

Theory of electron-positron interaction in simple metals: Application to lithium

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In spite of many efforts devoted to this phenomenon, the electron-positron interaction in metals remains an unsolved problem. The development of the partial density amplitude approach to the electronic structure of simple metals offers the possibility to perform direct many-body calculations of the electron-positron interaction in these materials. A theory of this interaction based on the hypernetted-chain approach of Gondzik and Stachowiak is proposed and applied to lithium. It leads to a nonlinear three-dimensional integro-differential equation for the enhancement amplitude. This equation is solved for two cases in which it reduces to one dimension and also for a model. The relation between the results obtained in this way for the annihilation rate and the predictions of the local density approximation and other approaches is studied. The role of core electrons in the interaction is commented.

I. INTRODUCTION

Theoretical investigations of the electron-positron interaction in metals have a long history.¹ However, the complexity of the problem prohibited direct calculations for a metal lattice (investigations in this direction have been performed in Refs. 2–9). On the other hand, puzzles connected with the e^+e^- interaction in an electron gas have gradually been solved (though major controversies still remain concerning the possibility of observing the Daniel-Vosko type distribution in the angular correlation experiment^{10,11,6,12,1}). The results obtained for jellium could be applied to real metals after introducing some oversimplifying approximations for e^+e^- correlations in metal lattices, like the local density approximation (LDA) and its generalizations such as the generalized gradient expansion and the weighted density approximation^{13–17,8} and the references quoted there, also Refs. 18 and 19. Many-body approaches started by Carbotte² used an assumption that can be called constant density potential approximation. In this last case it is assumed that the screened Coulomb potential of the e^+e^- interaction does not depend on the coordinates of the positron in the lattice and is equal to that occurring in jellium of density equal to the average density of conduction electrons in the metal. Recently Sormann⁵ resigned from this approximation, developing the so-called optimized Bloch-modified ladder theory. In the present work an approach is elaborated that allows us to compute effectively annihilation parameters from first principles. Calculations performed for a model (single spherical inhomogeneity in an electron gas) have already shown that the local density approximation for an e^+e^- interaction poorly describes the behavior of the annihilation rate.²⁰

We will express the influence of the positron on the density of conduction electrons in terms of the enhancement amplitude using an approach, which in the case of an electron gas, proved to be particularly simple and efficient.^{21–25} An equation for this quantity is derived in Sec. II. This equation has the form of a three-dimensional nonlinear integro-differential equation. In Sec. III this equation is solved in an approximation that neglects the angular dependence of the

electron distribution around the positron. The generalization of this approximation to anisotropic electron distributions would be affected by the nodes occurring in the density amplitude for conduction electrons. These nodes lead to singularities in the equation for the enhancement amplitude. Since such equations have never been solved, and they describe a phenomenon occurring in all real metals except possibly metallic hydrogen or metallic deuterium (though they are absent in the case of a positron in an electron gas), we devoted Sec. IV to the solution of the equation for the enhancement amplitude for the case of the positron on the nucleus in a form which includes the singularities. This case has a nearly spherical symmetry, and, on the other hand, e.g., Stott and Kubica²⁶ and recently Boroński and Jarlborg⁷ have shown that in lithium the positron density on the nucleus is appreciable.

II. DERIVATION OF THE BASIC EQUATION

This work is based consequently on the formalism elaborated for single impurities in an electron gas by Kallio, Pietenläinen and Lantto^{27,28} (KPL) and modified later by Gondzik and Stachowiak²¹ (see also Refs. 1 and 22–25). It is based on applying the variational principle to the Jastrow-type trial function:

$$\Psi(\mathbf{r}_p; \mathbf{r}_1, \dots, \mathbf{r}_N) = \prod_{k=1}^N R(|\mathbf{r}_k - \mathbf{r}_p|) \prod_{\substack{i,j \\ i < j}}^N f(|\mathbf{r}_i - \mathbf{r}_j|) \times \Phi(\mathbf{r}_1 \dots \mathbf{r}_N), \quad (1)$$

where \mathbf{r}_i and \mathbf{r}_p denote the electron and positron coordinates, respectively. R and f are variational functions-parameters and Φ is the Slater determinant of plane waves.

Following KPL we label this formalism HNC (hypernetted-chain) in spite of its rather distant relation to the HNC equation derived in the theory of liquids (additional details are given in Ref. 1). Its most characteristic feature is to solve a many-body problem in terms of a single density amplitude.

Because of its low mass a positron impurity in a metal differs from heavy impurities in two respects: by being delocalized and by experiencing recoil while interacting with the electrons. According to KPL this last effect can be accounted for in the Euler-Lagrange type equation for the density amplitude of electrons around the positron by replacing the electron mass by the reduced mass of the electron-positron system. Routine methods exist of computing the wave function of a positron in a lattice, and one can believe that they are approximately correct. As concerns electron wave functions, the problem is exceptionally complicated for two reasons:

(1) The effective electron-positron potential is unknown and likely to be different for each position of the positron in the lattice.

(2) The scattering of the electrons on the positron is for this reason complicated, the more so that the effective potential must be self-consistent.

According to Ref. 21, the screening of a positron in an electron gas can be computed from the Euler-Lagrange equation for the enhancement amplitude $w(\mathbf{r})$ (in atomic units, which will be used throughout the paper unless stated differently)

$$[-\nabla^2 + W(\mathbf{r})]w(\mathbf{r}) = 0, \quad (2)$$

where the electron density at distance r from the positron is

$$\rho(r) = w^2(r)\rho_0. \quad (3)$$

ρ_0 is the density of the electron gas into consideration and $w(r) \rightarrow 1$ at an infinite distance from the positron. $W(r)$ is the screened electron-positron potential,

$$W(r) = -\frac{1}{r} + W_p(r) + W_{xc}(r), \quad (4)$$

where $W_p(r)$ is the Coulomb potential of the electronic cloud screening the positron

$$W_p(r) = \rho_0 \int d\mathbf{r}' \frac{w^2(\mathbf{r}') - 1}{|\mathbf{r}' - \mathbf{r}|} \quad (5)$$

and $W_{xc}(r)$ is the exchange-correlation correction to the potential

$$W_{xc}(r) = V_{HL}\{w^2(r)\rho_0\} - V_{HL}\{\rho_0\}. \quad (6)$$

$V_{HL}\{\rho\}$ is the exchange-correlation term in the potential in an electron gas of density ρ . The potential $V_{HL}\{\rho\}$ is assumed in the form proposed by Hedin and Lundqvist.²⁹

Note that some well-known approaches to electron-positron interaction in jellium neglect the exchange-correlation term in the potential due to the screening cloud around the positron. Such is the case of the well-known approach of Kahana³⁰ in which the e^+e^- screened Coulomb potential is assumed in the static random phase approximation (RPA). This leads to the famous low density divergence of the annihilation rate caused by lack of self-consistency. But self-consistency alone cannot lead to the correct behavior of the annihilation rate at low electron densities. This is possible, as shown by Rubaszek and Stachowiak,³¹ only after introducing the exchange-correlation correction. In general, exchange-correlation by decreasing the effective electronic

charge in vicinity of the positron leads to a higher accumulation of electrons on the positron and hence to an increase of the annihilation rate. Omitting this term one gets only half of the spin-averaged positronium value of the annihilation rate in the limit of low electron densities.

