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Positron annihilation from F centers of alkali halide crystals

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Two-photon annihilation of a positron from an F center in an alkali halide crystal has been studied theoretically by accurately solving with the integral transform (generator coordinate method) the two-particle (electron-positron) problem for two different model crystal potentials (an effective Coulomb potential and the Krumhansl-Schwarz cavity potential) which are often used in the theoretical description of F centers. Calculations are presented of the stability and relevant expectation values of the positron-F-center system, the two-photon annihilation lifetime τ and the angular correlation distribution $N(\theta)$ for sixteen different alkali halides. Comparison is made with available experimental data and previous, less accurate calculations. It is demonstrated that a proper description of electron-positron correlation in the wave function is *necessary* in order to obtain meaningful results. We have also established a range of effective charges, Z^*, of the defect that can capture an electron-positron pair in a stable state. For the Krumhansl-Schwarz potential, the present highly accurate calculations show that all sixteen crystals have stable electron-positron states in the defect.

I. INTRODUCTION

In recent years, a number of experimental investigations[1,2] have been made of the annihilation of positrons from alkali halide crystals which contain substantial concentrations of F color centers. Typically, a significant change relative to that of the pure alkali halide crystal is found in the angular distribution of two-photon annihilation $N(\theta)$, i.e., a new and narrow component in the low-momentum (small angle θ) region appears. This has been attributed to annihilation of positrons trapped by the defect with the F-center electron, an (Fe^+) complex or center.

Several theoretical studies[3–9] have been reported wherein calculations have been made using model defect potentials in the two-particle Schrödinger equation (defined in Sec. II). These variational calculations have employed trial wave functions which were either uncorrelated or simply correlated for the electron-positron motion. One problem with uncorrelated and simply correlated treatments of the hydrogenic model potential is that with the exception of LiF, this model potential leads to (Fe^+) complexes that are unstable[7,8] with respect to free positronium [i.e., ground-state energies of the (Fe^+) complex which are greater than $E(\text{Ps})=-0.25$ hartree]. Thus in all these cases binding energies calculated relative to the ground-state energy of the F center are misleading.

Thus a major purpose of the present paper is to properly assess the role of interparticle correlation in the variational treatment of the problem, and in the calculation of the binding energy and the annihilation characteristics: the lifetime τ and the two-photon angular distribution $N(\theta)$.

Although experimental data are not available for all alkali halide systems, we shall present results for sixteen systems: LiH, LiF, LiCl, and LiI; NaF, NaCl, NaBr, and NaI; KF, KCl, KBr, and KI; and RbF, RbCl, RbBr, and RbI.

During the course of this work we have also investigated the problem of stable binding of an electron-positron pair to a Coulomb potential Z^*/r for a range of values of Z^*. It is well known[10] that for $Z^*=1$ the system is unbound. We shall show that for $0.45<Z^*<0.99$ the system becomes bound. This may be contrasted with the range ($0.7\leq Z^*\leq 0.9$) reported in earlier work[9] for a simply correlated wave function.

The definition of the problem and the methodological details can be found in Sec. II. The results are presented and discussed in Sec. III. Concluding remarks are made in Sec. IV.

Atomic units[11] are used throughout this paper except where otherwise indicated.

II. THEORETICAL AND COMPUTATIONAL CONSIDERATIONS

A. The Hamiltonian

The system under consideration is that of a normal F center plus a positron, an Fe^+ center. It consists of a positron-electron pair in the field of the defect. The Schrödinger equation to be solved for the (Fe^+) center problem is

$$H\psi(\mathbf{r}_+,\mathbf{r}_-)=E\psi(\mathbf{r}_+,\mathbf{r}_-) \tag{1}$$

with the Hamiltonian

$$H=-\tfrac{1}{2}\Delta_+-\tfrac{1}{2}\Delta_-+V(\mathbf{r}_+)-V(\mathbf{r}_-)-1/r_{+-}\ . \tag{2}$$

The potential $V(\mathbf{r})$ is the effective potential of an anion vacancy of the host alkali halide crystal. In the present study of S states, we assume that this potential is isotropic [$V(\mathbf{r})=V(r)$] and that the potential acting on the positron $V(r_+)$ and the electron $V(r_-)$ in the anion vacancy are of the same form.

