

Proceedings of the 30th Polish Seminar on Positron Annihilation, Jarnołtówek 1998

A NEW PROJECT FOR CALCULATIONS OF $e^+ - e^-$ INTERACTION IN REAL METALS

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A method based on numerical relaxation technique is proposed for solving directly the three-dimensional Euler-Lagrange equation for the density amplitude of conduction electrons in real metals disturbed by the positron. The method has been tested with good results for the positron in an inhomogeneous electron gas. The preliminary calculations of positron screening in lithium give hope for effective applications to real systems.

PACS numbers: 78.70.Bj, 71.10.-w, 71.60.+z

1. Introduction

Usually, calculating positron annihilation parameters in metals requires an introduction of certain approximations of different kinds. Many of them are connected with the difficulties of taking into account the fact that electron-positron ($e-p$) interaction depends on the positron position in the lattice of the crystal. The simplest and commonly used solution has been local density approximation (LDA) to the enhancement of the electronic density in the positron site (see a review by Šob [1]). These calculations benefit from the results for homogeneous electron gas. The so-called weighted density approximation (WDA) (see e.g., Rubaszek et al. [2]) or generalized gradient approximation (GGA) [3] also is borrowed from theoretical results calculated for the positron in an electron gas (see also [4, 5]). One should realize, however, that above approximations are not always justified, even if they lead to good agreement with experimental data. Moreover, in many cases the results of calculations using these approximations are far away from the experiment.

2. Method

Recently, some attempts have been undertaken in order to explain the complicated problem of $e-p$ interaction in inhomogeneous materials by a direct calculation of the density amplitude ζ of conduction electrons in the presence of the positron [6-9]. The equation for ζ proposed in Refs. [6] and (as a generalization of

the equation for the density amplitude for the homogeneous electron gas [10]) has the following form

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

where

$$W(\mathbf{r}_p, \mathbf{r}) = -\frac{1}{|\mathbf{r} - \mathbf{r}_p|} + W_p(\mathbf{r}_p, \mathbf{r}) + W_{xc}(\mathbf{r}_p, \mathbf{r}), \quad (2)$$

is e-p potential, and η is the Lagrange multiplier. The potential W_p in (2) reads

$$W_p(\mathbf{r}_p, \mathbf{r}) = \int d\mathbf{r}' \frac{\zeta^2(\mathbf{r}_p, \mathbf{r}') - \chi^2(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}. \quad (3)$$

This is the electrostatic part resulting from the perturbation of the electron density by the positron. The exchange-correlation part of the potential W (Eq. (2)) is assumed by using Hedin-Lundqvist formula [11] in the following way:

$$W_{xc}(\mathbf{r}_p, \mathbf{r}) = V_{\text{HL}}\{\rho_c(\mathbf{r}) + \zeta^2(\mathbf{r}_p, \mathbf{r})\} - V_{\text{HL}}\{\rho_c(\mathbf{r}) + \chi^2(\mathbf{r})\}, \quad (4)$$

ρ_c is the density of core electrons, χ is the density amplitude of conduction electrons unperturbed by the positron. It satisfies the following equation:

$$\left[-\frac{1}{2}\nabla^2 + V^0(\mathbf{r}) \right] \chi(\mathbf{r}) = \eta\chi(\mathbf{r}), \quad (5)$$

where $V^0(\mathbf{r})$ is the self-consistent lattice potential. In the density amplitude approximation [12] the function χ is related to the electron density of conduction electrons $\rho(\mathbf{r})$ in the following way:

$$\rho(\mathbf{r}) = \chi^2(\mathbf{r}). \quad (6)$$

If one assumes the density amplitude $\zeta(\mathbf{r}_p, \mathbf{r})$ in the form

$$\zeta(\mathbf{r}_p, \mathbf{r}) = w(\mathbf{r} - \mathbf{r}_p, \mathbf{r}_p)\chi(\mathbf{r}), \quad (7)$$

the following equation for the enhancement amplitude $w(\mathbf{r}, \mathbf{r}_p)$ (for positron considered at \mathbf{r}_p) can be obtained:

$$\rho(\mathbf{r} + \mathbf{r}_p) [-\nabla^2 + W(\mathbf{r}_p, \mathbf{r} + \mathbf{r}_p)] w(\mathbf{r}, \mathbf{r}_p) - \nabla\rho(\mathbf{r} + \mathbf{r}_p)\nabla w(\mathbf{r}, \mathbf{r}_p) = 0. \quad (8)$$

For convenience, we did the transformation $\mathbf{r} - \mathbf{r}_p \rightarrow \mathbf{r}$, $\mathbf{r}_p \rightarrow \mathbf{r}_p$. Of course, the enhancement amplitude w is independent of the electron state (as are the density amplitude ζ and χ).