The annihilation rates of Refs. 32, 33, and 31 are very close to each other. The approaches they use are very different, but they are similar in using three basic physical ingredients: momentum dependence of electron-positron scattering, self-consistency, and exchange-correlation correction.

Somewhat higher values of the annihilation rate follow from Eqs. (2)–(6). The reason is the Jastrow-type trial function that underlies them and that forces all the electronic states to scatter on the positron in the same way. However, the exchange-correlation term goes beyond the Jastrow-type trial function since it means that e^+e^- correlations depend on the positron coordinates. This guarantees the right behavior of the annihilation rate at high values of r_s . Omitting W_{xc} leads to underestimation of the annihilation rate.

The optimal trial function of the Jastrow type is obtained from the appropriate Euler-Lagrange equation derived by KPL. However, as we mentioned already, this trial function poorly includes electron-electron correlations. This is why Gondzik and Stachowiak replaced the very complicated functional $W(r)$ of KPL by the Kohn-Sham potential (4)–(6). This assumption greatly simplified the calculations and led to physically reasonable results. Its most spectacular confirmation has been provided by Ortiz,³⁴ who calculated the energy of electron-positron correlation in the Jastrow state using the variational Monte Carlo approach.^{34,25} The results of the approach of Eqs. (2)–(6) and of Ortiz are identical as concerns the energy of the e^+e^- correlation. This does not mean, of course, that these approaches are accurate. The approximations involved in using Eqs. (2)–(6) can be estimated owing to the formalism proposed in Ref. 22 (see also Refs. 23 and 25) and called perturbed HNC (PHNC) where the wave function of the system is described by a Slater determinant and the particular electronic states are allowed to scatter on the positron according to equations of the Kohn-Sham type. PHNC is, of course, free of all the approximations performed in HNC calculations. The annihilation rates following from PHNC are a little lower than those of Refs. 32, 33, and 31. This is probably due to allowing for nonorthogonality of one-electron wave functions (following the objections of Lowy and Jackson³²). They remain, however, higher than those of the Boroński and Nieminen formula.³⁵

In the present work an Euler-Lagrange type equation for the density amplitude $\chi(\mathbf{r})$ of conduction electrons in lithium is assumed in the form

$$\left[-\frac{1}{2}\nabla^2 + V^0(\mathbf{r}) \right] \chi(\mathbf{r}) = \eta\chi(\mathbf{r}), \quad (7)$$

where $\chi(\mathbf{r})$ is related to the electron density of conduction electrons $\rho_w(\mathbf{r})$ by the relation

$$\rho_w(\mathbf{r}) = \chi^2(\mathbf{r}). \quad (8)$$

The potential $V^0(\mathbf{r})$ is defined as a functional of χ :

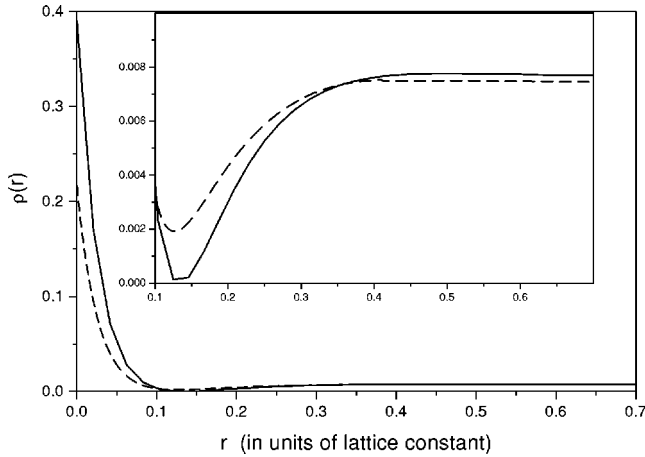


FIG. 1. The comparison of $\chi^2(\mathbf{r})$ (solid line) with $\rho_{FLAPW}(\mathbf{r})$ (dashed line) in Li for the (110) direction. The inset presents the magnified part of $\chi_0^2(r)$ and $\rho_{FLAPW}(r)$.

$$V^0\{\chi; \mathbf{r}\} = V_c(\mathbf{r}) + \int \frac{\chi^2(\mathbf{r}')}{|\mathbf{r}' - \mathbf{r}|} d\mathbf{r}' + V_{HL}\{\rho_c(\mathbf{r}) + \chi^2(\mathbf{r})\}, \quad (9)$$

$V_c(\mathbf{r})$ is the Coulomb potential of the core

$$V_c(\mathbf{r}) = \int \frac{-\rho_n(\mathbf{r}') + \rho_c(\mathbf{r}')}{|\mathbf{r}' - \mathbf{r}|} d\mathbf{r}', \quad (10)$$

$\rho_c(\mathbf{r})$ is the density of core electrons and $\rho_n(\mathbf{r})$ the density of the nuclear charge. The self-consistent potential $V^0\{\chi; \mathbf{r}\}$ is marked by a zero in order to indicate that it is self-consistent only in the meaning of the present approximation. η is the Lagrange multiplier. $\chi(\mathbf{r})$ has the same symmetry as the lattice.

Equation (7) is a nonlinear integrodifferential equation for $\chi(\mathbf{r})$ that we solve by linearization, assuming as approximate solution $\chi_0(\mathbf{r})$ the superposition of free atoms as done usually in band structure calculations, setting

$$\chi(\mathbf{r}) = \chi_0(\mathbf{r}) + \delta\chi(\mathbf{r}). \quad (11)$$

The correction $\delta\chi$ is found to be small, presenting no need for additional linearization. It is computed as a Fourier series over reciprocal lattice vectors \mathbf{G} :

$$\delta\chi(\mathbf{r}) = \sum_{\mathbf{G}} \tau_{\mathbf{G}} e^{i\mathbf{G} \cdot \mathbf{r}}. \quad (12)$$

A self-consistent solution of Eq. (7) is obtained by solving a set of algebraic equations for $\tau_{\mathbf{G}}$'s.