Of the models proposed for the description of the F

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center,[12] two models for $V(r)$ have been considered in most studies in the literature[3–5,7–9] for the Fe^+ problem and will be employed here.

1. Hydrogenic model

This potential may be written in the form

$$V(r)=Z^*/r \ , \tag{3}$$

where the parameter Z^* is empirically determined. We have used the values of Z^* given in Ref. 7. They were determined by solving the F-center problem with this potential to give the $1s$-$2p$ transition energy, $\Delta E_{1s\text{-}2p}=3(Z^*)^2/8$, and choosing Z^* to reproduce the maximum of the experimental F band, presumably due to the $1s$-$2p$ transition. As a result

$$Z^*=(8\Delta E_{1s\text{-}2p}/3)^{1/2} \ . \tag{4}$$

The experimental F-band maxima tabulated by Zahrt and Lin[13] were used in Ref. 7 to determine the values of Z^*. They may be found in our Table I.

2. Krumhansl-Schwarz (Refs. 13–15) cavity model

The potential for this model is of the form

$$V(r)=\begin{cases} V_0, & r<R \\ (k_0 r)^{-1}, & r\geq R \end{cases} \tag{5}$$

where R is the radius of the cavity and k_0 is the static dielectric constant of the crystal. The parameters of Ref. 13 for the KS model were used in our study and are listed in Table I.

TABLE I. Potential parameters describing the interaction of e^+ or e^- with an anion vacancy in alkali halide crystals.

Hydrogenlike model[a]		Cavity model (Krumhansl-Schwarz)[b]		
Crystal	Z^*	R	k_0	V_0
LiH	0.485	3.859	3.61	0.3775
LiF	0.697	3.794	1.92	0.4152
LiCl	0.562	4.84	2.75	0.3137
LiI	0.478	5.67	3.80	0.2616
NaF	0.598	4.37	1.74	0.3696
NaCl	0.512	5.317	2.25	0.2947
NaBr	0.475	5.63	2.62	0.2739
NaI	0.455	6.11	2.91	0.2507
KF	0.516	5.03	1.85	0.3202
KCl	0.464	5.94	2.13	0.2684
KBr	0.439	6.24	2.33	0.2527
KI	0.421	6.65	2.69	0.2340
RbF	0.505	5.33	1.93	0.3016
RbCl	0.441	6.18	2.19	0.2572
RbBr	0.411	6.47	2.33	0.2444
RbI	0.396	6.92	2.63	0.2261

[a] Due to Farazdel and Cade (Ref. 7).

[b] Due to Zahrt and Lin (Ref. 13).

B. Computational method

In our calculations, we used the method known in atomic, molecular, and related problems[16–18] as the integral-transform method and in nuclear physics as the generator-coordinate method.[19]

Given the problem of finding the eigenfunctions of an N-particle system with Hamiltonian H, the basic idea of the method is to systematically generate trial functions ψ by the prescription:

$$\psi(x_1,x_2,\ldots,x_N)=\psi(\mathbf{x}_N)=\int_{D_M} S(\mathbf{t}_M)\Phi(\mathbf{x}_N;\mathbf{t}_M)d\mathbf{t}_M \ , \tag{6}$$

where D_M is an M-dimensional integration domain for the parameter $(\mathbf{t})$ space, Φ is some known function (e.g., an exact eigenfunction for some model Hamiltonian), and the weight or shape function $S(\mathbf{t}_M)$ is to be determined. We shall assume that ψ, Φ, and S are all real.