Despite the approximation connected with dealing with density amplitude instead of working with the wave function, this type of approach has already turned out to be fruitful [10, 13-15].

However, in the case of the inhomogeneous electron density only simple models have been studied, e.g., some solutions have been obtained for the positron in the center of the spherically symmetric inhomogeneity or for the positron on the nucleus or, when the positron was shifted from a symmetric point the spherical averages were used [6-9]. Some new attempts of solving the problem for a simple anisotropic model by expanding $w(\mathbf{r})$ into a series of Legendre polynomials still meet calculational difficulties [16].

In this paper we propose to solve the problem by using a numerical relaxation method, often used for finding numerical solutions of differential equations [17]. This technique has successfully been applied by Puska and Nieminen [18, 19] to calculations of positron wave function in defects. The important advantage of the proposed numerical method is a possibility to apply it, when using Eq. (8), almost directly to arbitrary inhomogeneous systems, e.g., real metals.

Let us describe all the functions and operators in Eq. (8) within a large enough region forming, for convenience, a cube. The positron would always be in its center. Equation (8) is then discretized by using a regular mesh of n^3 points in which the values of the functions $w(r, r_p)$, $\rho(r + r_p)$, $W(r_p, r + r_p)$ are just w_{ijk} , ρ_{ijk} , W_{ijk} . We assume that $w(r, r_p) = 0$ at the borders of the cube. The Laplace operator at a given point $\{ijk\}$ is given by the functions w at this point and six neighboring points (h is the distance between the mesh points)

$$\nabla^2 w|_{ijk} = (w_{i+1,j,k} + w_{i,j+1,k} + w_{i,j,k+1} + w_{i-1,j,k} + w_{i,j-1,k} + w_{i,j,k-1} - 6w_{i,j,k})/h^2 \quad (9)$$

and scalar product of gradients in Eq. (8) is

$$G_{ijk} = [(w_{i+1,j,k} - w_{i-1,j,k})(\rho_{i+1,j,k} - \rho_{i-1,j,k}) + (w_{i,j+1,k} - w_{i,j-1,k})(\rho_{i,j+1,k} - \rho_{i,j-1,k}) + (w_{i,j,k+1} - w_{i,j,k-1})(\rho_{i,j,k+1} - \rho_{i,j,k-1})]/4h^2. \quad (10)$$

Then Eq. (8) can be presented in the following numerical form:

$$w_{ijk}^{m+1} = (w_{i+1,j,k}^m + w_{i,j+1,k}^m + w_{i,j,k+1}^m + w_{i-1,j,k}^m + w_{i,j-1,k}^m + w_{i,j,k-1}^m + G_{ijk}^m \rho_{ijk})/(h^2 W_{ijk}^m + 6), \quad (11)$$

where the index m corresponds to m -th iteration cycle. The function $w(r, r_p)$ at $r = 0$ can be found by using the cusp condition [20] which for the case of enhancement amplitude reads

$$\left. \frac{dw(r)}{dr} \right|_{r=0} = -\frac{1}{2}w(0) \quad (12)$$

and takes here a simple numerical form

$$w_{i_0,j_0,k_0}^m = \frac{w_{i_0+1,j_0,k_0}^m}{(1 - h/2)}. \quad (13)$$

Additionally, one requires full screening of the positron, i.e., electronic charge shifted by the positron should be equal to 1

$$\int [w^2(r) - 1]\rho(r)dr = 1. \quad (14)$$

This yields a prescription for normalization of w , if the integral (14) gives a constant a instead of 1 then

$$w_{ijk}^{\text{normalised}} = \sqrt{(w_{ijk}^2 - 1)/a + 1}. \quad (15)$$

The potential W_{ijk} is calculated initially on the base of some approximated values of w (e.g., from the solution for the positron in the homogeneous electron gas).