The comparison of the density of conduction electrons in Li obtained in this way with the results of full potential linearized augmented plane wave (FLAPW) calculations (performed according to the WIEN95 code³⁶) is shown in Fig. 1. The solution of Eq. (7) reproduces the main features of the distribution of conduction electrons.

The simplicity of the HNC approach to electron-positron interaction in jellium follows from the fact that Eq. (2) describes a single electronic wave of momentum $\mathbf{k} = \mathbf{0}$ scattering on the positron. In Eq. (7) a single electronic wave $\chi(\mathbf{r})$ describing conduction electrons scatters on a periodic array

of ions. Adding a positron to the metal will disturb the function $\chi(\mathbf{r})$. Taking into account the presence of the positron at \mathbf{r}_p we write the equation for the disturbed density amplitude of conduction electrons ζ in the form

$$\left[-\frac{1}{2}\nabla^2 + V^0(\mathbf{r}) + \frac{1}{2}W(\mathbf{r}_p, \mathbf{r}) \right] \zeta(\mathbf{r}_p, \mathbf{r}) = \eta \zeta(\mathbf{r}_p, \mathbf{r}), \quad (13)$$

where

$$W(\mathbf{r}_p, \mathbf{r}) = -\frac{1}{|\mathbf{r} - \mathbf{r}_p|} + W_p(\mathbf{r}_p, \mathbf{r}) + W_{xc}(\mathbf{r}_p, \mathbf{r}), \quad (14)$$

the Coulomb potential of the screening cloud

$$W_p(\mathbf{r}_p, \mathbf{r}) = \int d\mathbf{r}' \frac{\zeta^2(\mathbf{r}_p, \mathbf{r}') - \chi^2(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}, \quad (15)$$

the exchange-correlation contribution

$$W_{xc}(\mathbf{r}_p, \mathbf{r}) = V_{HL}\{\rho_c(\mathbf{r}) + \zeta^2(\mathbf{r}_p, \mathbf{r})\} - V_{HL}\{\rho_c(\mathbf{r}) + \chi^2(\mathbf{r})\}. \quad (16)$$

The \mathbf{r}_p parameter in Eqs. (13)–(16) simply means that the screening of the positron depends on its coordinates in the elementary cell. Note that for a homogeneous electron gas, Eq. (13) takes the form (2), while in the absence of the positron it takes the form (7). In Eqs. (13)–(16) the screening of the positron is described by one only function ζ depending on the positron coordinates \mathbf{r}_p as on a parameter. This makes achievement of self-consistency in the screening particularly easy. This formalism is likely to describe reality adequately when each conduction electron state gives approximately a similar contribution to the whole electron density and is scattered approximately in the same way by the positron. Alkali metals are the first candidates for applying such an approach. The experience of applying a similar formalism for screening the positron in an electron gas^{21–23,25} and computing the distribution of conduction electrons in lithium (Fig. 1) shows both the reliability and the efficiency of such an approach.

Let us remark that in Eq. (2) the coefficient 1 in the kinetic term is due to the reduced mass of the electron interacting with the positron. If we divide Eq. (2) by 2, the effect of reducing the mass is replaced by a corresponding decrease of the effective charge of the bare positron. However, the effective charge of the electronic cloud screening the positron is also reduced. So, while in Eq. (13) the effective charge of the bare positron is equal 1/2, its screening still needs one electronic charge which enters Eq. (13) with a coefficient 1/2.

Let us remark at this point that both in Eq. (7) and in Eq. (13) it is assumed that core states are not disturbed, neither by the lattice nor by the positron. While the first assumption is generally considered as a good approximation, the positron interaction with the core needs further studies and will be in the following the subject of additional comments.

Let us assume the density amplitude $\zeta(\mathbf{r}_p, \mathbf{r})$ in the form

$$\zeta(\mathbf{r}_p, \mathbf{r}) = w(\mathbf{r}_p, \mathbf{r} - \mathbf{r}_p) \chi(\mathbf{r}), \quad (17)$$

where $w(\mathbf{r}_p, \mathbf{r} - \mathbf{r}_p)$ will be called the enhancement amplitude. In the following we shall mostly use, for convenience,

the shortened notation $w(\mathbf{r}-\mathbf{r}_p)$, indicating instead its dependence on the distance between the electron and the positron.

Introducing Eq. (17) into Eq. (13) we have, owing to Eq. (7),

$$\chi[-\nabla^2+W(\mathbf{r}_p,\mathbf{r})]w-2\nabla\chi\nabla w=0. \quad (18)$$

We will also use Eq. (18) in the form

$$\chi^2[-\nabla^2+W(\mathbf{r}_p,\mathbf{r})]w-\nabla\chi^2\nabla w=0, \quad (19)$$

which is appropriate if $\chi(\mathbf{r})\neq 0$.

Replacing the Eq. (13) for the density amplitude ζ by Eq. (18) for the enhancement amplitude w has several advantages:

(1) We got rid of the lattice potential, which contains an infinite array of singularities.

(2) The region in which the enhancement amplitude $w(\mathbf{s})$ differs from unity is very limited unlike that for the density amplitude $\zeta(\mathbf{r})$ (otherwise we would have to refer to the supercell formalism).

(3) The success of the substitution (17) provides by itself information on the density amplitude ζ . One can expect that the function $w(\mathbf{s})$ is smooth enough, so the density of the screening cloud around the positron at a given distance from it is roughly proportional to the electron density in the absence of the positron, being modulated by the lattice.

Equation (18) is a nonlinear three-dimensional integrodifferential equation for w . We will solve it in some particular cases when it becomes one-dimensional.

III. ISOTROPIC APPROXIMATION

We will compute the function $w(\mathbf{r}_p,\mathbf{r}-\mathbf{r}_p)$ as a function of

$$\mathbf{s}=\mathbf{r}-\mathbf{r}_p. \quad (20)$$

In general the anisotropy of the distribution of conduction electrons $\chi^2(\mathbf{r}_p+\mathbf{s})$ as a function of \mathbf{s} will enforce an anisotropy of the function $w(\mathbf{s})$. We will limit ourselves in this section to an isotropic solution of Eq. (19). We will replace in this last equation the unperturbed density of conduction electrons $\chi^2(\mathbf{r}_p+\mathbf{s})$ by its average over directions of \mathbf{s} :

$$f(s)=\langle\chi^2(\mathbf{r}_p+\mathbf{s})\rangle_{\text{dir}}. \quad (21)$$

Of course, $f(s)$ depends on the vector \mathbf{r}_p , but here and below we will not indicate this dependence in the notation. Examples of the behavior of the function $f(s)$ for Li are shown in Fig. 2 for two values of \mathbf{r}_p .