Insertion of the ansatz (6) into the variational principle and variation with respect to $S(\mathbf{t}_M)$ yields a Fredholm-type integral equation which may be solved by approximate numerical integration. This produces the secular equations:

$$\sum_{i=1}^{L} W_i S(\mathbf{t}_i)[K(\mathbf{t}_i;\mathbf{t}'_j)-EI(\mathbf{t}_i;\mathbf{t}'_j)]=0 \tag{7}$$

for $j=1,2,\ldots,L$, where

$$I(\mathbf{t}_i;\mathbf{t}'_j)=\int d\mathbf{x}_N[\Phi(\mathbf{x}_N;\mathbf{t}_i)\Phi(\mathbf{x}_N;\mathbf{t}'_j)]_{\mathrm{sym}} \ , \tag{8}$$

$$K(\mathbf{t}_i;\mathbf{t}'_j)=\int d\mathbf{x}_N[\Phi(\mathbf{x}_N;\mathbf{t}_i)\hat{H}\Phi(\mathbf{x}_N;\mathbf{t}'_j)]_{\mathrm{sym}} \ . \tag{9}$$

$[f\hat{O}g]_{\mathrm{sym}}=f\hat{O}g+g\hat{O}f$, and hence the Hamiltonian kernel K and the overlap kernel I are Hermitian.

The W_i and $\mathbf{t}_i$ are weights and abscissas, respectively, for the numerical integration. We choose the sets $\{\mathbf{t}_i\}$ and $\{\mathbf{t}'_j\}$ to coincide, obtaining therefore a convergent series of upper bounds to the true energy.[17]

As we are interested in the ground state of the (Fe^+) system, we choose $\Phi(\mathbf{x}_N;\mathbf{t}_M)$ to be

$$\Phi(r_+,r_-,r_{+-};\alpha,\beta,\gamma) =(4\pi)^{-1}\exp(-\alpha r_+ -\beta r_- -\gamma r_{+-}) \ . \tag{10}$$

This choice of Φ corresponds to the variational ansatz:

$$\psi(r_+,r_-,r_{+-})=(4\pi)^{-1}\sum_{k=1}^{L} C_k\exp(-\alpha_k r_+ -\beta_k r_- -\gamma_k r_{+-}) \ . \tag{11}$$

In effect the nonlinear parameters (α,β,γ) are chosen to be the lattice points of a three-dimensional quadrature formula, and the linear coefficients are found by solving the secular equation.

Note that for the ansatz (11) all integrals required for the calculation of the energy and various other expectation values, and probability density functions, can be done analytically.[20–22]

It is evident that we may restrict our attention to finite values of α_k, β_k, and γ_k, and so we choose the integration domain of Eq. (6) to be a parallelotope in three space. Thus, we may write

$$\alpha_k \in [A_1, A_2], \quad \beta_k \in [B_1, B_2], \quad \gamma_k \in [G_1, G_2] \tag{12}$$

for $k = 1, 2, \ldots, L$. The parallelotope is not completely arbitrary. Since we are considering bound states, we must have

$$\min \beta_k > 0, \quad \min \gamma_k > 0 , \tag{13}$$

where the minimum is to be taken over all $k \in \{1, 2, \ldots, L\}$. We allow the α_k's to take on negative values for the same reasons as in atomic systems.[17,18,23,24]

In order to ensure that all of the required integrals exist, we impose the following constraints:

$$\min(\beta_k + \beta_l + \alpha_k + \alpha_l) > 0 , \tag{14a}$$

$$\min(\alpha_k + \alpha_l + \gamma_k + \gamma_l) > 0 , \tag{14b}$$

where the minimum in these constraints is to be taken over all $k \in \{1, 2, \ldots, L\}$ and all $l \in \{1, 2, \ldots, L\}$.

Thus, D_3 is chosen to be a parallelotope defined by Eq. (12), with A_1, A_2, B_1, B_2, G_1, and G_2 being variational parameters subject to the constraints of Eqs. (13) and (14).

Finally, it is necessary to choose a quadrature scheme for numerical integration of Eq. (7).[17] Monte Carlo methods[25,26] seem to be most appropriate because the number of quadrature points required for satisfactory accuracy is smaller than in traditional methods. We use one particular method[27] that was found to be quite successful.[17,18] In this method the quadrature points are pseudorandom numbers in the unit cube which may be mapped onto the required parallelotope by affine transformation. In this scheme the $3L$ nonlinear parameters are generated by the following equations:

$$\begin{aligned} \alpha_k &= (A_2 - A_1)\langle \tfrac{1}{2}k(k+1)\sqrt{2}\rangle + A_1 , \\ \beta_k &= (B_2 - B_1)\langle \tfrac{1}{2}k(k+1)\sqrt{3}\rangle + B_1 , \\ \gamma_k &= (G_2 - G_1)\langle \tfrac{1}{2}k(k+1)\sqrt{5}\rangle + G_1 \end{aligned} \tag{15}$$

for all $k = 1, 2, \ldots, L$. In the above $\langle x \rangle$ is defined to be the fractional part of x. Note that A_1, A_2, B_1, B_2, G_1, and G_2 are the variational parameters that define the parallelotope of Eq. (12).

