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Positrons in metals: A real-space approach

Pierre F. Maldague
IBM Thomas J. Watson Research Center,
Yorktown Heights, New York 10598
and Physics Department, Brandeis University,
Waltham, Massachusetts 02154 *

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A new, simple method for treating the short-range correlations in a one-positron, many-electron system is proposed. The method puts the emphasis on a real-space representation of wave functions. Our ultimate goal is to apply this method to situations in which the electron density is nonuniform, such as a metallic surface. In the present paper, we concentrate on the case of a uniform electron gas, which provides a useful test of the method. Our results for the total annihilation rate are well behaved at all densities, tend to the positronium limit at low density, and agree with experiment for simple metals. We show that the total rate is insensitive to electron-electron correlations, although such correlations could affect the momentum dependence of the partial annihilation rate measured in angular-correlation experiments. The extension of the method to nonuniform systems is briefly sketched.

I. INTRODUCTION

Few problems exhibit the importance of correlation effects as clearly as positron annihilation in metals. A basic quantity of interest in this problem is the annihilation rate R, given by[1,2]

$$R = \lambda \int d^3\vec{r}\, \langle \hat{\rho}_e(\vec{r}) \hat{\rho}_p(\vec{r}) \rangle \quad , \tag{1.1}$$

where $\hat{\rho}_e(\hat{\rho}_p)$ is the electron (positron) density operator and $\lambda = \pi e^4/m^2c^3$ is a constant. At zero temperature, the average in this expression should be taken in the ground state of the N-electron, one-positron system. If one neglects correlation effects entirely, this equation reduces to

$$R = \lambda \int d^3\vec{r}\, \rho_e(\vec{r}) |\Psi_p(\vec{r})|^2 \quad , \tag{1.2}$$

where $\rho_e(\vec{r}) = \langle \hat{\rho}_e(\vec{r}) \rangle$ and Ψ_p is the positron wave function. It is well known that rates computed from Eq. (1.2) are substantially smaller than found experimentally.[1,2] The reason is that the attractive Coulomb force between the positron and the electrons enhances the electron density near the positron, so that the correlated average in Eq. (1.1) can be much larger than the independent-particle prediction (1.2). Much work has been devoted in the last fifteen years to the theory of this "many-body enhancement" of the annihilation rate in a uniform electron gas, and the behavior of the total annihilation rate for metallic densities is now fairly well understood.

Many problems of experimental interest, however, are still awaiting a complete theoretical solution. Such problems include the effects of band structure on the annihilation rate, the momentum dependence of the partial rate R_p which is measured in angular-correlation experiments, the possible trapping of positrons near metal surfaces and more generally the behavior of positrons in nonhomogeneous electron systems. Our initial motivation for undertaking the present work was to develop a simple theory of electron-positron correlations that could be successfully applied to these problems, and to use the uniform electron gas as a test for determining the validity of the theory. Using our method in the simple case of a uniform electron gas turned out to be more fruitful than anticipated, however. Not only does our theory reproduce previous results for the annihilation rate at metallic densities, but it also provides a smooth transition between the high- and low-density limits, provides a new interpretation of the partial annihilation rate and sheds light on the respective roles of electron-positron and electron-electron correlations. We have therefore chosen to devote this paper to the case of a uniform electron gas; possible improvements and generalizations will be briefly sketched in Secs. IV and V.

In the absence of correlations, the electron density in the independent-particle formula (1.2) contains contributions from all occupied electron states. To lowest order, these are simply plane waves with wave vector $\vec{k}$ inside the Fermi sphere. If one assumes that the positron has thermalized before annihilating, its wave function is a plane wave with $\vec{k} = 0$. The effect of the electron-positron interaction is to cause transitions in which an electron is scattered from $\vec{k}$ to $\vec{k}+\vec{q}$, while the positron is scattered from $\vec{0}$ to $-\vec{q}$.

Because of the Pauli principle, the wave vector of the intermediate electron states must lie outside the Fermi sphere. In this way, Kahana[3] obtained a t-matrix equation for the electron-positron scattering amplitude, which can be used in Eq. (1.1) to calculate the annihilation rate R. The results are in good agreement with experiment for the nearly-free-electron (NFE) metals Al, Li, and Na, i.e., those NFE metals for which the electron-gas parameter r_s is less than about four. The same method can be used to calculate the partial annihilation rate R_p which is measured in angular-correlation experiments.[3,4] The results again agree with experiments on Na and Al,[5,6] although in the case of Al band-structure effects must be taken into account.

The t-matrix approximation is based on a high-density expansion in powers of r_s, and as with all electron-gas problems it is difficult to justify this expansion for values of r_s in the metallic range ($r_s \sim 2-6$). Many-body corrections to the t-matrix approximation have been investigated by Carbotte and Kahana,[7] who find a surprising amount of cancellation between some higher-order contributions. Sjölander and Stott[8] went beyond perturbation theory, and generalized to the electron-positron case the strong-coupling approach developed by Singwi *et al.*[9] to treat the electron gas. Their results for the total annihilation rate are very similar to those of Ref. 7.

The approaches outlined above suffer from a common problem. When the calculation is extended to values of r_s larger than four, one finds that the predicted rate R diverges. In particular, the rate predicted for $\mathrm{Cs}(r_s \simeq 6)$ is much higher than experiment. The breakdown of the theory was pointed out by Crowell *et al.*[10] for the t-matrix approximation, and by Sjölander and Stott for their own theory. A similar divergence shows up in apparently unrelated approaches such as that of Arponen and Pajanne,[11] which is based on a collective description of the electon gas. The divergence of the annihilation rate in these approximate theories is due to an excessive buildup of charge in the vicinity of the positron. The common origin of the divergence must lie in the neglect of the repulsion energy which would in reality prevent the excessive accumulation of electrons near the positron.

The first theory that succeeded in eliminating the divergence is that of Bhattacharyya and Singwi,[12] who added nonlinear corrections to the theory of Sjölander and Stott. The role played by the nonlinear correction is precisely to include correlation effects between the electrons which contribute to the density enhancement near the positron. Further insight into the nature of the divergence was provided by Lowy and Jackson,[13] who showed that the divergence arises from an incorrect description of bound states.

Although free of divergences, the theory of Bhattacharyya and Singwi does not reproduce the correct low-density limit for the annihilation rate. When $r_s \to \infty$, the many-body effects should become negligible. One expects that the positron will capture an electron to form positronium (Ps), which will move almost freely in the surrounding low-density gas. The annihilation rate should therefore tend toward the value $R = 2.0\ \mathrm{nsec}^{-1}$, the spin-averaged annihilation rate of free Ps. The theory of Bhattacharyya and Singwi, however, predicts a rate which tends continuously toward zero.[13] Leung *et al.*[14] proposed an alternative theory, which exploits the analogy between the problem of positron annihilation and the screening of a heavy impurity in an electron gas. Although their theory works well in the low-density limit and predicts a rate that tends toward the spin-averaged Ps value, the validity of the theory is difficult to assess in the high-density limit in view of the approximations used for the positron kinetic energy.

In Ref. 14, Leung *et al.* point out that the true low-density limit of the annihilation rate may be that of the Ps^- ion, as suggested by Ferrell[15] some time ago. Ferrell estimated the annihilation rate of Ps^- to be $\sim 3.2\ \mathrm{nsec}^{-1}$, 50% larger than the spin-averaged Ps rate. This estimate however, was based on a one-parameter variational wave function which yields a binding energy of about 0.20 eV. The correct value of the binding energy, obtained from a better variational wave function, is 0.33 eV.[16] A more accurate wave function for Ps^- was calculated by Ferrante[17] who finds that the electron density at the positron and the annihilation rate of Ps^- are nearly equal to the corresponding quantities in neutral Ps. Moreover, the small binding energy of Ps^- is reflected in its larger size. Since the binding of an extra electron presumably occurs when the density of the electron gas is comparable to that of the outer electron orbital in Ps^-, we will assume that the "correct" low-density limit of the annihilation rate for reasonable values of r_s is the spin-averaged Ps value.