As far as $f(s)$ is nodeless we can write Eq. (19) in the form

$$-w''-\left(\frac{2}{s}+\frac{f'}{f}\right)w'+W(s)w=0, \quad (22)$$

where the derivatives are taken with regard to s . Equation (22) is solved by linearization. We introduce as usual the substitution

$$w(s)=w_0(s)+\delta w(s), \quad (23)$$

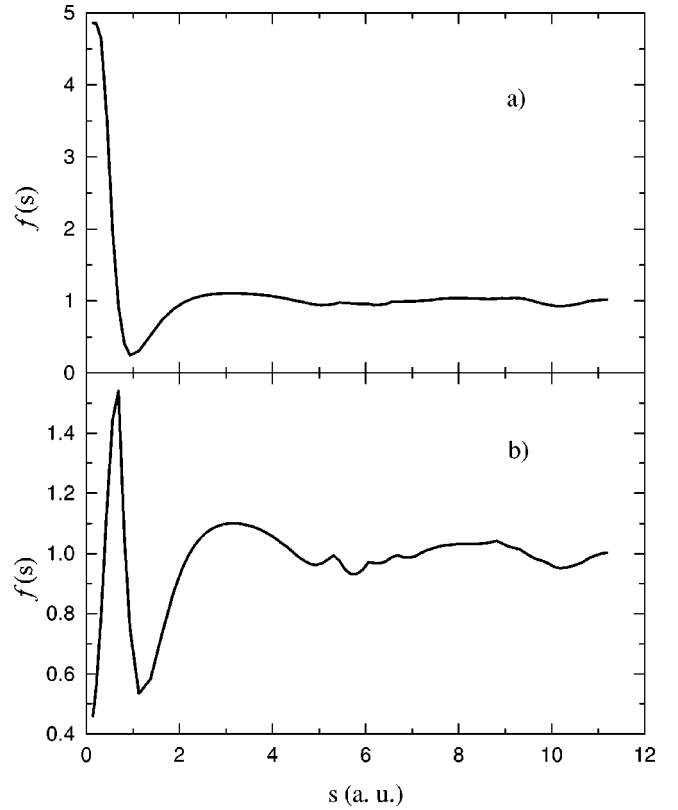


FIG. 2. The average electron distribution $f(s)$ for Li for the positron coordinates: (a) (0.38,0,0), (b) (0.68,0,0).

assuming that $w_0(s)$ is a known function close to the exact solution of Eq. (22) and $\delta w(s)$ is small, so terms of higher order in δw can be omitted.

The equation for δw is solved by assuming that the functions present in it (as well as their derivatives) are sufficiently well described by their values at a finite set of points s_i . This allows us to transform the equation into a set of linear algebraic equations for the values $\delta w(s_i)$. These are easily solved. By repeated linearizations we get an exact solution of Eq. (22), independent of the starting function $w_0(s)$. An additional test for the reliability of the solution is provided by controlling its stability with regard to increasing the density of points s_i .

Equation (22) was solved for Li for 35 values of \mathbf{r}_p along each of the principal axes. For positron positions not very close to the nucleus the solution is stable, i.e., it depends negligibly on the set of points s_i and does not depend on the choice of the starting function $w_0(s)$ (provided this choice is reasonable).

The values of the enhancement amplitude $w(\mathbf{r}_p,0)$ on the positron for \mathbf{r}_p along the principal axes are shown in Fig. 3. One general observation is that the value $w(0)$ varies relatively smoothly throughout the elementary cell (except for the enigmatic region close to the nucleus). This is due to the fact that the function $w(s)$ is characterized by some "rigidity." It is unable to follow small scale variations of the electron density, quite the contrary, to some extent, its behavior is opposite to what one could expect from the local density approximation as will be shown below. It samples rather a considerable region of the lattice enclosed within the screening radius. The crucial point is the size of the screening

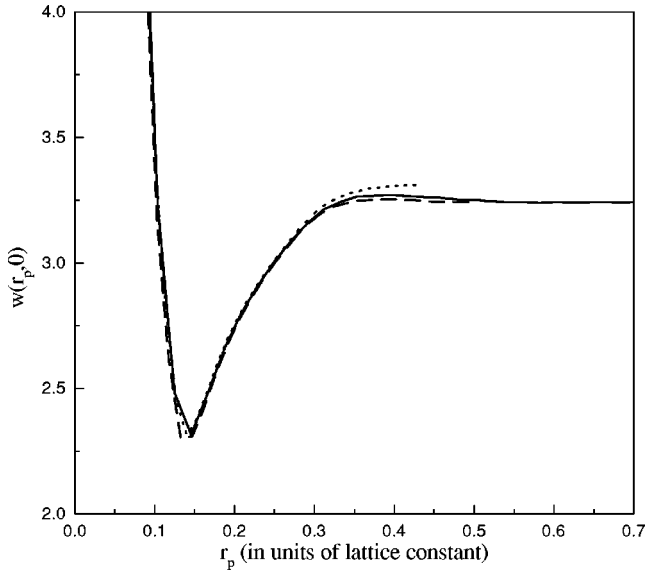


FIG. 3. The values of the enhancement amplitude $w(\mathbf{r}_p, 0)$ on the positron for \mathbf{r}_p along the main crystallographic directions as computed from Eq. (22). The lines are not smooth because we had to limit ourselves (for technical reasons) to $69 \times 69 \times 69$ points in the elementary cell.

cloud. This size is shown in Fig. 9 for the positron on the nucleus. It is visible that even in this extreme case the size of the screening cloud is comparable to the size of the atom. For this reason the amplitude of variation of the average electron density within the screening radius as a function of \mathbf{r}_p cannot be very big. As a result the local annihilation rate λ in surprisingly many cases is roughly proportional to the local electron density in absence of the positron. We should remember, however, that this behavior of λ results from the fact that the function $12/r_s^3$ dominates the function $w^2(\mathbf{r}_p, 0)$ in the following formula for the annihilation rate:

$$\lambda(\mathbf{r}_p) = \frac{12}{r_s^3(\mathbf{r}_p)} w^2(\mathbf{r}_p, 0), \quad (24)$$

where $r_s(\mathbf{r}_p)$ is the local value of the r_s parameter computed according to the formula

$$r_s(\mathbf{r}_p) = \left(\frac{3}{4\pi\chi^2(\mathbf{r}_p)} \right)^{1/3}. \quad (25)$$