In our calculations all overlap, energy and other integrals were computed with the recursion relationships given by Sack *et al.*[18,21,22] for the Z^*/r potential and with suitable modifications for the Krumhansl-Schwarz (KS) potential. The optimization of the variational parameters were carried out using Powell's algorithm[28] modified by the inclusion of a barrier function to handle the constraints given by Eqs. (13) and (14). All of the algorithms used for the solution of the secular equation were standard EISPACK routines.[29]

The present calculations were carried out on a Perkin-Elmer-3251 computer using a 64-bit mantissa. In the Appendix a study on the number of terms L required in the expansion of the wave function is reported and discussed. We note that in the atomic helium problem[17] this method yielded a truly compact and accurate wave function. With $L = 66$ and the analogous ansatz to Eq. (11), the ground-state energy was found to be $-2.903\,724\,363$ a.u. in comparison with the exact result of $-2.903\,724\,377$ a.u.

C. Other calculated quantities

The spin-averaged annihilation rate for two-photon annihilation, $\lambda_{2\gamma}$ (and the corresponding lifetime, $\tau_{2\gamma} = \lambda_{2\gamma}^{-1}$), is proportional to the electronic charge density ρ at the positron, i.e.,

$$\lambda_{2\gamma} = \pi \alpha^3 \rho , \tag{16}$$

where

$$\rho = \langle \psi(\mathbf{r}_+, \mathbf{r}_-) \,|\, \delta(\mathbf{r}_+ - \mathbf{r}_-) \,|\, \psi(\mathbf{r}_+, \mathbf{r}_-) \rangle \tag{17}$$

and α is the fine-structure constant.

The angular distribution $N(\theta)$ of the two emitted photons in the long-slit apparatus is

$$N(\theta) = \int \int |\chi(p_x, p_y, p_z = mc\theta)|^2 dp_x dp_y , \tag{18}$$

where $\mathbf{p} = (p_x, p_y, p_z)$ is the total momentum (in our case one positron and one F-center electron) of the annihilating pair or the two emitted photons and

$$\chi(\mathbf{p}) = \int \int \exp(-i\mathbf{p}\cdot\mathbf{r}_-)\delta(\mathbf{r}_+ - \mathbf{r}_-)\psi(\mathbf{r}_+, \mathbf{r}_-) d\mathbf{r}_+ d\mathbf{r}_- \tag{19}$$

is the probability amplitude of finding an electron-positron pair or the two emitted photons in the interval between $\mathbf{p}$ and $\mathbf{p} + d\mathbf{p}$. The full width at half maximum (FWHM) of $N(\theta)$ will be designated by Γ_N.

We have calculated the expectation values

$$\langle r_+ \rangle = \int r_+ \,|\, \psi(\mathbf{r}_+, \mathbf{r}_-) \,|^2 d\mathbf{r}_+ d\mathbf{r}_- , \tag{20}$$

$$\langle r_- \rangle = \int r_- \,|\, \psi(\mathbf{r}_+, \mathbf{r}_-) \,|^2 d\mathbf{r}_+ d\mathbf{r}_- , \tag{21}$$

and

$$\langle r_{+-} \rangle = \int r_{+-} \,|\, \psi(\mathbf{r}_+, \mathbf{r}_-) \,|^2 d\mathbf{r}_+ d\mathbf{r}_- \tag{22}$$

in order to characterize the distributions $|\psi(\mathbf{r}_+, \mathbf{r}_-)|^2$.