This paper is organized as follows. In Sec. II, we will outline our approach to electron-positron correlations. The approach is based on a variational principle, in which the energy of a trial wave function is minimized. The wave functions that we consider can be freely adjusted in the vicinity of the positron, where the influence of the positron potential is strongest. Far away from the positron, our wave functions reduce to the noninteracting ground state. Minimizing the energy leads to a set of coupled Schrödinger equations which are derived in Sec. III. There, we also present expressions for the induced-electron density and the partial annihilation rate. We conclude that Sec. III with a discussion of the relaxation energy, which involves collective excitations neglected in our simple variational treatment. An approximate expression for these collective contributions will be derived. The results of our calculations are discussed in Sec. IV.

II. OUTLINE OF THE PRESENT APPROACH

We now turn to the description of our approach to electron-positron correlations in a uniform electron gas. The main reason for studying the uniform case is that we want to get an estimate of the accuracy of our approximation by comparing our results to those of previous approaches, most of which deal with a uniform electron gas. The generalization to the nonuniform case will be outlined in Sec. IV.

In the following, we will use the "relative coordinate" representation introduced in Ref. 14. Let $\Psi(\vec{r}_0,\vec{r}_1,\cdots,\vec{r}_N)$ be the wave function of the one-positron, N-electron system. $\vec{r}_0$ is the coordinate of the positron, and $\vec{r}_1,\cdots,\vec{r}_N$ are the electron coordinates. We define a new wave function ϕ by the equation

$$\Psi(\vec{r}_0,\vec{r}_1,\cdots,\vec{r}_N)=\phi(\vec{r}_{\rm cm},\vec{r}_1-\vec{r}_0,\cdots,\vec{r}_N-\vec{r}_0) \quad , \tag{2.1}$$

where $\vec{r}_{\rm cm}$ is the center-of-mass coordinate.
The advantage of this transformation is that the positron coordinate drops out of the transformed Hamiltonian, which becomes ($\hbar=1$)

$$\hat{H}=\frac{\vec{P}_0^2}{[2(N+1)m]}+\frac{\sum_{i=1}^{N}\vec{p}_i^{\,2}}{2m}+\frac{\left(\sum_i \vec{p}_i\right)^2}{2m}$$
$$+\sum_{i>j}\frac{e^2}{r_{ij}}-\sum_i\frac{e^2}{r_i} \quad . \tag{2.2}$$

The first term is the center-of-mass kinetic energy; $\vec{P}_0$ is the (conserved) total momentum of the system and will be taken equal to zero in the following. The next two terms are the electron and positron kinetic energies, while the last two terms represent the electron-electron and electron-positron interaction, respectively. Were it not for the positron kinetic energy, the Hamiltonian (2.2) would describe an electron gas with a positively charged impurity at the origin.[14] However, the finiteness of the positron mass leads to recoil effects which must be taken into account.

Since the positron coordinate has dropped out of the problem, we will not mention it explicitly. The problem is now to determine the wave function of an inhomogeneous N-electron system with Hamiltonian (2.2). Fortunately, we do not need a complete solution of the N-electron problem. We are interested in quantities such as the annihilation rate, which only involve the short-distance behavior of the wave function. Hence, it is sufficient to find a wave function which is accurate near the origin.

Our philosophy will be to start from the Hartree-Fock (HF) description of the electron gas, and to modify the HF ground state so as to include correlation effects near the origin. The HF ground state, denoted by $|0\rangle$, is a Slater determinant in which all plane-wave states with wave vector k less than k_F are occupied. The first step of our analysis is to realize that the HF ground state can be equally well described in terms of *localized* states. To be specific, consider the state $|\sigma\rangle$ defined by

$$|\sigma\rangle=[2/\rho]^{1/2}\hat{\Psi}_\sigma(\vec{0})|0\rangle \quad , \tag{2.3}$$

where $\hat{\Psi}_0(\vec{r})$ is a Fermi field operator which annihilates an electron with spin σ and ρ is the average electron density. It is easy to see that $|\sigma\rangle$ is also a Slater determinant, in which one electron orbital is missing. The (normalized) wave function of the missing electron is

$$a_0(r)=[2/\rho]^{1/2}\int_{k<k_F}\frac{d^3\vec{k}}{(2\pi)^3}e^{i\vec{k}\cdot\vec{r}} \quad . \tag{2.4}$$

Obviously, the HF ground state can be recovered from $|\sigma\rangle$ simply by recreating an electron in the state (2.4). Thus we have

$$|0\rangle=\int d^3\vec{r}\,a_0(r)\,\hat{\Psi}_\sigma^\dagger(r)\,|\sigma\rangle \quad , \tag{2.5}$$

where $\hat{\Psi}^+$ is the field conjugate to $\hat{\Psi}$. The wave function a_0 is, in a sense, the most localized orbital that can be made out of occupied plane-wave states. Hence, it is natural to expect that a_0 will also be the orbital which is most affected by the presence of the positron at the origin. This suggests that an approximate ground state for the Hamiltonian (2.2) could be obtained by allowing the orbital in the right-hand side of Eq. (2.5) to be different from a_0. We are then led to the following ansatz for the ground state:

$$|b\rangle=\beta|0\rangle+\frac{1}{\rho^{1/2}}\sum_\sigma\int d^3\vec{r}\,b(r)\,\hat{\Psi}_\sigma^\dagger(\vec{r})\,\hat{\Psi}_\sigma(\vec{0})\,|0\rangle \quad . \tag{2.6}$$

Normalization requires that $\int d^3\vec{r}\,b^2(r)=1-\beta^2$, and for β to be determined unambiguously we must require that b be orthogonal to the occupied states

$$\int b(r)e^{-i\vec{k}\cdot\vec{r}}d^3\vec{r}=0, \quad k<k_F \quad . \tag{2.7}$$

We could now determine $b(r)$ by minimizing the expectation value of the Hamiltonian (2.2) in the trial state (2.6), subject to the above constraint for the wave function $b(r)$. However, in order to calculate the momentum dependence of the partial rate R_p it is necessary to consider the following generalization of (2.6):

$$|a,b\rangle=\beta|0\rangle+\frac{1}{2^{1/2}}\sum_\sigma\int d^3\vec{r}\,b(r)\,\hat{\Psi}_\sigma^\dagger(\vec{r})\int d^3\vec{r}'\,a(r')\,\hat{\Psi}_\sigma(\vec{r}')\,|0\rangle \quad , \tag{2.8}$$

where $a(r)$ is a normalized wave function. In Sec. III, we will show that the Fourier transform of this wave function is closely related to the partial annihilation rate. For $a(r)$ to be determined unambiguously, we must impose the requirement

$$\int d^3\vec{r}\, a(r) e^{-i\vec{k}\cdot\vec{r}} = 0, \quad k > k_F \ . \tag{2.9}$$

In view of Eq. (2.5), our earlier ansatz (2.6) is a special case of Eq. (2.8), obtained by setting $a(r) = a_0(r)$.