The self-consistent solution can be obtained by successive iterations of (11) and the potential is found on a base of results of the preceding iteration. In each iteration cycle Eqs. (13) and (15) are used. Unfortunately, the more elegant method as linearization of the problem and solving a set of corresponding linear equations as was done in e.g. Refs. [6] and [8] for spherically symmetric w is, however, in practice not feasible within this scheme since it is strongly limited by the capacity of the computer.

3. Calculations and results

We have initially assumed that the sufficient diameter of the considered cubic region may be ~ 40 a.u. The maximum h we use is ~ 0.5 a.u. In calculations in Refs. [10] and [6] for the homogeneous electron gas the maximum radius r for the function $w(r)$ was ~ 20 a.u. For such distances this function vanishes and then gives the practical limits for the integrations (e.g., in the requirement (14)). The accuracy of the calculations depends much on the way of computing the function w close to $r = 0$. In order to achieve enough accuracy we enlarge the number of considered points at the cusp by constructing additional overlapping cubic regions, each with regular mesh of points, condensing them always twice for the regions closer and closer to $r = 0$. Fortunately, the value of the derivative of $w(r)$ near to $r = 0$ is proportional to $-1/2$ only so the cusp possesses a quite gentle character $w(r)|_{r \rightarrow 0} = w(0)[1 - (1/2)r]$. When calculating the screening charge according to Eq. (14) we use, however, the following ansatz to improve the accuracy:

$$\int [w^2(r) - 1] \chi^2(r) dr = \int \{ [w^2(r) - 1] \chi^2(r) - [w^2(0) - 1] \exp(-r - r^2/10) \} dr + \int [w^2(0) - 1] \exp(-r - r^2/10) dr. \quad (16)$$

The similar method is applied to calculations of the electrostatic part of the potential W_p . The computation of the potential is the most time-consuming part of the calculations (it requires performing $n^3 \times n^3$ operations).

3.1. Tests

In order to check the quality of the calculations we have, at the beginning, performed the test for the case of the positron in the homogeneous electron gas and compared the results with the ones obtained with the method of Gondzik and Stachowiak [10]. Of course, the number of iterations needed to achieve self-consistency depends on the starting function w_0 , generally we need from a few dozens up to a few hundreds of iterations to get a satisfactory result ($\sim 1\%$). The result of the test is presented in Fig. 1. Similarly, we got quite good agreement with the results of calculations ([6] and [8]) for different sizes of spherically symmetric Gaussian-type inhomogeneity with the positron situated in the center.

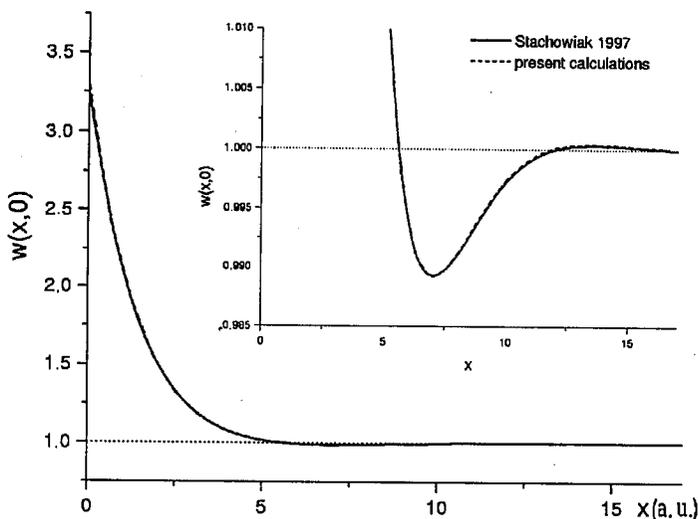


Fig. 1. The enhancement amplitude $w(r, 0)$. The solid line is the result of calculations according to Ref. [6]. The dashed line corresponds to present calculations. The inset is the magnified part of the figure.