III. RESULTS AND DISCUSSION

We begin the discussion with our results obtained for hydrogenic potentials. In Table I are given the relevant potential parameters used in the calculations. The fully converged results for the lifetimes τ, the expectation values $\langle r_+ \rangle$, $\langle r_- \rangle$, $\langle r_{+-} \rangle$ along with the total energy $E(\mathrm{Fe}^+)$ are shown in Table II. Also shown are the binding energies of the positron, defined as $E_B(e^+) = E(F) - E(\mathrm{Fe}^+)$. $E(F)$, the energy of the F center, is equal to $-(Z^*)^2/2$ in this model. It should be pointed out that only for the eleven cases mentioned in Table II, the (Fe^+) system was found to be stable. This can be contrasted with the results of Refs. 7 and 8, wherein only LiF was found to be stable against dissociation into free positronium. In fact, at this stage it is quite interesting to examine the stability of a bound electron-positron pair in the effective hydrogenic potential Z^*/r. It is well known[10] that if $Z^* = 1$ the system is unbound. In general,

TABLE II. Results of the present calculations for the (Fe^+) complex with the hydrogenic model potential (Z^*/r). In the case of KBr, KI, RbCl, RbBr, and RbI, the complex (Fe^+) was found not to be bound with respect to free positronium, i.e., $-E(\mathrm{Fe}^+)<0.25$, and therefore, our results for these systems are not included in this tabulation.

Crystal	$\langle r_+ \rangle$	$\langle r_- \rangle$	$\langle r_{+-} \rangle$	$-E(\mathrm{Fe}^+)$	$-E_B(e^+)$[a] (eV)	$-E_B(\mathrm{Ps})$[b] (eV)	τ (nsec)
LiH	8.09	7.41	3.26	0.2522	3.663	0.060	0.5610
LiF	5.28	2.51	4.78	0.2992	1.532	1.339	1.118
LiCl	5.45	4.10	3.67	0.2614	2.816	0.310	0.6840
LiI	8.68	8.06	3.23	0.2517	3.742	0.046	0.5520
NaF	5.16	3.49	3.89	0.2685	9.747	0.503	0.7670
NaCl	6.58	5.67	3.39	0.2546	3.361	0.125	0.5949
NaBr	9.01	8.42	3.22	0.2516	3.774	0.044	0.5486
NaI	13.92	13.57	3.11	0.2505	4.060	0.014	0.5246
KF	6.44	5.49	3.41	0.2550	3.622	0.136	0.6006
KCl	10.63	10.18	3.15	0.2509	4.144	0.025	0.5311
RbF	7.11	6.30	3.33	0.2536	3.431	0.098	0.5790

[a] Binding energy of a positron to an F center, $E_B(e^+)=E(F)-E(\mathrm{Fe}^+)$.

[b] Binding energy of positronium to the anion vacancy is $E(\mathrm{Ps})-E(\mathrm{Fe}^+)=-0.25h-E(\mathrm{Fe}^+)$.

this system can be unstable with respect to the following channels: (1) into a defect and free positronium; (2) into a free positron and an electron bound to the defect, i.e., the F center. For the system to be stable it is necessary that its total energy be less than the energies of both of these channels. In Fig. 1 the behavior of $E(\mathrm{Fe}^+)$ as a function of Z^* is shown along with the free positronium energy (-0.25 a.u.) and that of an F center $[(-Z^*)^2/2]$. An important result that emerges from our study is that the system is stable for the range of Z^* values $0.45 \leqq Z^* < 1$. We found that for $Z^*=0.99$ the system is stable so that it can be conjectured that it will remain so for $Z^*=1-\delta$ for any small positive δ. Earlier calculations reported by Boeve and Arefev[9] gave the stability range as $0.7 \leqq Z^* \leqq 0.9$. In their calculations, they employed only a one-term expansion of the correlated wave function.