The function

$$\lambda_{IPM}(\mathbf{r}_p) = \frac{12}{r_s^2(\mathbf{r}_p)} \quad (26)$$

is the independent particle model (IPM) annihilation rate. For details concerning the independent particle model see Ref. 37. The function $w^2(\mathbf{r}_p, 0)$ exhibits properties that up to now were not investigated so carefully and are in some cases even contradictory to the local density approximation. The local annihilation rate for conduction electrons according to Eq. (24) is presented (by the full curve) in Fig. 4 and compared to the analogical quantity under an assumption which we shall call *constant enhancement approximation* (CEA),

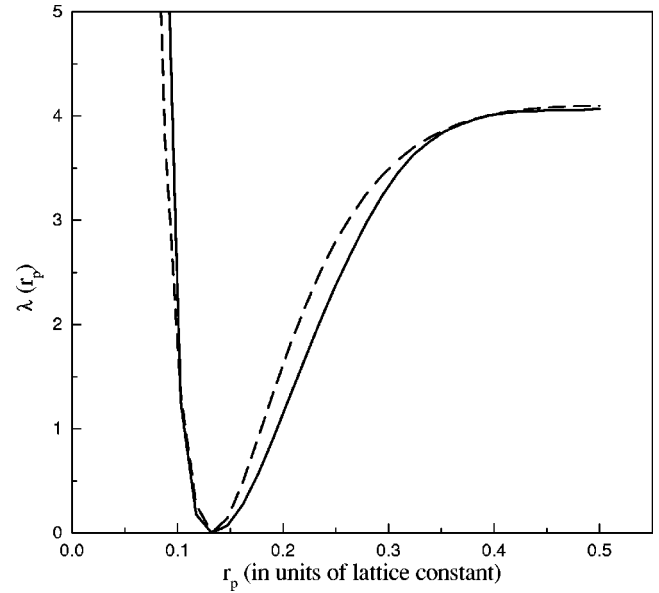


FIG. 4. Local annihilation rates for conduction electrons and positron coordinates along the (100) direction: as follows from Eq. (22) (full curve) and from formula (27) (dashed curve).

which consists in replacing the IPM annihilation rate (26) [e.g., Ref. 33, relation (6.8)] by the formula

$$\lambda_{CEA}(\mathbf{r}_p) = \frac{12}{r_s^3(\mathbf{r}_p)} w_j^2(0), \quad (27)$$

presented by the dashed curve in Fig. 4. $w_j(0)$ is the enhancement amplitude for jellium of density $\rho_0 = 2/a^3$ computed from Eq. (2), where a is the lattice constant. The annihilation rates are expressed as usual in units of $10^9/s$. The local density approximation for the annihilation rate is difficult to present on Fig. 4 since it concerns the whole electron population (core included), however note that it never falls below 2, which is the value for spin-averaged free positronium.¹

In the constant enhancement approximation the spatial dependence of the enhancement does not distort the wave functions of conduction electrons (as observed by positrons) since the enhancement factor is constant throughout the lattice. If one neglects the momentum dependence of the enhancement, which is not considered in the present work this approximation is similar as concerns the angular correlation experiment to the *independent particle model* (IPM). Of course, the predictions of these approximations concerning the positron lifetime would be quite different, but who applies IPM while interpreting the positron lifetime?

Calculations have also been performed for the positron at the center of a model inhomogeneity in which the electron density is given by the formula

$$f(s) = \rho_0 \{ 1 + \sigma \exp[-(s/s_0)^2] \}, \quad (28)$$

where ρ_0 is the density of the electrons for infinite s . Here we will assume it to be equal to the average density of conduction electrons in lithium. σ determines the amplitude of the inhomogeneity and can be positive or negative, provided it is bigger than -1 . The parameter s_0 determines the width of the inhomogeneity. Note that the node of $\chi(r)$ in lithium

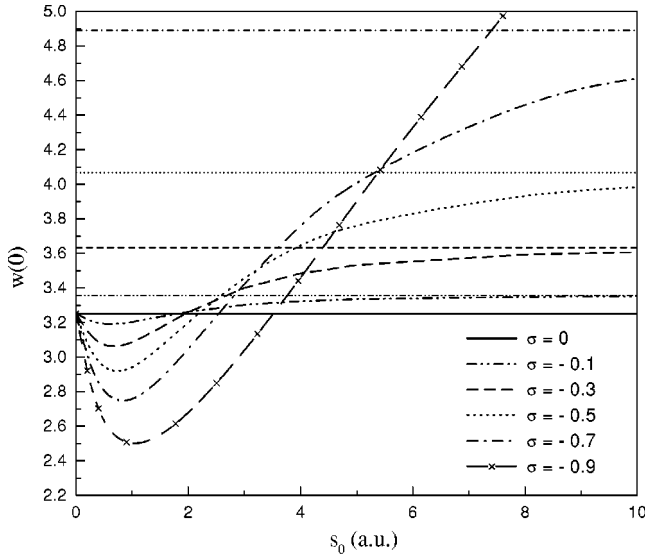


FIG. 5. The enhancement amplitude $w(0)$ (for the positron at the center of the inhomogeneity characterized by parameters σ and s_0) as a function of the parameter s_0 for several negative values of σ (case of a hole in jellium).

occurs at about 0.9 a.u. from the nucleus, the distance between nearest neighbors is equal to 5.713 a.u. This gives an idea about the typical size of the inhomogeneities in this metal. The results are shown in Fig. 5 and Fig. 6. The most striking observation (Fig. 4 for $\sigma < 0$ and Fig. 5 for $\sigma > 0$) is that our enhancement amplitudes as functions of s_0 are not monotonic. This feature is characteristic for all values of σ . The difference between figures corresponding to our approach and the predictions of the local density approximation increases with s_0 for small values of this parameter, with an extremum at about $s_0 = 1$. Then, for $s_0 > 1$ the enhancement amplitudes start to bend towards the appropriate LDA value, approaching it asymptotically.