Equation (2.8), together with the restrictions (2.7) and (2.9), describes the class of trial ground states which will be used to calculate the annihilation rate. Before getting into the actual calculation of the orbitals a and b (which will be discussed in Sec. III), we wish to make a few qualitative remarks on the implications of Eq. (2.8). Our ansatz has a simple interpretation when $\beta = 1 - \epsilon$ is close to unity. To order ϵ, the trial state (2.8) can be obtained from the uniform HF ground state by replacing the orbital $a(r)$ (which is doubly occupied) by the distorted orbital

$$(1-\epsilon/2)a(r) + b(r)/2^{1/2} \ .$$

Since only one electron of each spin is involved in this replacement, it is clear that the short-range correlations built into the trial state (2.8) have single-particle character.

The screening effect, which is responsible for the accumulation of electrons near a positively charged impurity, is usually described in terms of collective excitations (e.g., the random-phase approximation (RPA) or its nonlinear generalizations[2,12]). Yet, our ansatz (2.8) only contains a single electron-hole excitation. To try and reconcile these seemingly contradictory descriptions, it is useful to simplify the problem and let the positron mass become infinite. The problem then reduces to determining the response of the electron gas to the Coulomb field of a proton. Let us further simplify the problem by looking at the high-density limit, where perturbation theory can be used. To first order in the electron-proton potential, an occupied plane-wave state $|\vec{k}\rangle$ is transformed into a scattering state $|\vec{k})$ given by

$$|\vec{k}) = |\vec{k}\rangle + \frac{1}{V}\sum_{k'>k_F} \frac{\tilde{V}_{k-k'}}{k^2/2m - k'^2/2m}|\vec{k}') \ ; \tag{2.10}$$

where

$$\tilde{V}_{k-k'} = -4\pi e^2/|\vec{k}-\vec{k}'|^2$$

and V is the volume of the system. In RPA, $\tilde{V}$ also contains the Coulomb potential of the induced charge which must be determined self-consistently. Equation (2.10) can be rewritten as an equation for the perturbed many-body ground state $|1\rangle$ in terms of the unperturbed ground state $|0\rangle$

$$|1\rangle = |0\rangle + \frac{1}{V}\sum_{\sigma}\sum_{k<k_F}\sum_{k'>k_F} \frac{\tilde{V}_{k-k'}}{k^2/2m - k'^2/2m} \times \hat{a}^{\dagger}_{k',\sigma}\hat{a}_{k,\sigma}|0\rangle \ . \tag{2.11}$$

Here, $\hat{a}_{k,\sigma}$ annihilates an electron with wave vector $\vec{k}$ and spin σ. When one calculates the induced-electron density $\tilde{\rho}_q$ to the same order in $\tilde{V}$, the only terms in Eq. (2.11) which contribute to $\tilde{\rho}_q$ are those with $\vec{k}' - \vec{k} = \vec{q}$. For $q >> k_F$, the induced density could equally well be calculated from the approximate ground state

$$|1'\rangle = |0\rangle - \frac{1}{V}\sum_{\sigma}\sum_{k<k_F}\sum_{k'>k_F} \frac{\tilde{V}_{k'}}{k'^2/2m}\, \hat{a}^{\dagger}_{k',\sigma}\hat{a}_{k,\sigma}|0\rangle \ , \tag{2.12}$$

since k is less than k_F and can be neglected compared to k'. The sums over k and k' in Eqs. (2.10)–(2.12) are now decoupled, so that the state $|1'\rangle$ is of the separable form (2.8). Thus, we find that the ansatz (2.8) for the perturbed ground state can account for the large q components of the induced-charge density, hence for the short-range behavior of the wave function in real space.

If one is interested in the small q (long-range) components of the induced density, however, it is clear that Eq. (2.12) no longer provides a valid approximation. In particular, the total induced charge in the state $|1'\rangle$ vanishes [as it does in our ansatz (2.8)], since $\tilde{\rho}_q \to 0$ as $q \to 0$. To obtain the correct limit $\tilde{\rho}_q \to 1$, one must allow for collective excitations of the electron gas in which all the orbitals away from the proton are pulled in so as to screen the Coulomb potential at large distance. These collective excitations are represented by the terms with $\vec{k} \sim \vec{k}'$ in Eq. (2.11), which cannot be approximated by a separable electron-hole excitation as in (2.12).

To summarize the above discussion, we can say that a state containing a single excitation such as Eq. (2.12) provides a correct description of the short-range behavior of the exact ground state. However, one should add to Eq. (2.12) a "collective contribution" which is negligible close to the proton but is dominant away from it. A more quantitative discussion of these two contributions will be given in Sec. IV, where we will compare our results to those of Almbladh *et al.*[18] Whether or not the collective contribution can be neglected depends on the quantity one is interested in. It is clear from the definition (1.1) that the total annihilation rate is a local quantity for which a wave function of the form (2.8) should provide an adequate approximation. Other quantities, such as the relaxation energy, depend in an essential way on long-wavelength density excitations.[19] A simple approximation for the collective contribution to the relaxation energy will be discussed in Sec. III.

III. EFFECTIVE SCHRÖDINGER EQUATIONS

The goal of this section is to derive effective Schrödinger equations for the electron and hole orbitals that enter the trial ground state (2.8). These equations will be derived by requiring that the expectation value of the Hamiltonian in the trial ground state be a minimum. This expectation value can be expressed in terms of averages of products of field operators in the noninteracting ground state, whose evaluation is straightforward if somewhat lengthy. We may without restriction we will assume that a and b are real, and since our problem has spherical symmetry we will assume that both have s-like character. The relevant averages can be written as follows:

$$H_e = \frac{1}{2m} \int d^3\vec{r} \left[\left(\frac{d}{dr} b(r) \right)^2 - (1-\beta^2) \left(\frac{d}{dr} a(r) \right)^2 \right] , \tag{3.1}$$

$$H_p = \frac{1}{2m} \int d^3\vec{r} \left[\left(\frac{d}{dr} b(r) \right)^2 + (1-\beta^2) \left(\frac{d}{dr} a(r) \right)^2 \right] , \tag{3.2}$$

$$H_{ep} = -2(2)^{1/2}\beta \int d^3\vec{r}\, a(r) b(r) \frac{e^2}{r} - \int d^3\vec{r}\, b(r)^2 \frac{e^2}{r} + (1-\beta^2) \int d^3\vec{r}\, a(r)^2 \frac{e^2}{r} , \tag{3.3}$$

$$\begin{aligned} H_{ee} = &- \int d^3\vec{r} d^3\vec{r}'\, b(r) b(r') \frac{e^2}{|\vec{r}-\vec{r}'|} G(|\vec{r}-\vec{r}'|) + (1-\beta^2) \int d^3\vec{r} d^3\vec{r}'\, a(r) a(r') \frac{e^2}{|\vec{r}-\vec{r}'|} G(|\vec{r}-\vec{r}'|) \\ &- \int d^2\vec{r} d^3\vec{r}' \frac{e^2}{|\vec{r}-\vec{r}'|} b(r)^2 a(r)^2 + 2 \int d^3\vec{r} d^3\vec{r}'\, a(r) b(r) \frac{e^2}{|\vec{r}-\vec{r}'|} a(r') b(r') . \end{aligned} \tag{3.4}$$

In these equations, $H_e(H_p)$ is the expectation value of the electron (positron) kinetic energy, and $H_{ee}(H_{ep})$ is that of the electron-electron (electron-positron) Coulomb interaction. We have subtracted from H_e and H_{ee} the corresponding HF energies of the uniform N-electron system, so as to obtain expressions that are independent of the volume. We have assumed that the electron gas is immersed in a uniform background of positive charge in order to avoid electrostatic divergences in H_{ep} and H_{ee}. We have also used the fact that $a(r)$ is normalized to unity and $b(r)$ to the smaller quantity $1-\beta^2$, and we have taken into account the Pauli principle restrictions (2.7) and (2.9). The various terms in Eqs. (3.1)–(3.4) have the following interpretation:

(a) H_e is the change in kinetic energy due to the excitation of an electron from the orbital a to the orbital b.

