Positron-electron correlation energy in an electron gas according to the perturbed-hypernetted-chain approximation

E. Boroński and H. Stachowiak

W. Trzebiatowski Institute of Low Temperature and Structure Research, Polish Academy of Sciences, P.O. Box 937,
50-950 Wrocław 2, Poland

The electron-positron correlation energy in an electron gas has been calculated many times, and quite different results have been obtained. Therefore we found it useful to compute correlation energies using the more recent approach of Stachowiak labeled PHNC (perturbed hypernetted chain) which is self-consistent and describes quite well the experiment as concerns partial annihilation rates, at least in simple metals. This approach passed the additional test successfully, leading to reasonable results for the correlation energy in the whole range of investigated electron densities ($0.1 < r_s < 25$). [S0163-1829(98)05704-X]

The positron annihilation method has become an important tool for investigation of the electronic structure of solids. In particular, in the last few years it has been commonly used in studies of semiconductors and high-T_c superconductors, in addition, many metals have been investigated (see, e.g., Refs. 1–3 and references therein). However, because of the electron-positron interaction which itself affects the information on electronic structure coming from experiments, it is necessary to pursue studies widening the theoretical ground for proper interpretation of the data.

Since the work of Kahana[4,5] (cf. also Ref. 6), who proposed using the Bethe-Goldstone equation for the positron in an electron gas, and was the first to obtain reasonable theoretical results for $2 < r_s < 4$, many approaches have been presented describing the electron-positron (e-p) interaction in jellium (see Ref. 7). We believe that three features should be required from a good theory of this effect, namely, self-consistency, determination of momentum dependence, and presence of three-particle correlations. As far as we know only four approaches satisfy these requirements. They are due to Lowy and Jackson[8] (see also Lowy[9]) who used the Lippman-Schwinger equation, Arponen and Pajanne (AP) (Refs. 10 and 11) who diagonalized the Hamiltonian rewritten in terms of Sawada boson operators, Rubaszek and Stachowiak[12] who obtained a self-consistent solution of the Bethe-Goldstone equation developing the approach started by Kahana,[5] and Stachowiak,[13] who used the approach labeled conventionally perturbed hypernetted chain (PHNC).

All these approaches, except the one of Arponen and Pajanne, use the two-body approximation, separating the motion of the positron and one particular electronic state in their mutually screened Coulomb potential from the whole system. These approaches lead to different predictions concerning measurable quantities. Differences are quite drastic in the case of partial annihilation rates. For more information on this subject cf. Refs. 7, 14 and 15, also Fig. 1.

Among authors of the four theories mentioned above, however, only AP calculated electron-positron correlation energies (in the limited range of $1.5 < r_s < 8$, and additionally, for the positronium ion Ps^-). The electron-positron correlation energy may be defined as the change of the energy

of a system consisting of many electrons after introducing a positron. In the present paper we will compute this quantity within two approaches [HNC (Ref. 16), and PHNC (Ref. 13)]. The reasons for performing such calculations are the following.

(i) The most reliable calculations of this quantity up to now are based on the AP approach. Nobody up to now has succeeded in finding satisfactory values for the correlation energies within any of the two-body theories mentioned above, although it follows from experimental values that some of them give a better description of momentum dependent enhancement. Hyodo, McMullen, and Stewart[14] wrote, ''among those calculations, Kahana . . . and some of the ex-
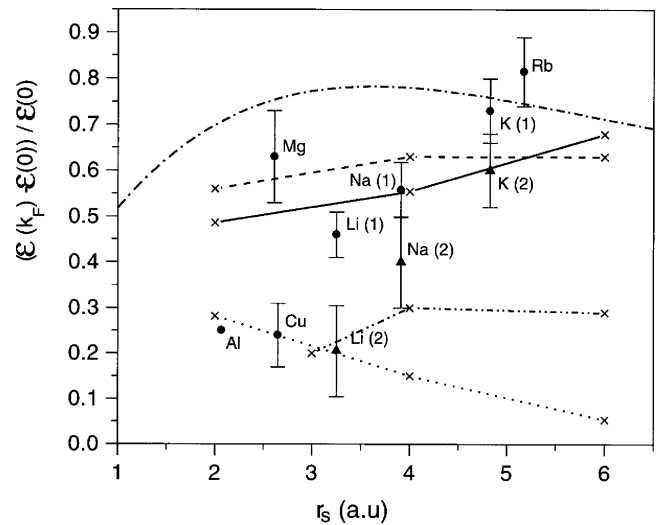