Such a behavior of $w(s)$ can be explained in the following way for $\sigma < 0$: Since for very narrow holes ($s_0 \rightarrow 0$) the

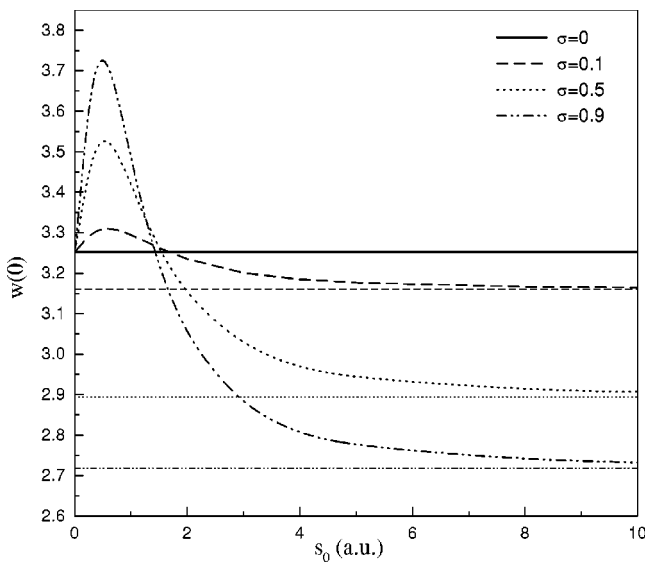


FIG. 6. The enhancement amplitude $w(0)$ as a function of the parameter s_0 for different positive values of σ .

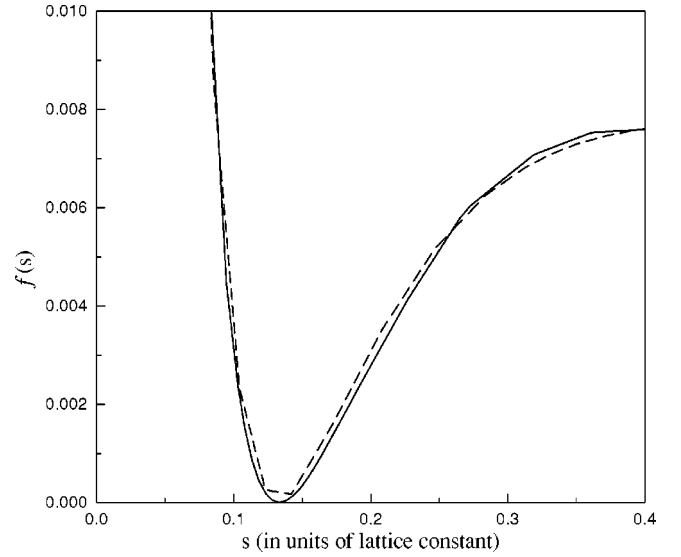


FIG. 7. The function $f(s)$ for $\mathbf{r}_p = \mathbf{0}$ (dashed curve) compared to the function $\tilde{\chi}^2(s)$ (full curve).

distribution of the screening charge resembles the one calculated for $\sigma = 0$, the enhancement amplitude for this case is only weakly affected by the presence of the inhomogeneity. When the hole in the density increases, the screening charge concentrates on the edge of the hole. The inhomogeneity is still too narrow for the screening cloud around the positron to build up inside of it and the correlation keeps the additional charge within the region of higher density—at the edge in case of a hole. However, for a wider hole the screening charge can find a place inside and can fit into it. In this case the screening charge distribution near the center of the hole starts to rise conforming to the expectations of LDA. The same kind of arguments holds for $\sigma > 0$.

IV. INTERACTION IN THE PRESENCE OF NODES

Let us write Eq. (18) in the form

$$[-\nabla^2 + W(\mathbf{r}_p, \mathbf{r})]w - 2\frac{\nabla\chi}{\chi}\nabla w = 0. \quad (29)$$

The density amplitude $\chi(\mathbf{r})$ got one nearly spherical node in each elementary cell, which leads to singularities in Eq. (29).

Let us note that singularities of this kind, unlike in an electron gas, will occur in each metal. For this reason we consider as a necessary step forward in the theory of e^+e^- interaction in metals to solve Eq. (29) at least in one case in which singularities are accounted for.

Let us consider the positron on the lithium nucleus: $\mathbf{r}_p = \mathbf{0}$. Calculations for this case (e.g. Refs. 26 and 7) have shown that in metallic lithium the positron density on nuclei is appreciable. Equation (29) takes the form

$$-w'' - \left(\frac{2}{s} + 2\frac{\tilde{\chi}'}{\tilde{\chi}}\right)w' + W(s)w = 0, \quad (30)$$

where differentiation is over $s = |\mathbf{r} - \mathbf{r}_p|$. We approximated for simplicity the density amplitude $\chi(\mathbf{s})$ by a function of spherical symmetry $\tilde{\chi}(s)$ shown in Fig. 7, neglecting in this

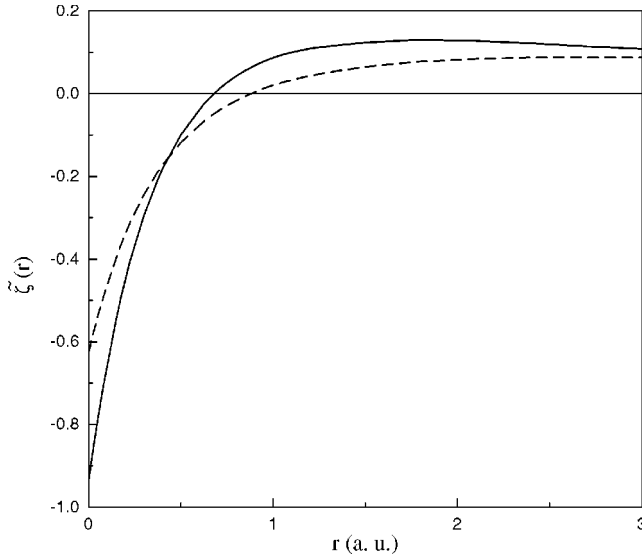


FIG. 8. Density amplitude of conduction electrons for the positron on the nucleus $\tilde{\zeta}(s)$ (full curve) and in the absence of the positron $\tilde{\chi}(s)$ (dashed curve).

way its angular dependence which is small for small values of s and not very important for high values [for large s , $\tilde{\chi}(s)$ approaches $\sqrt{2/a^3}$].

The function $\tilde{\chi}(s)$ has a node at σ_1 while it should be expected that the node of

$$\tilde{\zeta}(s) = w(s)\tilde{\chi}(s) \quad (31)$$

occurs at a different value of s equal σ_0 . In order to satisfy Eq. (31) the enhancement amplitude $w(s)$ must have a singularity at σ_1 . We assume the solution of Eq. (30) in the form

$$w(s) = \frac{\omega_{-1}}{s - \sigma_1} + \tau(s). \quad (32)$$

This allows to solve Eq. (30) exactly.