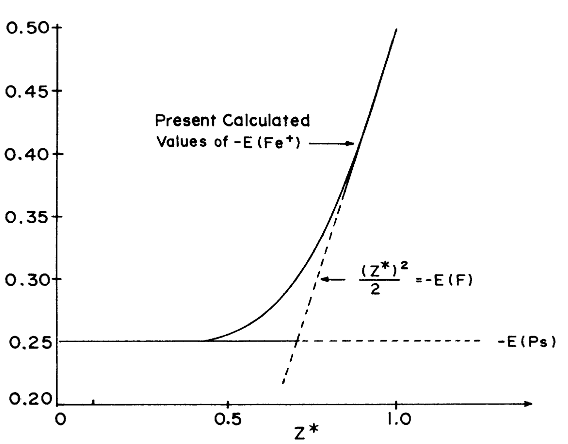

FIG. 1. Comparison of the present calculated values with the Z^*/r potential for the (Fe^+) complex, $E(\mathrm{Fe}^+)$, with those for the F center, $E(F)$.

More insight can be gained by examining Fig. 2(a), where the behavior of the expectation values $\langle r_+ \rangle$, $\langle r_- \rangle$, and $\langle r_{+-} \rangle$, is shown as a function of Z^*. The physical picture which emerges from these results is as follows. (1) In the limit $Z^* \to 1$ the system tends to behave like a free positron and an F center with $\langle r_+ \rangle$ and hence $\langle r_{+-} \rangle$ increasing rather sharply and $\langle r_- \rangle$ slowly approaching the hydrogenic limit $(Z^*=1)$. However, it should be noted that the total energy for $Z^*<1$ is always below the hydrogenic energy. (2) As Z^* decreases $\langle r_+ \rangle$ decreases and goes through a minimum at $Z^*=0.65$. With further decrease of Z^*, the system starts to approach the free positronium limit with $\langle r_{+-} \rangle$ decreasing to reach the $\langle r_{+-} \rangle$ value of free positronium. The total energy in our calculation is just below the free positronium energy (-0.25 a.u.) for $Z^*=0.45$. The lower limit has not been fully determined in our calculations since it is quite difficult to represent almost free positronium loosely attached to the defect center using our ansatz. As a result the sys-

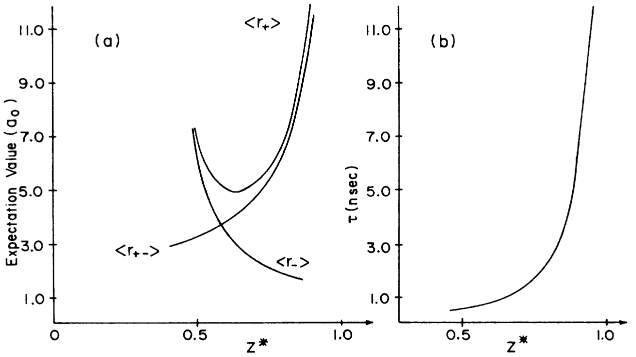

FIG. 2. (a) Behavior of $\langle r_+ \rangle$, $\langle r_- \rangle$, and $\langle r_{+-} \rangle$ with Z^* for the Z^*/r potential. (b) Behavior of the lifetime τ with Z^* for the Z^*/r potential.

TABLE III. Results of the present calculations for the (Fe^+) complex with the Krumhansl-Schwarz (cavity-model) potential.

	$\langle r_+ \rangle$	$\langle r_- \rangle$	$\langle r_{+-} \rangle$	$-E(Fe^+)$	$E_B(e^+)$[a] (eV)	$E_B(Ps)$[b] (eV)	τ (nsec)
LiH	6.12	3.40	5.00	0.296 56	2.638	1.2669	1.30
LiF	6.46	3.37	5.42	0.296 78	1.616	1.2729	1.31
LiCl	7.12	4.43	5.12	0.277 30	2.280	0.7429	0.98
LiI	7.98	6.08	7.15	0.273 81	2.841	0.6475	0.83
NaF	6.74	4.08	5.04	0.282 95	1.459	0.8966	1.11
NaCl	7.07	4.86	4.58	0.267 82	2.041	0.4849	0.83
NaBr	7.28	5.42	4.18	0.265 51	2.264	0.4220	0.77
NaI	7.92	6.29	4.05	0.261 71	2.495	0.3186	0.71
KF	7.05	4.90	4.48	0.271 00	1.682	0.5714	0.86
KCl	7.76	6.11	3.98	0.261 52	2.348	0.3135	0.70
KBr	7.86	6.36	3.90	0.260 34	2.348	0.2814	0.70
KI	10.14	7.82	6.85	0.264 45	2.721	0.3932	1.02
RbF	7.21	5.08	4.52	0.269 58	1.886	0.5328	0.73
RbCl	7.56	6.01	3.94	0.259 28	2.190	0.2525	0.68
RbBr	7.62	6.27	3.75	0.258 00	2.343	0.2177	0.67
RbI	8.35	7.13	3.64	0.255 76	2.571	0.1567	0.62