FIG. 1. Relative enhancement factors $[\varepsilon(k_F) - \varepsilon(0)]/\varepsilon(0)$. The dotted curve refers to the original results of Arponen and Pajanne (Ref. 11). The solid curve is obtained by renormalizing these values according to Boronski (Ref. 37). The dashed curve corresponds to the results of Rubaszek and Stachowiak (Ref. 38). The dashed-dotted curve refers to the PHNC (Ref. 13). The dashed-double-dotted curve corresponds to the results of Lowy (Ref. 9). The references to the experimental values are as follows: Al—Ref. 39; Cu—Ref. 40; Mg—Ref. 41; Li(1), Na(1), K(1), and Rb—Ref. 42; Li(2), Na(2), and K(2)—Ref. 43.

TABLE I. The values of electron-positron correlation energies calculated within the PHNC for given electronic densities expressed in terms of r_s.

r_s	0.1	0.2	0.3	0.5	0.7	0.9	1.0	1.3	1.5	1.8	2.0
E_c	-3.414	-2.515	-1.915	-1.460	-1.192	-1.026	-0.966	-0.834	-0.774	-0.705	-0.670

r_s	2.5	3.0	3.5	4.0	5.0	6.0	8.0	10.0	15.0	20.0	25.0
E_c	-0.603	-0.557	-0.524	-0.501	-0.470	-0.451	-0.437	-0.437	-0.452	-0.468	-0.483

tensions . . . give results close to the present result . . . ;'' see also Ref. 15.

(ii) Experimental needs connected with practical applications in studies of the electronic structure create the necessity to develop the theory of electron-positron interaction for real (i.e., different from the electron gas) metals. Up to now, existing applications were connected mostly with *ad hoc* approximations difficult to check, like the constant density potential or the local-density approximations.[17] The applicability of these approximations is problematic.[18] Unlike many other approaches (cf. Ref. 19), the HNC and PHNC, because of their simplicity and efficiency, give the hope to perform *direct* calculations of annihilation characteristics in metal lattices. In particular, the PHNC leads to reasonable annihilation rates and enhancement factors for the electron gas[13] (if agreement with measurements performed in simple metals can be considered as a good criterion). In the present work we will perform the correlation energy test for this approach. Let us point out that the real problem is not the e-p interaction in an electron gas (this is more or less understood) but in more complicated systems particularly including transition metals where theoretical predictions fail to lead to agreement with experiment.[20]

The importance of performing the correlation energy test for the PHNC is visible, if only from recent literature. Barbiellini *et al.*,[21] while studying positron annihilation in real metals, applied the approach of Arponen and Pajanne,[10] which otherwise disagrees with experiment, only because in approaches agreeing with experiment correlation energy calculations have not been performed: "However, in order to use an enhancement factor consistent with the correlation energy, we introduce the function . . . based on the data by Arponen and Pajanne."

Quite a few papers devoted to correlation energy calculations have been published earlier, but either they applied simple linear-response theories[22–24] or were incomplete or not self-consistent,[25–27] and the results often diverged. Recently, Ortiz and Ballone[28] undertook an attempt to compute e-p correlations by the variational Monte Carlo method.

Thus in this work we present electron-positron correlation energies calculated by using the methods from Refs. 16 and 13, and compare them with the ones calculated from earlier theories. These methods are numerically exceptionally efficient. Some authors consider that such an advantage is not essential. However, the efficiency of the PHNC approach made it possible to calculate annihilation parameters in a much wider range and a computer time shorter by a factor of 100 in comparison with the self-consistent Bethe-Goldstone approach.[12] Only this allowed us to perform the calculations presented in this paper *at all*. The formula for the correlation energy is based on the Feynman-Hellman theorem and reads (see, e.g., Refs. 29–31)

$$E_c^{+-}(n_0) = -\frac{3e^2}{r_s^3}\int_0^1 dZ \int_0^\infty dr\ r[g_{+-}(r,n_0,Z)-1],$$

(1)

where $g_{+-}(r,n_0,Z)$ is the electron-positron correlation function for a charge Z of the positive particle and an electron density n_0. Z is considered here as a coupling constant of the interaction. The above formula solves the problem, the point is in the practical determination of $g(r,n,Z)$. The scheme of the calculations is the following.

(i) We compute self-consistently for given n_0 and Z the HNC density amplitude $w(r)$, the appropriate correlation function $g(r,n,Z)$ and the potential $V_0(r)$ according to Eqs. (10), (11), and (22)–(27) in Ref. 16.