(b) The positron kinetic energy can be written as $1/2m\,\langle(\vec{k}_e+\vec{k}_h)^2\rangle$, where $\vec{k}_e$ and $\vec{k}_h$ are the electron and hole wave vectors in the trial state (2.8). Since the cross-term $\langle\vec{k}_e\cdot\vec{k}_h\rangle$ vanishes for spherically symmetric orbitals, we are left with a contribution similar to Eq. (3.1) except that the hole term is now positive.

(c) The last two terms in H_{ep} represent the enhancement in Coulomb attraction resulting from the replacement of $a(r)$ by $b(r)$. The first term is what we call the "hybridization" contribution to the energy. This term arises because the occupied states are no longer plane-wave states. At high densities, β is near unity and b is of the order of the small quantity $(1-\beta^2)^{1/2}$ due to the normalization condition. The hybridization term is then linear in $(1-\beta^2)^{1/2}$ and provides the dominant contribution to the potential energy.

(d) The quantity $G(r-r')$ in the expression for H_{ee} is defined as

$$\langle \hat{\psi}_\sigma^+(\vec{r}) \hat{\psi}_\sigma(\vec{r}') \rangle = [\rho/2]^{1/2} a_0(|\vec{r}-\vec{r}'|) ,$$

see Eq. (2.3). The first term is the energy arising from exchange between the excited electron and N electrons inside the Fermi sea. The second term is minus the exchange energy of the orbital $a(r)$, which becomes empty upon excitation of an electron. The third term is the direct interaction between the excited electron and the positive background. In the uniform state, the background charge is exactly cancelled by that of the electrons. Since an orbital $a(r)$ has been removed, however, the excited electron sees a net charge $ea^2(r)$. Finally, the last term serves two purposes: first, it partially cancels the exchange energy of the excited electron, and accounts for the fact that the excited electron can undergo exchange processes with only N-1 electrons rather than N as the first term in H_{ee} indicates. Second, it accounts for the repulsion energy between two excited electrons of opposite spin.

Having calculated the energy of our trial state (2.8), we may now derive the effective Schrödinger equations satisfied by $a(r)$ and $b(r)$. Neglecting H_{ee} for the moment, the effective Schrödinger equations for a and b are obtained by minimizing

$$H_e+H_p+H_{ep}-E\left[\beta^2+\int d^3\vec{r}\,b^2(r)\right]$$

and

$$H_e+H_p+H_{ep}-E'\int d^3\vec{r}\,a^2(r)\ ,$$

respectively. E and E' are Lagrange multipliers which take care of the normalization conditions. In view of the Pauli principle restrictions (2.7) and (2.9), it is convenient to rewrite Eqs. (3.1)–(3.3) in terms of the Fourier transforms

$$\tilde{b}(k)=[\rho/2]^{1/2}\int d^3\vec{r}\,e^{-i\vec{k}\cdot\vec{r}}b(r) \tag{3.5}$$

and

$$\tilde{a}(k)=[\rho/2]^{1/2}\int d^3\vec{r}\,e^{-i\vec{k}\cdot\vec{r}}a(r)\ . \tag{3.6}$$

The effective Schrödinger equations are then obtained by taking functional derivatives with respect to $b(k)$ for $k>k_F$, $a(k)$ for $k<k_F$ and β. The equation for β is simply

$$E\beta=-2^{1/2}\int d^2\vec{r}\,a(r)b(r)(e^2/r)\ , \tag{3.7}$$

and the momentum-space equations for a and b are

$$\left[\frac{k^2}{2m}+\epsilon_{ep}\{a\}\right]\tilde{b}(k)-\int_{k'>k_F}\frac{d^3\vec{k}'}{(2\pi)^3}\tilde{b}(k')\frac{4\pi e^2}{|\vec{k}-\vec{k}'|^2}-2^{1/2}\beta\int_{k'<k_F}\frac{d^3\vec{k}'}{(2\pi)^3}\tilde{a}(k')\frac{4\pi e^2}{|\vec{k}-\vec{k}'|^2}=E\tilde{b}(k),\quad k>k_F\ , \tag{3.8}$$

$$(1-\beta^2)\int_{k'<k_F}\frac{d^3\vec{k}'}{(2\pi)^3}\tilde{a}(k')\frac{4\pi e^2}{|\vec{k}-\vec{k}'|^2}-2^{1/2}\beta\int_{k'>k_F}\frac{d^3\vec{k}'}{(2\pi)^3}\tilde{b}(k')\frac{4\pi e^2}{|\vec{k}-\vec{k}'|^2}=E'\tilde{a}(k),\quad k<k_F\ . \tag{3.9}$$

The positive quantity $\epsilon_{ep}\{a\}$ is defined by

$$\epsilon_{ep}\{a\}=\int d^3\vec{r}\,\frac{e^2}{r}a(r)^2\ . \tag{3.10}$$

We now discuss the contributions of H_{ee} to the effective Schrödinger equations. The first two terms in Eq. (3.4) yield contributions

$$[\epsilon_x\{a\}-\epsilon_x(k)]\tilde{b}(k)$$

and

$$(1-\beta^2)\epsilon_x(k)a(k)$$

to the left-hand sides of Eqs. (3.8) and (3.9), respectively. The exchange energy $\epsilon_x(k)$ is given by

$$\epsilon_x(k)=\frac{e^2}{\pi}\left[k_F+\tfrac{1}{2}(k_F^2/k-k)\ln\frac{k+k_F}{|k-k_F|}\right]\ , \tag{3.11}$$

and $\epsilon_x\{a\}$ is the average of $\epsilon_x(k)$ calculated in the hole state $\tilde{a}(k)$

$$\epsilon_x\{a\}=\frac{2}{\rho}\int\frac{d^3\vec{k}}{(2\pi)^3}\tilde{a}(k)^2\epsilon_x(k)\ . \tag{3.12}$$

The third term in H_{ee} gives rise to local potentials in the equations for a and b. These potentials are

$$V_{\text{loc,a}}(r)=\int d^3\vec{r}'\,b(r')^2\frac{e^2}{|\vec{r}-\vec{r}'|}\ , \tag{3.13}$$

$$V_{\text{loc,b}}(r)=\int d^3\vec{r}'\,a(r')^2\frac{e^2}{|\vec{r}-\vec{r}'|}\ . \tag{3.14}$$

Finally, the last term in H_{ee} gives rise to nonlocal potentials $V_{nl,a}$ and $V_{nl,b}$. The Fourier transforms of these potentials are given by

$$\tilde{V}_{nl,a}(\vec{k},\vec{k}')=\frac{16\pi e^2}{\rho}\int\frac{d^3\vec{q}}{(2\pi)^3}\frac{\tilde{b}(\vec{k}+\vec{q})\tilde{b}(\vec{k}'-\vec{q})}{q^2}\ , \tag{3.15}$$

$$\tilde{V}_{nl,b}(\vec{k},\vec{k}')=\frac{16\pi e^2}{\rho}\int\frac{d^3\vec{q}}{(2\pi)^3}\frac{\tilde{a}(\vec{k}+\vec{q})\tilde{a}(\vec{k}'-\vec{q})}{q^2}\ . \tag{3.16}$$