[a] Binding energy of a positron to an *F* center, $E_B(e^+)=E(F)-E(Fe^+)$.

[b] Binding energy of positronium to the anion vacancy, $E_B(Ps)-E(Fe^+)=-0.25h-E(Fe^+)$.

tem may be formally stable to a slightly lower value of Z^* than 0.45.

The behavior of the lifetime τ as a function of Z^* is shown in Fig. 2(b). As expected the lifetime τ monotonically decreases with Z^*. This curve could be used to find lifetimes of positrons for any defect modeled by an effective hydrogenic potential. We defer the comparison of our lifetimes with experiment until after the discussion of our results for the KS potentials.

The results for the KS potentials are presented in Table III. There is a marked increase in the lifetimes as compared to those for the Z^*/r potential.

Our binding energies $E_B(e^+)$ are lower than those found in Ref. 7 by a factor of 2. This is due to the sufficiently correlated nature of the wave functions employed in the present work. This fact highlights the importance of electron-positron correlations in such systems. It is also gratifying to see that for the KS potential the (Fe^+) complex is stable for all of the listed alkali halides. In Ref. 7 only LiH, LiCl, NaF, KF, and RbF were found to be stable with respect to free positronium for this potential.

In Ref. 7, the correlated calculation for those systems for which the cavity radius is larger than 6.18 a.u. (e.g., KBr, KI, RbCl, RbBr, and RbI) yielded the free positronium energy and wave function. No such effect was found in the present calculation for all sixteen crystals.

Our results for lifetimes (τ) and FWHM (Γ_N) of the angular correlation curves are compared in Table IV with the available experimental data. The lifetimes obtained

TABLE IV. Comparison of theory and experiment. The lifetimes obtained in the most correlated calculations of Ref. 7 are given in parentheses.

	τ (nsec)			Γ_N[a] (mrad)		
Crystal	Expt.[b]	KS	Z^*r^{-1}	Expt.[b]	KS	Z^*r^{-1}
NaCl	1.09±0.02 1.1 ±0.1	0.831 (0.61)	0.595	3.7±0.2	2.5	1.8
KCl	0.99±0.02 1.04	0.701 (0.64)	0.531	5.0±0.2 5.3	2.0	0.9
KBr	1.06±0.02	0.696 (NB)	NB[c]	5.0±0.2	2.1	NB
KI	1.46±0.04	1.02 (NB)	NB	4.2±0.2	1.7	NB

[a] Γ_N indicates full width at half maximum of $N(\theta)$, the two-photon angular correlation curve.

[b] Experimental data is taken from the compilation in Refs. 1 and 2.

[c] NB denotes not bound with respect to the free positronium state.

with the KS potential show much better agreement with experiment due to the better representation of the actual potential by the KS model. The improvement in the lifetimes obtained with the present method over that of Ref. 7 is evident (numbers in parentheses). This shows once again the importance of properly accounting for electron-positron correlation.

The values of Γ_N (FWHM) obtained for both potentials are lower than the experimental values by almost a factor of 2. The inclusion of correlation narrows the angular correlation curve, an observation consistent with Ref. 7. Since $\langle r_+ \rangle$, $\langle r_- \rangle$, and $\langle r_{+-} \rangle$ are comparable to the size of the F center, we believe that inclusion of nearest-neighbor interactions is essential for realistic calculation of angular correlations. It appears that the electron-positron pair momentum distribution is more sensitive to this interaction than are the lifetimes.