$\tilde{\zeta}(s)$ is obtained by introducing the solution for $w(s)$ into formula (31). The results are shown in Fig. 8. The enhancement amplitude $w(0)$ is equal 1.50. Since the density of conduction electrons on the nucleus is equal in this formalism to 0.389, $w(0)$ for jellium of this density should be equal 1.51 (in perfect agreement with the local density approximation, provided the density of core electrons is omitted).

The density of the electronic cloud screening the positron is shown in Fig. 9 by the full curve. The density of core electrons is also shown (by the dashed curve). We have also calculated the density of the core polarized by the positron on the nucleus. Note that in our formalism the effective charge of the bare positron is equal to 0.5 electronic charge [Eqs. (13) and (14)], according to the result obtained within the theory of liquids by KLP. For this reason, in order to estimate the polarization of the core by the positron, we assumed that for positron coordinates coinciding with the Li nucleus the effective charge at the nucleus becomes equal to 3.5 [this result can be obtained if one substitutes 0 instead of r_p in Eqs. (13) and (14)]. In such an approach the density of

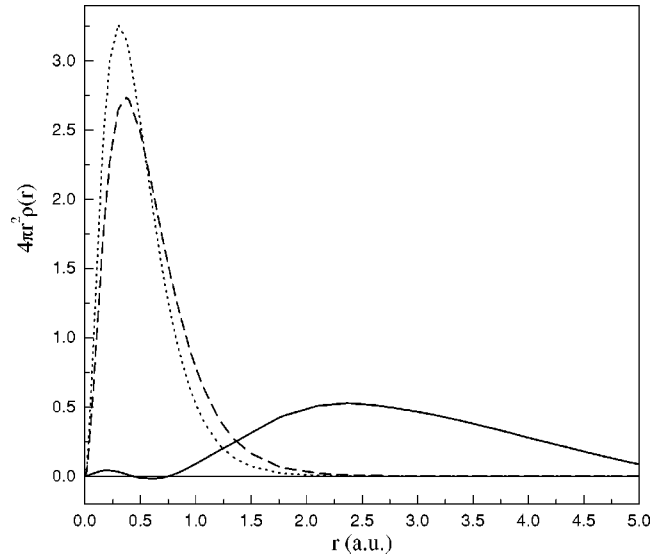


FIG. 9. Electron densities multiplied by $4\pi r^2$. The full curve shows the density of the electronic cloud screening the positron, the dashed curve the density of core electrons and the dotted curve the density of electrons in the core polarized by the positron on the nucleus.

the polarized core was estimated by solving the problem of an atom with a nucleus of charge 3.5 and 4 electrons (one electron corresponding to the screening charge around the positron). Calculations were performed with the help of the commonly used Desclaux program for atoms. The density of the polarized core is shown in Fig. 9 (by the dotted line).

From the above results it is visible that the computations performed in Sec. III are approximate. Indeed, electron-positron interaction displaces the nodes of the density amplitude for conduction electrons. For positron coordinates on the nucleus the node surface approaches the nucleus. Also for other values of the positron coordinates the position of the nodes will be affected. In order to describe the displacement of the node in terms of enhancement amplitude it is necessary to allow in this last function for a singularity as in Eq. (32). Though this problem needs deeper investigations, let us suppose that when the positron coordinates approach the nucleus (from infinity) the node will first withdraw before this particle, but finally the positron coordinates will cross the node. In the absence of the positron the node is situated at a distance σ_1 from the nucleus. In presence of the positron the node surface cuts the straight line connecting the nucleus and the positron at $\sigma_0(r_p)$. The positron coordinate reaches the node surface at some value of r_p , $r_p^0 = \sigma_0(r_p^0) < \sigma_1$. The enhancement amplitude $w(r_p, 0)$ has a singularity on both sides of the node at σ_1 and equals zero at $r_p = r_p^0$. This is not at all the behavior of the enhancement amplitude shown in Fig. 3. Indeed, the amplitudes obtained in Sec. III have a shallow minimum right at σ_1 , i.e., in lithium at a distance 0.13 of the lattice constant from the nucleus.

This shows that it is necessary to perform calculations for a positron in an anisotropic surrounding. This, of course, will make the problem infinitely more complicated mathematically.

V. CONCLUSIONS

Presentation of the electronic structure of simple metals by means of partial density amplitudes introduces simplifi-

cations that make feasible computations of the effect of the electron-positron interaction in metal from first principles, without using approximations that reduce this problem to that of the e^+e^- interaction in an electron gas like the local density, the generalized gradient,⁸ or the weighted density⁹ approximations or very complicated and controversial many-body approaches.²⁻⁵ Of course, the validity of our results depends on the possibility to represent the distribution of conduction electrons in metals by means of a density amplitude. There is no reason to suppose that this cannot be done for all alkali metals.³⁸

In this paper the results of appropriate calculations for lithium are presented. Note that the possibility of describing conduction electrons by means of a single function depends on whether such an assumption is close indeed to reality. If the contribution of higher orbital momentum states (p and d) to the density of conduction electrons is too high, one can wonder if the approach to the electronic structure presented in this paper can be applied to such materials. Anyway, even lithium is infinitely more complicated than the electron gas, so studies of the e^+e^- interaction in this metal are worthwhile.

The appropriate equation (18) for the enhancement amplitude was solved in several cases in which it reduces to one dimension. For definitive conclusions it is necessary to get a more general solution of Eq. (18), which would take account of the anisotropy of the electron distribution around the positron. Note, however, that the problem of screening the pos-

itron in an anisotropic surrounding cannot be avoided if one needs an adequate description of the e^+e^- interaction in metals. Its solution could not be a simple generalization of the calculations performed in Sec. III. Research in this direction is in progress.

The main results of the calculations are the following. For a positron on the nucleus the node surface in the density amplitude approaches the nucleus in comparison to its position in the absence of this particle. The screening cloud is situated mainly outside the core region. So it is a quite good approximation to assume that the core is polarized by a bare positron. The local annihilation rate agrees with the local density approximation provided only the density of conduction electrons is accounted for. For a positron outside the nucleus as well as for models our results agree with the predictions of the local density approximation only for very extended inhomogeneities.

Arguments are found in favor of the constant enhancement approximation for conduction electrons, which as concerns angular correlation is equivalent to the independent particle model. These last results need confirmation by calculations that would allow for anisotropic solutions of the e^+e^- interaction problem.

