix of order 20 was more than sufficient to provide the convergence to the exact eigenvalues with a precision better than 10^{-8}, in the energy range of interest. Furthermore, it is possible to determine the coefficients of the states entering the vibronic state of interest by exploiting a simple property inherent in the algebraic transformations of the Lanczos approach;[14] in fact, such coefficients for a given starting initial state can be calculated (within an inessential phase factor) from the diagonalization of the small tridiagonal matrix.

For convenience we give explicitly the recursion procedure for our model Hamiltonian. Let $|f_0\rangle,|f_1\rangle,\ldots$ indicate the $\nu+1$ normalized functions of the recursion hierarchy; let $|F_{\nu+1}\rangle$ be the (unnormalized) function defined as

$$|F_{\nu+1}\rangle=\mathscr{H}|f_\nu\rangle-a_\nu|f_\nu\rangle-b_\nu|f_{\nu-1}\rangle\ . \tag{8}$$

The next pairs of parameters $b_{\nu+1}$ and $a_{\nu+1}$ are given by the normalization and by the expectation value of the Hamiltonian on the state $|F_{\nu+1}\rangle$, namely

$$\begin{aligned}b^2_{\nu+1}&=\langle F_{\nu+1}|F_{\nu+1}\rangle\ ,\\ a_{\nu+1}&=\frac{\langle F_{\nu+1}|\mathscr{H}|F_{\nu+1}\rangle}{\langle F_{\nu+1}|F_{\nu+1}\rangle}\ .\end{aligned} \tag{8a}$$

After determining $b^2_{\nu+1}$, we construct the normalized function $|f_{\nu+1}\rangle$ of the recursion hierarchy as

$$|f_{\nu+1}\rangle=|F_{\nu+1}\rangle/b_{\nu+1}$$

or, equivalently,

$$b_{\nu+1}|f_{\nu+1}\rangle=|F_{\nu+1}\rangle\ .$$

In the case of our vibronic model

$$|F_\nu\rangle=\sum_{\substack{j=1-4\\ l,m,n}}c^{(\nu)}_{jlmn}|\phi_j;lmn\rangle\ , \tag{9}$$

where $j=1,2,3,4$ stands for s,x,y,z, respectively. Using Eqs. (8), (8a), and (9), we obtain the following explicit recursion relation for the coefficients $c^{(\nu)}_{jlmn}$:

$$\begin{aligned}
c_{1lmn}^{(\nu+1)}=\{&c_{1lmn}^{(\nu)}[E_s+(l+m+n+\tfrac{3}{2})\hbar\omega_{\mathrm{LO}}]\\
&+\gamma(\sqrt{l+1}c_{2\,l+1\,m\,n}^{(\nu)}+\sqrt{l}c_{2\,l-1\,m\,n}^{(\nu)}+\sqrt{m+1}c_{3\,l\,m+1\,n}^{(\nu)}\\
&\quad+\sqrt{m}c_{3\,l\,m-1\,n}^{(\nu)}+\sqrt{n+1}c_{4\,l\,m\,n+1}^{(\nu)}+\sqrt{n}c_{4\,l\,m\,n-1}^{(\nu)})\}/b_\nu-\frac{a_\nu}{b_\nu}c_{1lmn}^{(\nu)}-\frac{b_\nu}{b_{\nu-1}}c_{1lmn}^{(\nu-1)}\ ,\\
c_{2lmn}^{(\nu+1)}=\{&\gamma(\sqrt{l+1}c_{1\,l+1\,m\,n}^{(\nu)}+\sqrt{l}c_{1\,l-1\,m\,n}^{(\nu)})+c_{2lmn}^{(\nu)}[E_p+(l+m+n+\tfrac{3}{2})\hbar\omega_{\mathrm{LO}}]\}/b_\nu-\frac{a_\nu}{b_\nu}c_{2lmn}^{(\nu)}-\frac{b_\nu}{b_{\nu-1}}c_{2lmn}^{(\nu-1)}\ ,\\
c_{3lmn}^{(\nu+1)}=\{&\gamma(\sqrt{m+1}c_{1\,l\,m+1\,n}^{(\nu)}+\sqrt{m}c_{1\,l\,m-1\,n}^{(\nu)})+c_{3lmn}^{(\nu)}[E_p+(l+m+n+\tfrac{3}{2})\hbar\omega_{\mathrm{LO}}]\}/b_\nu-\frac{a_\nu}{b_\nu}c_{3lmn}^{(\nu)}-\frac{b_\nu}{b_{\nu-1}}c_{3lmn}^{(\nu-1)}\ ,\\
c_{4lmn}^{(\nu+1)}=\{&\gamma(\sqrt{n+1}c_{1\,l\,m\,n+1}^{(\nu)}+\sqrt{n}c_{1\,l\,m\,n-1}^{(\nu)})+c_{4lmn}^{(\nu)}[E_p+(l+m+n+\tfrac{3}{2})\hbar\omega_{\mathrm{LO}}]\}/b_\nu-\frac{a_\nu}{b_\nu}c_{4lmn}^{(\nu)}-\frac{b_\nu}{b_{\nu-1}}c_{4lmn}^{(\nu-1)}\ .
\end{aligned}\tag{10}$$

These relations, for any given starting state, allow one to construct a small-order tridiagonal matrix, which is then diagonalized using standard programs.