We may summarize our results in terms of the contributions $(V_{ee}\tilde{b})(k)$ and $(V_{ee}\tilde{a})(k)$ which must be added to the left-hand sides of Eqs. (3.8) and (3.9) when the electron-electron contribution to the energy is included. We find

$$(V_{ee}\tilde{a})(k)=(1-\beta^2)\epsilon_x(k)\tilde{a}(k)+\int_{k'<k_F}\frac{d^3\vec{k}'}{(2\pi)^3}[\tilde{V}_{nl,a}(\vec{k},\vec{k}')-\tilde{V}_{\text{loc},a}(\vec{k}-\vec{k}')]\tilde{a}(k')\ , \tag{3.17}$$

$$(V_{ee}\tilde{b})(k)=[\epsilon_x\{a\}-\epsilon_x(k)]\tilde{b}(k)+\int_{k'>k_F}\frac{d^3\vec{k}'}{(2\pi)^3}[\tilde{V}_{nl,b}(\vec{k},\vec{k}')-\tilde{V}_{\text{loc},b}(\vec{k}-\vec{k}')]\tilde{b}(\vec{k}')\ , \tag{3.18}$$

where $\tilde{V}_{\mathrm{loc},a}$ and $\tilde{V}_{\mathrm{loc},b}$ are the Fourier transforms of the local potentials (3.13) and (3.14).

The variational ground state (2.8) is completely determined by the effective Schrödinger equations (3.7)–(3.9), with the electron-electron contributions (3.17) and (3.18). Once these equations have been solved, it remains to express the experimentally observable quantities in terms of the electron and hole orbitals. In the remainder of this section, we derive expressions for the partial annihilation rate, the total rate, the induced density, and the relaxation energy.

The partial annihilation rate R_p is the contribution to the total annihilation rate from electron-positron pairs having total momentum p. In our relative coordinate representation, R_p is given by

$$R_p = \lambda \int d^3\vec{r}\, e^{-i\vec{p}\cdot\vec{r}} \sum_\sigma \langle \phi | \hat{\Psi}_\sigma^\dagger(\vec{0})\, e^{-i\vec{P}_{el}\cdot\vec{r}}\, \hat{\Psi}_\sigma(\vec{0}) | \phi \rangle \ , \tag{3.19}$$

where $\vec{P}_{el}$ is the total momentum operator for electrons and $|\phi\rangle$ is the exact ground state. The meaning of this equation is clear: after annihilation, the electronic wave function is $\hat{\Psi}_\sigma(\vec{0})|\phi\rangle$. The momentum of the annihilating pair must be the total momentum $\vec{P}_{\mathrm{tot}}$ of the $(N+1)$-particle system (which we assumed to be zero), minus the amount of momentum left in the system after annihilation, which is precisely the quantity expressed in the right-hand side of Eq. (3.19). With our ansatz (2.8) for the electronic wave function, the expression for R_p can be reduced to expectation values of products of field operators in the noninteracting ground state. A straightforward but lengthy calculation yields

$$\begin{aligned} R_p = {} & 2\lambda\beta^2\theta(k_F - p) + \frac{4\lambda\beta}{\rho^{1/2}} b(0)\tilde{a}(p) + \frac{2\lambda}{\rho} b(0)^2\tilde{a}(p)^2 \\ & + \lambda \int d^3\vec{r}\, e^{-i\vec{p}\cdot\vec{r}} [2G(r)A_2(r) - a(r)^2] B_2(r) \ . \end{aligned} \tag{3.20}$$

As before, the Green function $G(r)$ is defined as $\langle \hat{\Psi}_\sigma^\dagger(\vec{r})\hat{\Psi}_\sigma(\vec{0})\rangle$. The functions A_2 and B_2 are defined by

$$A_2(r) = \frac{2}{\rho} \int \frac{d^3\vec{k}}{(2\pi)^3} e^{i\vec{k}\cdot\vec{r}} \tilde{a}(k)^2 \ , \tag{3.21}$$

$$B_2(r) = \frac{2}{\rho} \int \frac{d^3\vec{k}}{(2\pi)^3} e^{i\vec{k}\cdot\vec{r}} \tilde{b}(k)^2 \ . \tag{3.22}$$

Note that normalization requires

$$A_2(0) = 1, \quad B_2(0) = 1 - \beta^2 \ .$$

The total annihilation rate is obtained by integrating R_p, which yields

$$\begin{aligned} R = \lambda[& \rho + b(0)^2 + 2(2)^{1/2}\beta a(0) b(0) \\ & - (1-\beta^2) a(0)^2] \ . \end{aligned} \tag{3.23}$$

Next, we calculate the induced-charge density $\delta\rho(r)$ defined by

$$\delta\rho(r) = \sum_\sigma \langle \phi | \hat{\Psi}_\sigma^\dagger(\vec{r}) \hat{\Psi}_\sigma(\vec{r}) | \phi \rangle \ . \tag{3.24}$$

The calculation is again straightforward, and one finds after some algebra

$$\begin{aligned} \delta\rho(r) = {} & 2(2)^{1/2}\beta a(r) b(r) + b(r)^2 \\ & - (1-\beta^2) a(r)^2 \ . \end{aligned} \tag{3.25}$$

We conclude this section with a discussion of the relaxation energy E_c of the positron. E_c can be defined as the amount by which the ground-state energy changes when the electron-positron interaction is turned on. Since we have subtracted the energy of the uniform HF ground state from the energy averages (3.1)–(3.4), the relaxation energy E_1 predicted by our variational principle is

$$E_1 = H_e + H_p + H_{ep} + H_{ee} \ . \tag{3.26}$$

A little reflection shows that the right-hand side of this equation is in fact equal to E, the eigenvalue of the Schrödinger equation (3.8). Unfortunately, relaxation energies calculated in this way turn out to be too high. Formally, we may use the Feynman formula and write E_c in the form

$$E_c = -\int_0^e de' \left\langle \int d^3\vec{r}\, \frac{e}{r} (\hat{\rho}_e(\vec{r}) - \rho) \right\rangle_{e'} \ , \tag{3.27}$$

where the subscript indicates that the average should be calculated assuming the positron has charge e'. It is clear from this expression that E_c is a sensitive function of the induced density far away from the positron, unlike the total rate (1.1) which only involves the electron density at the origin. As discussed in Sec. II, our ansatz underestimates the induced density far from the positron, hence also the absolute magnitude of the relaxation energy.

To see how this defect of our theory could be corrected, it is instructive to look at the high-density limit. Since the RPA should become exact in this limit, Eq. (3.27) becomes

$$E_c = \frac{1}{2} \int \frac{d^3\vec{q}}{(2\pi)^3} \frac{4\pi e^2}{q^2} \left(\frac{1}{\epsilon(q)} - 1 \right) . \tag{3.28}$$

Here,

$$\epsilon(q) = 1 + (4\pi e^2/q^2)\pi(q)$$

is the RPA dielectric function, but $\pi(q)$ differs slightly from the Lindhard function due to the recoil energy of the positron

$$\pi(q) = 8m \int \frac{d^3\vec{r}}{(2\pi)^3} \frac{n_{\vec{k}}(1 - n_{\vec{k}+\vec{q}})}{(\vec{k}+\vec{q})^2 + q^2 - k^2} . \tag{3.29}$$