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- ¹H. Stachowiak and A. Rubaszek, in *Positrons at Metallic Surfaces*, edited by A. Ishii (Trans. Tech. Publications, Aedermannsdorf, 1993), p. 7.
- ²J.P. Carbotte, Phys. Rev. **144**, 309 (1966).
- ³J.P. Carbotte and A. Salvadori, Phys. Rev. **162**, 290 (1967).
- ⁴H. Sormann and W. Puff, in *Positron Annihilation: Proceedings of the 7th International Conference on Positron Annihilation, New Delhi, India 1985*, edited by P.C. Jain and R.M. Singru (World Scientific, Singapore, 1985).
- ⁵H. Sormann, Phys. Rev. B **54**, 4558 (1996).
- ⁶E. Boroński and T. Jarlborg, in *Positron Annihilation*, Vols. 105–110 of Materials Science Forum, edited by Zs. Kajcsos and Cs. Szeles (Trans. Tech., Basel, Switzerland, 1992), p. 607.
- ⁷E. Boroński and T. Jarlborg, Nukleonika **42**, 33 (1997).
- ⁸B. Barbiellini, M.J. Puska, T. Torsti, and R.M. Nieminen, Phys. Rev. B **51**, 7341 (1995).
- ⁹A. Rubaszek, Z. Szotek, and W.M. Temmerman, Phys. Rev. B **58**, 11 285 (1998).
- ¹⁰A.A. Manuel, D. Vasumathi, B. Barbiellini, A. Shukla, P. Suortti, and T. Chiba, in *Proceedings of the International Conference on Positron Annihilation (ICPA-11), Kansas City*, edited by Y.C. Jean, M. Eldrup, D.M. Schrader, and R.N. West, Mater. Sci. Forum **255-257**, 760 (1997).
- ¹¹J.P. Carbotte, in *Positron Solid State Physics*, edited by W. Brandt and A. Dupasquier (North-Holland, Amsterdam, 1983), p. 32.
- ¹²T. Hyodo, T. McMullen, and A.T. Stewart, Phys. Rev. B **33**, 3050 (1986).
- ¹³A. Rubaszek, Phys. Rev. B **44**, 10 857 (1991).
- ¹⁴E. Bonderup, J.U. Andersen, and D.N. Lowy, Phys. Rev. B **20**, 883 (1979).
- ¹⁵B. Chakraborty, Phys. Rev. B **24**, 7423 (1981).
- ¹⁶S. Daniuk, G. Kontrym-Sznajd, J. Mayers, A. Rubaszek, H. Stachowiak, P.A. Walters, and R. West, in *Positron Annihilation*, edited by P.C. Jain, R.M. Singru, and K.P. Gopinathan (World Scientific, Singapore, 1985), pp. 43, 279.
- ¹⁷S. Daniuk, G. Kontrym-Sznajd, A. Rubaszek, H. Stachowiak, J. Mayers, P.A. Walters, and R. West, J. Phys. F: Met. Phys. **17**, 1365 (1987).
- ¹⁸T. Jarlborg and A.K. Singh, Phys. Rev. B **36**, 4460 (1987).
- ¹⁹K.O. Jensen, J. Phys.: Condens. Matter **1**, 10 595 (1989); S. Daniuk, M. Šob, and A. Rubaszek, Phys. Rev. B **43**, 2580 (1991); B. Barbiellini, P. Genoud, and T. Jarlborg, J. Phys.: Condens. Matter **3**, 7631 (1991); M.J. Puska, *ibid.* **3**, 3455 (1991).
- ²⁰H. Stachowiak, in *Positron Annihilation*, Proceedings of the International Conference on Positron Annihilation (ICPA-10), Beijing, 1994, edited by Yuan-Jin He, Bi-Song Cao, and Y.C. Jean, Mater. Sci. Forum **175-178**, 209 (1995).
- ²¹J. Gondzik and H. Stachowiak, J. Phys. C **18**, 5399 (1985).
- ²²H. Stachowiak, Phys. Rev. B **41**, 12 522 (1990).
- ²³H. Stachowiak and J. Lach, Phys. Rev. B **48**, 9828 (1993).
- ²⁴H. Stachowiak, Phys. Rev. B **56**, 4545 (1997).
- ²⁵E. Boroński and H. Stachowiak, Phys. Rev. B **57**, 6215 (1998); see also in *Positron Annihilation*, Proceedings of the International Conference on Positron Annihilation (ICPA-10), Beijing, 1994, edited by Yuan-Jin He, Bi-Song Cao, and Y.C. Jean, Mater. Sci. Forum **175-178**, 891 (1995).
- ²⁶M.J. Stott and P. Kubica, Phys. Rev. B **11**, 1 (1975).

- ²⁷A. Kallio, P. Pietiläinen, and L. Lantto, Phys. Scr. **25**, 943 (1982).
- ²⁸P. Pietiläinen and A. Kallio, Phys. Rev. B **27**, 224 (1983).
- ²⁹L. Hedin and B.J. Lundqvist, J. Phys. C **4**, 2064 (1971).
- ³⁰S. Kahana, Phys. Rev. **129**, 1622 (1963).
- ³¹A. Rubaszek and H. Stachowiak, Phys. Rev. B **38**, 3846 (1988).
- ³²D.N. Lowy and A.D. Jackson, Phys. Rev. B **12**, 1689 (1975).
- ³³J. Arponen, and E. Pajanne, Ann. Phys. (N.Y.) **121**, 343 (1979).
- ³⁴G. Ortiz, Ph.D. thesis, Ecole Polytechnique Fédérale de Lausanne (1992); see also G. Ortiz and P. Ballone, Europhys. Lett. **23**, 7 (1993); G. Ortiz and P. Ballone, Phys. Rev. B **50**, 1391 (1994).
- ³⁵E. Boroński and R.M. Nieminen, Phys. Rev. B **34**, 3820 (1986).
- ³⁶P. Blaha, K. Schwarz, P. Dufek, and R. Augustyn, WIEN95, Technical University of Vienna, 1995 [improved and updated Unix version of the original copyrighted WIEN code, which was published by P. Blaha, K. Schwarz, P. Sorantin, and S.B. Trickey, Comput. Phys. Commun. **59**, 399 (1990)].
- ³⁷S. Berko in *Positron Solid-State Physics*, Proceedings of the International School of Physics ‘‘E. Fermi,’’ Course LXXXIII, edited by W. Brandt and A. Dupasquier (North-Holland, New York, 1983), pp. 64–145.
- ³⁸G. Banach, Ph.D thesis (in Polish), Institute of Low Temperature and Structure Research, Polish Academy of Sciences, 2000.