(ii) On the basis of $w(r)$ and according to Eqs. (2.1)–(2.5), (4.12), and (5.1) in Ref. 13 we obtain the PHNC positron-electron correlation functions $g^{\text{PHNC}}(r,n,Z)$. It is worthwhile to remark at this point that the physical assumptions underlying the HNC and PHNC are quite different. We consider as an achievement of the theory of liquids [especially as concerns the works of Kallio, Pietiläinen, and Lantto (KPL) (Ref. 27)] that the e^+-e^- correlation function $w^2(r)-1$ could be obtained in such a simple way with quite good accuracy (this concerns in particular the approach of Gondzik and Stachowiak[16]). This fact is the source of the efficiency of the PHNC. The main difference between the approach of Ref. 16 and KPL consists in the neglect of the Pauli term in the potential (in the meaning of Ref. 32), while
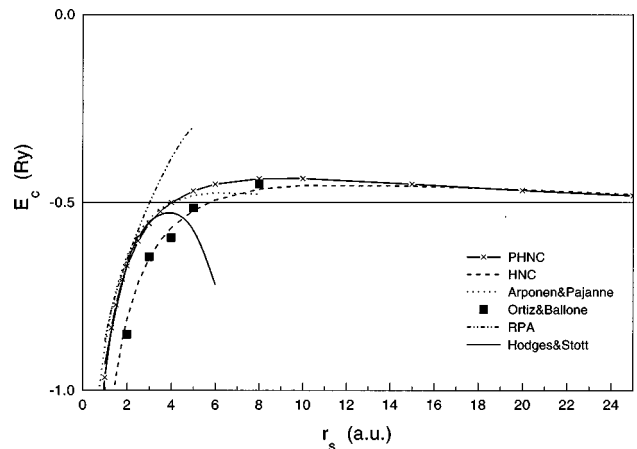

FIG. 2. The electron-positron correlation energy as a function of r_s. The curves denoted as PHNC (full curve plus small crosses) and HNC (dashed curve) show the results obtained in this work. The dotted curve corresponds to the interpolated results of Arponen and Pajanne (Ref. 10). The full squares refer to Monte Carlo calculations of Ortiz and Ballone (Ref. 28). The curve RPA corresponds to linear-response result (Ref. 22). The curve Hodges&Stott is the result of Ref. 23.
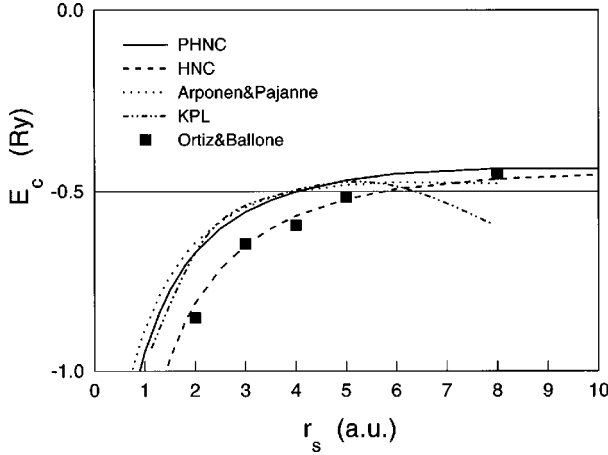
FIG. 3. Magnified part of Fig. 2 for $0 < r_s \lesssim 8$. The curves are labeled as in Fig. 2, but instead of the results of RPA (Ref. 22) and Hodges and Stott (Ref. 23), we present the curve corresponding to the results of Ref. 27.

the trial function of KPL neglects the influence of the positron on the $e^+ - e^-$ correlation function.

(iii) According to formula (1), we find the correlation energy.

In this way, we calculated the density distribution for 22 values of r_s ($0.1 < r_s < 25$), (see also Table I) and for 11 values of Z ($0.005 < Z < 1$) according to the HNC and PHNC (in total for 242 values of the pair r_s and Z). The results are presented in Figs. 2 and 3.

Concerning the HNC results, Fig. 2 shows that the corresponding curve behaves quite well for lower densities, while the values of E_c seem to tend asymptotically to the value for the Ps atom. In the high-density limit, however, the absolute values of the correlation energy are higher than the random-phase-approximation (RPA) results: the difference between the RPA and HNC increases when r_s approaches zero. This result is not surprising since it is known that the HNC (cf. Balescu[33] and Croxton[34]) is a low-density approximation.