Equation (3.28) is similar to the expression used by Bergersen and Carbotte.[19] Taking $r_s \rightarrow 0$ in this equation, we find $E_c \cong -\frac{1}{2}(e^2 q_{\text{TF}})$, where $q_{\text{TF}} \sim 1/(r_s)^{1/2}$ is the Thomas-Fermi wave vector. The main contribution to E_c comes from the region $q \sim q_{TF} << k_F$ in Eq. (3.28), which implies that most of the correlation energy arises from long wavelength, collective excitations in the electron gas. The relaxation energy predicted by Eq. (3.26), however, only includes short-range excitations and tends to a constant as $r_s \rightarrow 0$. To see this, observe that the kinetic energy term in Eq. (3.8) dominates the other terms containing $\tilde{b}(k)$, and assuming $\tilde{a}(k) = 1$ one finds

$$\tilde{b}(k) \cong (2)^{1/2} \beta m \epsilon_x(k)/k^2, \quad k > k_F . \tag{3.30}$$

The normalization condition then gives

$$1 - \beta^2 = \left(\frac{2}{\rho} \right) \int \frac{d^3\vec{k}}{(2\pi)^3} \tilde{b}(k)^2 = 0(r_s^2) ,$$

so that $\beta \approx 1$. Equation (3.7) for β then yields

$$E \cong -\left(\frac{4m}{\rho} \right) \int_{k>k_F} \frac{d^3k}{(2\pi)^3} \epsilon_x(k)^2/k^2 . \tag{3.31}$$

Rewriting the integral in terms of the variable $x = k/k_F$, one finds that $E = 0(1)$ as $r_s \rightarrow 0$. If one now uses Eq. (3.27) to calculate $\tilde{a}(k)$ from Eq. (3.9), one finds that $\tilde{a}(k)$ depends weakly on k, with a maximum at k_F. We should in principle go back to the equation for $\tilde{b}$ and iterate until self-consistency is achieved, but it is clear that only quantitative changes would result.

In the low-density limit, it is easy to see that $\beta \rightarrow 0$ and Eq. (3.8) reduces to the Schrödinger equation of *Ps* in terms of center-of-mass coordinates. As discussed in the Introduction, the only effect of collective excitations in this limit would be to transform *Ps* into Ps^-. Since the binding energy of Ps^- is small, the relaxation energy essentially reduces to the single excitation contribution $E_1 = -\frac{1}{4}$ a.u.

For intermediate values of r_s, the contribution E_{coll} of collective modes to the relaxation energy can be estimated from the following physical argument. Let us write the total induced density as

$$\rho_{\text{ind}}(r) = \rho_1(r) + \rho_c(r) , \tag{3.32}$$

where ρ_1 is calculated with our ansatz (2.8) and ρ_c is a collective contribution. Although ρ_1 alone leads to a vanishing induced charge [i.e., $-e \int d^3\vec{r} \rho_1(r) = 0$], it tends to cancel the charge density $e\delta(\vec{r})$ of the positron at short distances. We may look at ρ_c as the response of the electron gas to the charge density $e\delta(\vec{r}) - e\rho_1(r)$. Since the potential induced by this charge density is much weaker then the bare potential $-e^2/r$, we may calculate ρ_c in linear response theory. The dielectric constant that relates ρ_c to the potential should in principle take into account the presence of a particle-hole excitation in the wave function (2.8). However, in the high-density limit where the collective contribution is important, we have seen that $\beta \simeq 1$ in Eq. (2.8), so that the variational ground state essentially reduces to the uniform HF ground state. We may then use the RPA dielectric function and write ρ_c in the form

$$\tilde{\rho}_c(q) = \left(1 - \frac{1}{\epsilon(q)} \right) (1 - \tilde{\rho}_1(q)) , \tag{3.33}$$

where $\tilde{\rho}_c$ and $\tilde{\rho}_1$ are the Fourier transforms of ρ_c and ρ_1. The corresponding approximation for the collective contribution to the relaxation energy is then

$$E_{\text{coll}} = \frac{1}{2} \int \frac{d^3\vec{q}}{(2\pi)^3} \frac{4\pi e^2}{q^2} \times [1 - \tilde{\rho}_1(q)]^2 \left(\frac{1}{\epsilon(q)} - 1 \right) . \tag{3.34}$$

In the high-density limit, $\tilde{\rho}_1(q)$ is much less than unity for $q \leq k_F$, so that one recovers Eq. (3.28). At low densities, $\tilde{\rho}_1(q)$ rapidly increases to a value close to unity, while the last factor in the integrand of Eq. (3.34) tends to zero as q/k_F becomes large. Since $k_F \sim 1/r_s$ tends to zero, we see that E_{coll} vanishes in that limit.

For intermediate values of r_s, we do not expect the RPA formula (3.34) to be accurate. First, the distortion in the wave function (2.8) near the origin becomes important, so that the dielectric function used to calculate ρ_c should be that of the distorted state rather than the noninteracting state. Second, the presence of an extra "screening charge" will add new contributions to the effective Schrödinger equations for the electron and hole orbitals. We will show in Sec. IV that such corrections could significantly modify the momentum dependence of the partial annihilation rate. Although some of the simplicity of our ansatz would be lost, including these corrections would be worth the effort since much work has been devoted to the observation of partial rates. We hope to return to these problems in a future publication.

IV. RESULTS AND DISCUSSION

An often used criterion for deciding the validity of a theory of positron annihilation is whether the theory can reproduce the experimental annihilation rates in simple metals. Less attention has been paid to the partial annihilation rate R_p, which will be discussed in this section along with other quantities of interest. Following tradition, we have plotted in Fig. 1 the annihilation rate R as a function of r_s for metallic densities. The continuous curve was calculated by solving numerically the effective Schrödinger equations derived in Sec. III. For the electron orbital b we use a mesh of 200 points separated by 0.2 a.u. in real space. The values of the Fourier-transformed hole orbital were specified at 20 equally spaced points between 0 and k_F. The Coulomb contributions (3.17) and (3.18) were included in the calculation, and the annihilation rate was calculated from Eq. (3.23). The experimental results of Weisberg and Berko[3] for Al, Mg, Li, Na, and K are indicated by open circles. The full circle is the rate for Al reported by MacKenzie *et al.*,[20] which is in better agreement with recent experiments.[21] Although the agreement between theory and experiment is quite good, the experimental data contain contributions from band structure and core annihilation effects which are not included in the theory. Bhattacharyya and Singwi estimate[12] that core annihilation contributes about 15% to the total annihilation rate of the heavier elements. Correcting the data of Weisberg and Berko in this manner, we obtain the squares in Fig. 1 which are in better agreement with the theory of Ref. 12 than with ours. Band structure and core annihilation effects are not quantitatively understood, however, and one will have to wait for satisfactory treatments of such effects before using experimental data as an accurate test of electron gas theories. Given that uncertainty, our theory does as well as previous ones (Refs. 3, 8, 12–14) for $r_s < 4$. At low densities, our theory yields a rate which tends smoothly toward the *Ps* value, in contrast to the *t*-matrix approximation[3,10] which leads to a divergence and to the theory of Bhattacharyya and Singwi which yields a vanishing rate.[13]

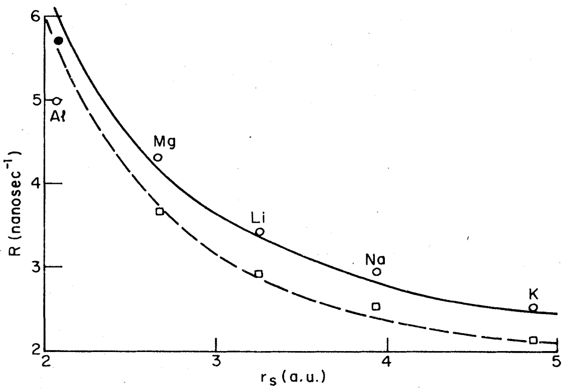

FIG. 1. Annihilation rate in nsec^{-1} as a function of r_s in atomic units. Continuous curve: present theory; dashed curve: theory of Ref. 12. The circles are the experimental values of Weisberg and Berko (Ref. 2); the squares represent the valence electron contribution as estimated by Bhattacharyya and Singwi (Ref. 12).