To give an idea of the reliability of the method used, we have verified that about 20 iterations in a cluster containing up to 20 phonons in each direction provide an accuracy of better than 10^{-8} for the eigenvalues of interest. This precision is almost 3 orders of magnitude better than what was really necessary. For actual calculations we have used clusters whose size was checked to be more than sufficient for our purpose.

In performing the calculations, we take $E_p - E_s = 90$ meV as assumed by Ham and Grevsmühl.[7] In a theoretical estimation of Wood and Öpik,[15] the difference $E_p - E_s$ of the relaxed F center in KCl is calculated to be 80 meV. Because of some uncertainty in the theoretical treatment, and in order to compare our new method with the perturbative procedure of Ham, we have taken the value of 90 meV. Similarly, the value of $\hbar\omega_{\mathrm{LO}}$ is taken to be 26.8 meV, and the coupling constant $\gamma = 19.4$ meV.

In Table I we report our calculations in the absence of external fields. The most significant physical quantities are δE, the difference between the first-excited vibronic state and the ground state, and $\tau_r(0)/\tau_r(p)$, the ratio between the radiative lifetimes of the ground vibronic state and the electronic state $2p$. Finally, we also give the ratio $R = \tau_r(0)/\tau_r(1)$ concerning the ratio of the radiative lifetimes of the ground vibronic state and the first-excited state. A comparison with the previous results of Ham shows that a nonperturbative approach is desirable, even if the relaxed-F-center problem in KCl falls in the intermediate-coupling region.

TABLE I. Comparison between our results and the calculations of Ham and Grevsmühl (Ref. 7) in the absence of external fields. The energies are in meV.

KCl	Recursion method[a]	Perturbative method[b]
δE	19.37	17.65
$\tau_r(0)/\tau_r(p)$	12.3	12.1
$\tau_r(0)/\tau_r(1)$	1.73	2.4

[a] This work.
[b] Reference 7.

III. EXTERNAL PERTURBATION EFFECTS

A. Effect of a magnetic field

We now study the effects of external perturbations on the vibronic model outlined in the preceding section. In order to consider the effect of the magnetic field, it is convenient to take as basis functions for the electron Hamiltonian, besides $|p_z\rangle$, the states

$$|p_+\rangle = -\frac{1}{\sqrt{2}}(|p_x\rangle + i|p_y\rangle)$$

and

$$|p_-\rangle = \frac{1}{\sqrt{2}}(|p_x\rangle - i|p_y\rangle) .$$

The relevant part of the Hamiltonian due to the presence of the magnetic field H is thus

$$\mathcal{H}_m = g_L \mu_B H(|p_+\rangle\langle p_+| - |p_-\rangle\langle p_-|) .$$

We have calculated the induced circular polarization neglecting spin-orbit terms:

$$P_m(0) = \frac{I_+(0) - I_-(0)}{I_+(0) + I_-(0)} . \tag{11}$$

We find a satisfactory agreement with the experimental data of Fontana and also of Baldacchini *et al.*[16] In particular, expression (11) turns out to be linear in H; with the value $g_L = 1$ we obtain

$$P_m(0) = -(8.9\times 10^{-8})H \quad (H \text{ in G}) .$$

The experimental results[16] of the circularly-polarized dichroism and the previous theoretical results[7] of Ham are summarized in Table II. Incidentally, we remark that the experimental situation is still somewhat uncertain[17] and more accurate experiments would be desirable.

B. External electric field

In the presence of an external electric field, say in the z direction, the interaction Hamiltonian becomes

$$\mathcal{H}_E = eEz ,$$

where E is the local electric field and e is the absolute value of the electronic charge. Because of this interaction

TABLE II. Comparison between our results, the calculations of Ham, and the experimental results in the presence of external fields (magnetic, electric, and uniaxial stress).