3.2. Inhomogeneous electron gas

It is very interesting to investigate the dependence of $w(r, r_p)$ on r_p in the case when the positron coordinates change with respect to the inhomogeneity. We have found solutions for the simple model where the electron density is given by the following function:

$$\rho(r) = \rho_0 \{1 + \sigma \exp[-(r/s_0)^2]\}. \quad (17)$$

The obtained function is evidently anisotropic, although this anisotropy is much weaker than it could be expected (Fig. 2). In Fig. 3 we present the dependence of contact value $w(0, r_p)$ on the positron distance from the center of the inhomogeneity. This dependence is strong and for the inhomogeneity of relatively small size ($s_0 = 1$ a.u., $\sigma = \pm 0.7$) and for small values of r_p it is even contradictory to what could be suggested by the LDA approximation. This effect appears owing to the role of the gradient term in Eq. (8) which is sensible to quick variations of the density usually produced by an external potential. Only when the positron distance from the inhomogeneity becomes large (order of a few sizes of the inhomogeneity) the behavior of the function $w(0, r_p)$ is in good agreement with the LDA results.

3.3. Lithium

We have solved Eq. (8) for a more realistic case. We have chosen metallic lithium. In the approximation used the density amplitude is equal to the square root of the density of conduction electrons. This density has been calculated with the FLAPW method (full potential linearized augmented-plane wave) formalism (Blaha et al. [21]) with frozen core approximation. Of course, no nodes occur in the density amplitude calculated in that way. The result of our preliminary calculations

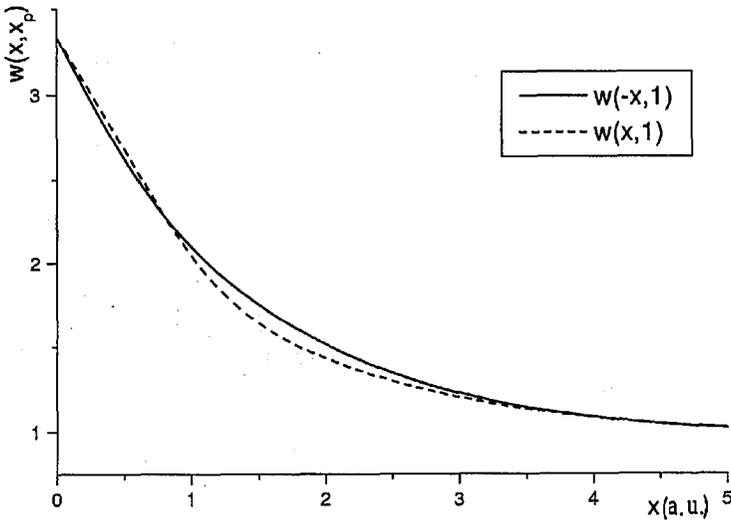


Fig. 2. Left and right hand side (x , $-x$) of the enhancement amplitude $w(r, r_p)$ (along $[1,0,0]$ direction) for $r_p = (1, 0, 0)$ a.u., $\sigma = -0.7$, $s_0 = 1$ a.u.

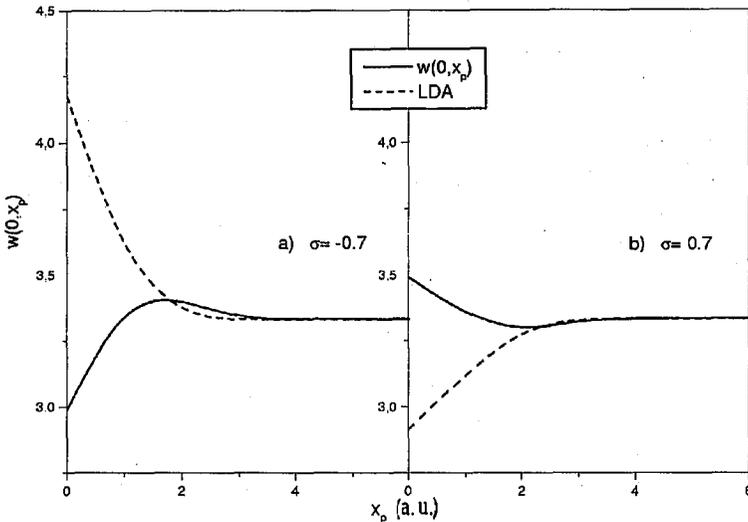


Fig. 3. The contact function $w(0, r_p)$ (along $[1,0,0]$ direction). The dashed and solid lines correspond to LDA approximation and the presented method, respectively; (a) $\sigma = -0.7$, $s_0 = 1$ a.u., (b) $\sigma = 0.7$, $s_0 = 1$ a.u.