As concerns PHNC, in the high-density limit the E_c curve fits the RPA results well, while in the low-density limit it approaches the value -0.5 Ry (Ps), reproducing the low-density results of HNC. No divergence of the kind presented in Ref. 35 is observed up to $r_s = 25$ [$E_c(r_s = 25) = -0.483$ Ry] which is the highest value of r_s for which calculations have been performed.

For practical purposes we can interpolate our values for E_c (corresponding to the PHNC approximation) with the formula

$$E_c(r_s) = \frac{a}{(r_s - d)^2} + \frac{b}{(r_s - d)} + c. \qquad (2)$$

The parameters a, b, c, and d are given in Table II. The above formula is valid for $r_s > 0.1$. Since our values approach the RPA results of Arponen[36] for small r_s, when $r_s < 0.1$ one can use the formula obtained by this author:

$$E_c(r_s) = -1.56/\sqrt{r_s} + (0.051 \ln r_s - 0.081)\ln r_s + 1.14. \qquad (3)$$

TABLE II. The parameters a, b, c, and d corresponding to various ranges of electronic densities expressed in terms of r_s. The parameters in the fourth line provide a slightly worse interpolation, but are valid in a wider range of r_s.

Range	a	b	c	d
$0.1 < r_s < 1$	0.033 921 278	-0.696 800 3	-0.350 969 5	-0.07
$1 < r_s < 8$	-2.385 415	0.173 216 7	-0.456 261 3	-1.0
$8 < r_s < 25$	-0.262 390 1	3.958 436	-5.858 743	-4.0
$0.2 < r_s < 8$	-1.428 222	-0.171 569 8	-0.392 759 4	-0.67

It remains a question whether the PHNC and HNC absolute values of E_c are not too small for lower densities. The only results we could compare to are those of Arponen and Pajanne. Comparing to Table II in Ref. 10, it appears that for $r_s > 5$ the PHNC results are close to the AP data calculated in one-boson Tamm-Dancoff approximation (only one electron and the positron are excited at a given moment). This corresponds in some sense to neglecting the possibility of binding two electrons to the positron (this occurs in the case of the Ps$^-$ ion).

One particular property of the PHNC and AP curves is that they cross the value of -0.5 Ry corresponding to free positronium almost exactly at the same electron density ($r_s = 4$). The PHNC curve exhibits the same peculiar feature as the AP curve, namely, the energy does not monotonously approach the Ps or Ps$^-$ value (the correlation energy for a proton is a monotonous function of the electron density[11]), but has a minimum (in absolute values) connected , according to the AP interpretation, to the positron recoil.

As concerns the results of other authors presented in Figs. 2 and 3: most of these approaches do not yield correct correlation energies for $r_s > 6$. That is, the first attempts[22,23] based on the linear response of the electron system to the positron led to correlation energies vanishing to zero with r_s. On the other hand, results based on the theory of Sjölander and Stott[25] exhibit a divergence of the correlation energy for $r_s > 6$. The results of Ortiz and Ballone[28] are given only in a few points in the range $1 < r_s < 8$ and, generally, are close to our HNC values and quite far from Kallio, Pietiläinen, and Lantto[27] for higher densities (in this region KPL results are, astonishingly, quite close to our PHNC values). This is surprising if one takes into account that in Refs. 27 and 28 the same trial function is used, while in our calculations the potential in the Euler-Lagrange equation is replaced by the Kohn-Sham potential (this replacement can be justified in view of the article of Holas and March[32] with the Pauli potential neglected). This is an argument in favor of the approach used by Gondzik and Stachowiak.[16]

Summarizing, we show results for correlation energy in an electron gas obtained within the HNC and PHNC approaches. The values given by the latter method are presented in convenient form using an interpolation formula. They are satisfactory in the whole range of electron densities and are obviously better than the results denoted in Figs. 2 and 3 as RPA, Hodges&Stott, and KPL because these curves exhibit wrong tendencies for higher r_s, and are only fragmentary. As concerns the comparison to the results of Arponen and Pajanne, our values agree quite well. Thus in the

light of the discussion of shortcomings of other approaches we consider our results as an additional confirmation of the consistency and efficiency of PHNC. They can be useful in calculations of positron annihilation parameters in real metals (some intermediate quantities as, e.g., $g[|\mathbf{r}_+ - \mathbf{r}_-|, n_-(\mathbf{r}_+), Z]$ needed to calculate the correlation energy may be used in projects basing on better approximations than the local-density approximation). The PHNC itself gives hopes to be a convenient tool for future applications in real metals.

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