IV. CONCLUDING REMARKS

The present calculations have demonstrated that one must include a high degree of electron-positron correlation in the wave function of the (Fe$^+$) system in order to obtain reliable results for assessing the stability of the (Fe$^+$) system with respect to dissociation into either (i) a positronium atom (Ps) and a defect (-0.25 hartree) or (ii) into a free positron and an electron bound to the defect (i.e., a neutral F center). For the model parameters determined[13] for the description of the optical electron of the F center, our calculations show that for *all* sixteen crystals considered herein, the (Fe$^+$) complex is stable with respect to both channels (i) and (ii) for the Krumhansl-Schwarz potential. For the hydrogenic model potential (Z^*/r) we find that the (Fe$^+$) complex is not bound with respect to (i) for KBr, KI, RbCl, RbBr, and RbI and is bound for the remaining eleven crystals.

These results may be contrasted with earlier correlated calculations which showed that for the Z^*/r potential[7,8] only LiF is stable with respect to channel (i) while for the KS potential[7] only LiH, LiF, LiCl, NaF, NaCl, and KF exhibited such stability

The more realistic KS model yields the more satisfactory results with regard to stability and with regard to the positron lifetimes for the only four crystals (NaCl, KCl, KBr, and KI) for which experimental data is known to us. However, the two-photon angular correlation function, $N(\theta)$ calculated for both models is much narrower than the experimental data, but here again the results for the KS model are significantly closer to experiment. Clearly the calculated results could be improved in comparison with experiment by choosing the model parameters on the basis of the positron data. Such a choice would destroy the connection with the description of the optical data. The development of even more realistic models [including the effects of pickoff, screening of the electron-positron interaction by band electrons, the lattice structure, and electron- (positron) phonon interactions] could lead to improved results. Of course, it would be useful to have experimental data available for the other systems considered in the present study.

An interesting result of the present work is that the electron-positron system is bound in the Coulomb potential for $0.45 < Z^* < 1.0$. This is an additional demonstration[10] of the nonexistence of a bound state between a hydrogen atom and a positron.

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APPENDIX

In this appendix we report the results of a study of the number of terms, L, required in the expansion of the wave function

$$\psi(r_+,r_-,r_{+-})=(4\pi)^{-1}\sum_{k=1}^{L} C_k \exp(-\alpha_k r_+ -\beta_k r_- -\gamma_k r_{+-}) \quad (\mathrm{A1})$$

to obtain convergence of the Fe$^+$ energy and the lifetime τ. In Table V, data is presented for the Z^*/r potential for $Z^*=0.65$. In this case, E(Fe$^+$) and τ have converged, i.e., 60 terms do not change the results to the reported number of significant figures. It is clear that single-term wave functions are unreliable in this case. For the KS potential, $L=45$ was found to be sufficient.

TABLE V. Results calculated for the Z^*/r potential with $Z^*=0.65$.

L	$-E$(Fe$^+$)	$E_B(e^+)$	τ (nsec)
1	0.2632	0.0520	1.900
10	0.2814	0.0702	0.972
20	0.2818	0.0706	0.957
40	0.2821	0.0709	0.936
50	0.2822	0.0710	0.935

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[24] A detailed discussion for atomic systems can be found in Ref. 17. We only note here that the exact solution $\psi(\mathbf{r}_+,\mathbf{r}_-)$ of the Schrödinger equation (1) for $V(r)=Z^*/r$ would be expected to satisfy the cusp conditions: $(\partial/\partial r_+)\ln\bar{\psi}|_{r_+=0}=Z^*$, $(\partial/\partial r_-)\ln\bar{\psi}|_{r_-=0}=-Z^*$, and $(\partial/\partial r_{+-})\ln\bar{\psi}|_{r_{+-}=0}=\frac{1}{2}$, where $\bar{\psi}$ denotes the spherical average of ψ about the singularity in question. In the atomic helium problem (Ref. 17), the cusp conditions were satisfied quite well (the electron-nuclear cusp ratio was $-2.000\,183$ instead of the exact value of -2, while the electron-electron ratio was $0.495\,80$ instead of the exact value of 0.5 in the case of the $L=66$ wave function).

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