The total annihilation rate is proportional to the total electron density at the position of the positron. Much more information can be obtained from the induced density $\delta\rho$ at finite distance from the positron. According to Eq. (3.25), $\delta\rho$ is the sum of (a) a hybridization contribution $2(2)^{1/2}\beta a(r)b(r)$, (b) an electron contribution $b(r)^2$, and (c) a hole contribution $-(1-\beta^2)a(r)^2$. These three contributions are plotted in Fig. 2 for $r_s = 3.1$ (top) and 4.1 (bottom). The hybridization contribution dominates as $r_s \rightarrow 0$, since in that limit $\beta \rightarrow 1$ and $b(r)$ is normalized to the small quantity $1-\beta^2$. For $r_s \sim 3$, the electron and hybridization contributions are about equal. The electron orbital remains quite localized, and resembles the wave function of *Ps* (for which $\rho(0) = \frac{1}{8}\pi = 0.040$). The oscillations in $b(r)^2$ at $r \gtrsim r_s$ come from the requirement that $b(r)$ be orthogonal to the occupied electron states. The hole orbital, on the other hand, becomes less and less lo-

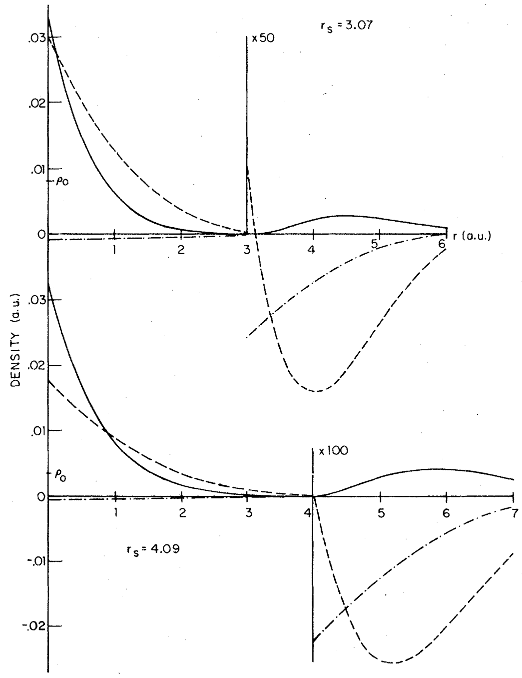

FIG. 2. Contributions to the induced density from hybridization (dashed curves), electron (continuous curves) and hole (dash-dotted curves) terms. For comparison, the electronic density at the origin in positronium is 0.040 a.u.

calized as r_s increases. The reason for this behavior is not difficult to find: it is the result of a competition between the "hybridization energy" (the first term in the expression (3.3) for the average electron-positron interaction) and the Coulomb repulsion between the positron and the hole (the last term in the same expression). At high densities, β is large and the hybridization energy dominates, leading to a localized hole. At low densities, $\beta \rightarrow 0$ and the repulsion term dominates, pushing the hole orbital away from the positron.

The above discussion sheds light on the rather remarkable agreement between the rate predicted by many different theories in the range $2 < r_s < 4$. Since electron-gas correlations are important in that range, one would have expected that differences in the treatment of correlation effects would lead to a significant spread in the predicted values of the annihilation rate. The induced densities depicted in Fig. 2 suggest that correlation effects should in fact play a marginal role, since the electron orbital is localized within a region smaller than a sphere of radius r_s. To confirm this interpretation, we have repeated our calculations without the Coulomb contributions (3.17) and (3.18) to the Schrödinger equations. We find that the resulting change in R is small (about 1% for $r_s = 2$ and 5% for $r_s = 4$), although individual terms in Eqs. (3.17) and (3.18) have a large effect on R. The smallness of the overall change must be due to a large amount of cancellation among the electron-electron contributions. There is an interesting parallel between this result and that obtained by Carbotte and Kahana,[7] who also found a near cancellation of higher-order contributions to the t-matrix approximation. In view of the considerable differences between our approach and that of Ref. 7, it is likely that the cancellation is not accidental, but reflects the fact that the annihilation rate is dominated by short-range excitations.

As a further test of our approximation, we have calculated the induced-charge density in the limit of an infinite positron mass. The Schrödinger equations (3.8) and (3.9) and the Coulomb contributions (3.17) and (3.18) are easily adapted to this case, and have been solved numerically for $r_s = 2.2$. If we add to Eq. (3.25) the contribution (3.33) of collective excitations, we obtain an induced-charge density which agrees with the results of Almbladh *et al.*[18] to within 5% at distances from the origin less than 2 a.u. Repeating the calculation for $r_s = 4$, we again obtain good agreement at short distances, although the Friedel oscillations in our calculated induced density are smaller than found in Ref. 18. This is probably due to the inadequacy of the RPA formula (3.33) at low density. As discussed in Sec. III, the collective contribution to the charge density is small in that regime, so that the error introduced by the RPA at short distances is not significant.

Returning to positron annihilation, let us discuss the partial rate R_p given by Eq. (3.20). It is convenient to use the "enhancement factor" $\epsilon(p)$,[3] defined as R_p/R_p^0 where $R_p^0 = 2\lambda$ is the Sommerfield partial rate. Our results are qualitatively similar to those of Refs. 3 and 7, where it is found that; (i) $\epsilon(p)$ increases monotonically from 0 to k_F, and (ii) $\epsilon(p)$ is negligible for $p > k_F$. However, the maximum in our $\epsilon(p)$ curves is much more pronounced than found in Ref. 3. According to a recent investigation of the Kahana equation,[22] the extrapolation procedure used in Ref. 3 is incorrect and the t-matrix formalism predicts a singularity in $\epsilon(p)$ for $p = k_F$. This would be consistent with our numerical results. Interestingly, the enhancement factor calculated by Kahana[3] is in very good agreement with the angular correlation experiments of Donaghy and Stewart[23] on Na. The sharper maximum found here and in Ref. 22 thus spoils the agreement between theory and experiment. The quantity that is measured experimentally is (apart from a normalization constant)

$$N_p = \int_p^{k_F} dk\, k\,\epsilon(k) \Big/ \int_0^{k_F} dk\, k\,\epsilon(k) \quad . \tag{4.1}$$

For a free-electron gas, $\epsilon(k) = 1$ and the above equation becomes $N_p^{(0)} = 1 - (p/k_F)^2$. The continuous curve in Fig. 3 is the deviation $N_p - N_p^{(0)}$ from free-electron behavior predicted by our theory for $r_s = 4$. The dashed curve is the prediction of the t-matrix theory as calculated by Kahana,[3] which agrees very well with the experimental data.[23] The dash-dotted curve shows the result of the improved t-matrix calculation of Ref. 22. As in our discussion of the total annihilation rate, we must emphasize that the experimental determination of N_p involves the substraction of a (p dependent) core contribution and that there are uncertainties associated with this procedure. However, the discrepancy between our results and

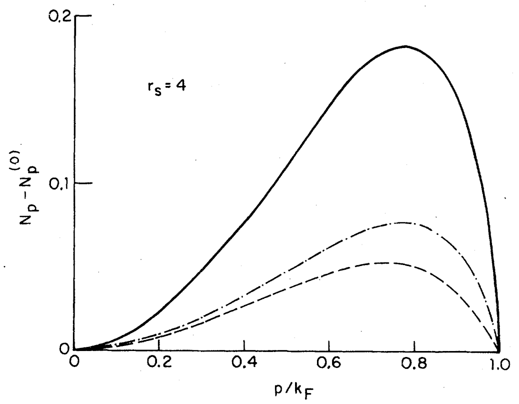

FIG. 3. Deviation from free-electron behavior of the normalized counting rate, Eq. (4.1), for $r_s = 4$. Continuous curve: present theory; dashed curve: theory of Ref. 3; dash-dotted curve: theory of Ref. 22.

those obtained in the t-matrix formalism is significant and deserves some discussion.