for the positron situated at the center of the face of the elementary cell (bcc) can be seen in Fig. 4 (x, y plane). We have magnified the lower part of the figure to see better the details beside the peak. At first view, a quite strong anisotropy and the places where ions occur are clearly seen. The anisotropy becomes large for $r \geq a_0/2$ (a_0 is the lattice constant, here $a_0 = 6.597$ a.u.). It is evident that along some directions the function w vanishes relatively quickly and along the others

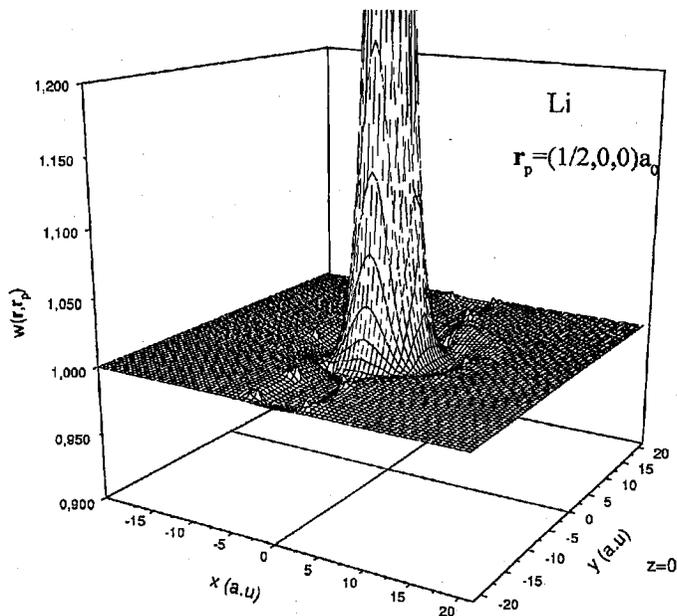


Fig. 4. The enhancement amplitude $w(r, r_p)$ for lithium ($z = 0$ plane). The positron coordinates are $r_p = (1/2, 0, 0)a_0$ (face of the elementary cell).

the e-p interaction propagates much far away than it could be expected on the base of the results for the electron gas of corresponding density. At the places of ions we can observe quick changes of $w(r)$ like small “craters”, they result from the great variations of the electron density near the ions and the gradient term in Eq. (8) is responsible for this effect. The calculations performed without this term do not create these bumps. The value of $w(r, r_p)$ in the peak is about 5% lower than calculated for the electron gas of density corresponding to the mean value of the real density in the cell and $\sim 3\%$ less than expected on the base of LDA.

4. Conclusions

We present a practical scheme making possible the self-consistent calculation of the enhancement amplitude (so, in this approximation the e-p correlation function) for the arbitrary system with inhomogeneous density of itinerant electrons. The real metals are the main goal of the calculations. The method gives a possibility to investigate the influence of nonlocal effects on the enhancement. The results show clearly that the figures obtained when the enhancement amplitude is actually the **functional** of the electron density may be in some cases contradictory to those found within LDA (and probably also WDA) when the enhancement is only a function of local (or an effective) value of the electron density at the positron. This has been also confirmed by other recent works concerning this problem [8, 16].

In application to lithium the above method (though preliminary and not fully accurate yet) was able to yield for the first time an image of the enhancement calculated directly in the real metal, without using homogeneous electron gas results

and without making any additional approximations in Eq. (8). This 3-dimensional enhancement amplitude exhibits an interesting and complicated structure impossible to get with conventional methods. For now, the weakness of the method is its relatively slow convergence, however, some studies are undertaken to make it better. Also, it is important to improve the accuracy of the calculations in the neighborhood of ions in the lattice. The comparison to experiment can be possible after calculating the function $w(r, r_p)$ for sufficiently large number of points r_p in the elementary cell of the metal.

Acknowledgments

This work benefited from grant No. 2 P03B 099 12 of the Committee for Scientific Research (Poland).

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