KCl	Recursion method[a]	Perturbative method[b]	Expt. results
$P_m(0)$	$-(8.9\times10^{-8})H$	$-(10.2\times10^{-8})H$	$-(9\pm1)\times10^{-8}H$[c] $-(11\pm1)\times10^{-8}H$[c] $-(16\pm1)\times10^{-8}H$[d]
$P_E(0)$	$(4.14\times10^{-7})\alpha^2F^2$	$(2.24\times10^{-7})\alpha^2F^2$	$(1.0\pm0.3)\times10^{-5}F^2$[e]
$P_{\text{stress}}(0)/X$	5.4×10^{-3} (kg/mm^2)$^{-1}$	5.1×10^{-3} (kg/mm^2)$^{-1}$	9.9×10^{-3} (kg/mm^2)$^{-1}$[f] $(2.27\pm0.13)\times10^{-3}$[g]

[a] This work.
[b] Reference 7.
[c] Reference 16.
[d] Reference 17.
[e] Reference 9.
[f] Reference 20.
[g] Reference 21.

the states $2s$ and $2p$ are coupled regardless of the vibronic interaction, the state $2s$ being coupled with the state $2p_z$. We can thus write the additional term due to the presence of an electric field in the form

$$\mathscr{H}_E = D\cdot E(\,|s\rangle\langle p_z| + |p_z\rangle\langle s|\,)\ ,$$

where

$$D\cdot E = \langle s\,|\,eEz\,|\,p_z\rangle\ .$$

The change in energy of the ground vibronic state is found to be negative and quadratic in $D\cdot E$.

The polarization induced by the electric field is given by

$$P_E(0) = \frac{I_z(0) - I_x(0)}{I_z(0) + I_x(0)}\ .$$

This is positive and quadratic in $D\cdot E$. For convenience the dipole D is measured units of a dipole with length 1 Å; the dimensionless parameter is indicated with α. We also assume that there is no difference between the local field and the bulk field. The results are given in Table II. The comparison with the experimental data gives for α a value between 4 and 5, as predicted by Bogan and Fitchen.[9]

We have also calculated the temperature dependence of the polarization induced by the electric field. We suppose the system to be in the condition such that $kT \ll \delta E$. Then we can write

$$\frac{P(T)}{P(0)} = \frac{1 + C(T)e^{-\delta E/kT}}{1 + 3Re^{-\delta E/kT}}\ ,$$

where δE is the difference between the ground and the excited vibronic state (in the absence of a perturbation), $C(T)$ is only slightly dependent on temperature, and R has been defined in Sec. II. It is possible to show that the induced polarization decreases with the temperature and can be approximated with an exponential of the form

$$\frac{P(0)}{P(T)} - 1 \sim Ae^{-\beta/T}\ ,$$

with $\beta = 227$ K, $A = 3.8 = 3R - C(T)$; this behavior is again in good agreement with that of Bogan and Fitchen.[9]

C. Uniaxial stress

Finally, we have also considered with our procedure the effect of a uniaxial stress on the luminescence of the relaxed F center. Suppose that a uniaxial stress X is applied in the z direction of the crystal. This gives a deformation

$$\epsilon_\theta = X/(c_{11} - c_{12})\ ,$$

where c_{11} and c_{12} are the elastic constants of the isotropic solid. The most important effect of the uniaxial stress is to lift the degeneracy of the $2p$ levels. In the presence of a uniaxial stress we must include in our formalism the additional Hamiltonian

$$\mathscr{H}_{\text{stress}} = V\epsilon_\theta(-\,|p_z\rangle\langle p_z| + \tfrac{1}{2}\,|p_x\rangle\langle p_x| + \tfrac{1}{2}\,|p_y\rangle\langle p_y|\,)\ ,$$

where $3V\epsilon_\theta/2$ is the energy difference of the states p_x and p_z due to the stress. It is possible to obtain the polarization and its dependence on the temperature via the standard tridiagonalization procedure used throughout this paper. If we take for V the value that can be deduced by the experiments of Schnatterly[18] and Hetrick,[19] we obtain

$$P_{\text{stress}}(0) = (5.4\times10^{-3})X \quad (X \text{ in } +\text{kg/mm}^2)\ .$$

This must to be compared with a value of $(5.1\times10^{-3})X$ calculated by Ham[7] and with an experimental value of $(9.9\times10^{-3})X$ of Hetrick and Compton[20] and $(2.27\times10^{-3})X$ of Akiyama *et al.*[21] We have also found that $P(T)$ increases slightly with the temperature, but this increase is less than 10% $[P_{\text{stress}}(T)/P_{\text{stress}}(0) - 1 \simeq 9\%$ per 100 K], in agreement with the experimental data.

IV. CONCLUSIONS

In this paper we have considered a nonperturbative procedure, based on the recursion method, for investigating the vibronic model of impurity centers. The procedure has been successfully applied to study the luminescence features of the F center in KCl, which is known to be in the intermediate-coupling situation. In this case, as shown in this paper, the differences between a perturbative and a nonperturbative approach are significant, but not dramatic. The major advantage of our procedure lies in the fact that it can be applied with about the same amount of labor to strongly coupled electron-phonon systems. Furthermore, other possible extensions of the vibronic model can be treated within the approach of the present paper. For instance, it would be possible to take into account quadratic terms in electron-phonon coupling (if relevant), to include other modes, or spin-orbit interaction, or to study more complicated centers such as F_A centers,[22,23] which are of large importance for color-center lasers.

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