The excessive bulge predicted by our theory is a consequence of the rapid increase of the enhancement factor $\epsilon(p)$ near k_F. To understand the physical origin of this increase, let us go back to the expression (3.20) for the partial rate. The main contributions to R_p are the terms involving $b(0)\tilde{a}(p)$, whose momentum dependence is governed by that of the hole orbital $\tilde{a}(p)$. At low densities, one finds that this orbital tends to be concentrated in a thin shell $p \simeq k_F$ in momentum space. This corresponds to an extended orbital in real space, and we saw in our discussion of the induced density that the origin of this behavior is the repulsion energy between the positron and the hole. One possible reason for the excessive delocalization of the hole orbital may be that some of the correlation effects neglected in our theory tend to cancel the positron-hole repulsion. The effective Schrödinger equations do not account for the collective contribution ρ to the induced-charge density [Eq. (3.32)], which ensures that the total induced charge is $-e$. Physically, one expects that ρ_c will be spread over a region of size $\sim r_s$ around the positron. Although the additional Coulomb potential due to ρ_c would be too weak to affect the localized electron orbital $b(r)$, it would be strong enough to prevent the hole orbital from escaping to infinity in the low-density limit.

We may summarize our discussion of the annihilation rate as follows. The total annihilation rate depends only on the shape of the electron wave function near the origin, which in turn is determined by the strong electron-positron attraction. In particular, the total rate is insensitive to the shape of the hole orbital. We have repeated our calculation, assuming that $\tilde{a}(p)=1$ [or equivalently using a trial ground state of the form (2.6)]. For $r_s=4$, we find that R increases by a mere 1%. The partial rate, on the other hand, directly reflects the momentum dependence of the hole orbital. Since the hole extends over a region of space of radius comparable to r_s, the partial rate provides a probe of electron correlations away from the positron. Our discussion suggests that an improved theory of the partial rate could be obtained from a more careful treatment of collective effects, such as outlined at the end of Sec. III.

Next, we discuss the relaxation energy E_c of the positron. According to the discussion in Sec. III, E_c is the sum of the single-excitation contribution E_1 given by Eq. (3.26) and a collective contribution E_{coll} given by Eq. (3.34). We find that for $2 \leq r_s \leq 5$ the single-excitation contribution is nearly constant, $E_1 \simeq -0.29$ Ry. This is consistent with our analysis of the high-density limit carried out in Sec. III. When our estimate (3.34) of the collective contribution is added, we obtain the curve shown in Fig. 4 where the result of Bhattacharyya and Singwi[24] is also plotted for comparison. The agreement is not bad, given the crudeness of our estimate of the collective contribution. It is also interesting to compare our value of E_1 to that obtained by Bergersen,[25] who also used a variational method. His trial function can be described in terms of ours in the following way. If we set $\beta=0$ in our ansatz (2.8) and made the special choice $\tilde{a}(k)^2 \propto \delta(k-k_F)$ for the hole wave function, we obtain the s-wave contribution to Bergersen's trial function. The remaining contributions are similar, but involve partial waves of higher order. There are two main differences between his and our wave functions: (a) due to the lack of hybridization ($\beta\equiv 0$), Bergersen's relaxation energy tends to zero as $r_s \rightarrow 0$ while ours remains finite; (b) the presence of higher-order partial waves in his wave function yields a lower positron kinetic energy, and in the region $r_s > 4$, where hybridization becomes less important, his relaxation energy is lower than our E_1. Since higher-order partial waves do not contribute to the density enhancement at the origin, it is clear that Bergersen's wave functions include some of the collective effects which we have estimated in Eq. (3.34). This again points out the need for an improved treatment of correlation effects.

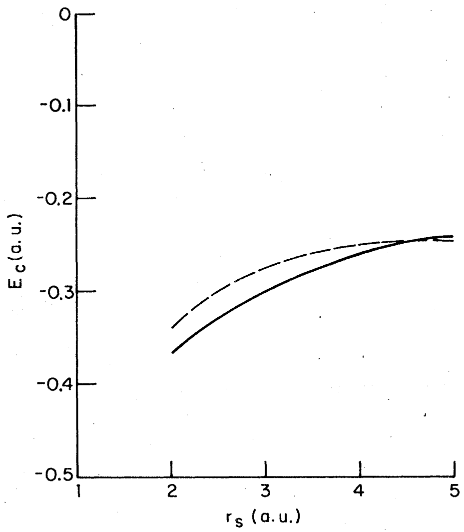

FIG. 4. Relaxation energy (in a.u.) as a function of r_s. The continuous curve is obtained from the present theory, the dashed curve from Ref. 24.

Finally, let us briefly indicate how our variational method could be applied to nonuniform systems. The relative coordinate representation which we have used up to now must of course be abandoned. In the spirit of the Kohn-Sham local density approximation,[26] it is natural to look first at systems in which the electron density is a slowly varying function of position. In that case, one expects that the electronic wave function will be largely determined by the local electron density at the position of the positron. A natural generalization of our simple ansatz (2.6) is then (neglecting spin for simplicity)

$$|\phi\rangle=\int d^3\vec{r}\,\phi(\vec{r})\,\hat{\Psi}_p^{\dagger}(\vec{r})\left[\beta_{\rho(r)}+\int d^3\vec{r}\,'\,b_{\rho(\vec{r}\,')}(r')\,\hat{\Psi}_e^{\dagger}(\vec{r}+\vec{r}\,')\,\hat{\Psi}_e(\vec{r})\right]|0\rangle\quad, \tag{4.2}$$

where b_ρ is the variational wave function appropriate for a uniform gas of density ρ and the subscripts differentiate between electron and positron field operators. Minimizing the energy leads to an effective Schrödinger equation for the wave function of the "dressed" positron. To leading order, the contribution of electron-positron correlations to this effective Schrödinger equation is a local potential $E_r[\rho(\vec{r})]$. Gradient terms will give rise to negative contributions arising from the polarizability of the dressed positron as well as positive contributions coming from the density dependence of the variational wave function. We plan to explore these ideas further in a future publication.

V. CONCLUSION

The main result of this investigation is that a wave function of the form (2.8) containing a single electron-hole excitation can account for most of the correlation effects which are important in positron annihilation. Our approach provides new insight into the nature of these correlations and their effect on observable properties. The success of our theory in explaining the behavior of the total annihilation rate stems from the fact that the density enhancement resulting from the electron-positron attraction is highly localized in space. The relaxation energy and the partial annihilation rate are more sensitive to the long-range tail of the induced density, for which a single-excitation wave function is not adequate. We have argued, however, that collective contributions to the induced density could be treated in linear response theory, since the positron potential is partially screened by the electron-hole excitation present in our variational wave function. A generalization of our variational calculation, including such collective excitations, should shed light on the momentum dependence of the partial annihilation rate.

Since our method is variational, it lends itself to systematic improvement. The emphasis in this paper has been on the uniform electron gas, which best illustrates the basic physical principles. Most important applications, however, involve nonuniformities which arise from band-structure effects or from the presence of surfaces or vacancies. It is hoped that the simplicity and the flexibility of our approach will make it a useful tool for the quantitative analysis of such nonuniform system.

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