NUKLEONIKA 2010;55(1):9–12

ORIGINAL PAPER

Positron lifetime calculations for some elements on the base of the GGA-PHNC approximation

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Abstract. In studies of several interesting materials by positron annihilation technique the theoretical values of positron lifetimes in bulk and defected elements are useful. These lifetimes are often calculated within the GGA approximation. In this approximation the results of Arponen and Pajanne calculations for the positron in an electron gas are used. It is known, however, that when using the LDA approximation for the calculations of positron lifetime in real metals, Arponen and Pajanne values yield worse agreement with experimental data than the results obtained on the basis of PHNC formalism. Therefore, in this paper the appropriate PHNC formulas are incorporated into the GGA approximation and the calculations have been performed for some metals: bulk as well as containing monovacancies. The comparison of the lifetimes found in this way to the ones based on the previous method is promising for the applicability of the new algorithm.

Key words: positron annihilation • electronic structure

Introduction

In studies of properties of several materials the measurement based on positron annihilation is one of widely applied techniques. However, the interpretation of the results of, e.g. angular correlation of annihilation quanta or Doppler broadening often requires some additional theoretical calculations describing the positron behaviour in the investigated sample. In general, this requires knowledge of the corresponding electronic structure and the proper description of the electron-positron (e-p) interaction. Since this many-body problem is highly complicated, several approximations should be introduced. One of the most common is the local density approximation (LDA) for the enhancement of the electron density on the positron due to e-p interaction. There are few formulas [2, 3, 12] based on many-body theories for the positron in jellium that briefly though approximately describe this enhancement. They work reasonably well for several materials and the corresponding positron lifetimes calculated for many pure and defected (vacancies) elements agree even very well with the data found experimentally. However, in many cases the complicated configuration of electronic states of the atomic core and the magnitude of the core density requires more advanced treatment of these states than the local approximation of these states by the electron gas. Then, the applied approximations are the GGA

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Received: 12 June 2009
Accepted: 8 October 2009

(generalized gradient approximation) [2], and the WDA (weighted density approximation) [6, 9]. The last one is probably the best [11], however, its complication makes that it cannot be used commonly. In this paper one advocates for the formulas based on the perturbed hypernetted-chain (PHNC) approach [4, 12] that can be introduced into the GGA to improve some of its deficiencies. In section 'Theory' the idea of the GGA is explained and the formulas that are used by the GGA are commented. Section 'Calculations and results' presents some results for the positron lifetime in several elements (for the positron in the bulk and trapped in a monovacancy) calculated within the GGA according to the new prescription. The comparison of the lifetimes to ones found within the popular version of the GGA, and additionally to LDA-based lifetimes, is done. The conclusions are presented in section 'Conclusions'.

Theory

In a homogeneous electron gas we have for the annihilation rate λ_0 (in units of 10^9/s) the following formula

$$\lambda_0 = 16\pi n\, g(r_s,0) \tag{1}$$

where $g(r_s,0)$ is a contact value of the e-p correlation function and r_s relates to the electron density n of the electron gas by $n = 4\pi r_s^3/3$. In the subsequent formulas we shall use the values of $g(r_s,0)$ corresponding to the three approaches belonging to: 1) Boroński and Nieminen [3], 2) Stachowiak *et al*. [10, 12], 3) Arponen and Pajanne [1, 2].

The most widely used is the formula of Boroński and Nieminen (BN) [3]

$$g_{\mathrm{BN}}(r_s,0) = 1 + 1.23r_s + 0.8295r_s^{3/2} - 1.26r_s^2 + 0.3286r_s^{5/2} + r_s^3/6 \tag{2}$$

There is also the formula presented by Barbiellini [2] interpolating the results of Arponen and Pajanne (AP) for the homogeneous electron gas [1]

$$g_{\mathrm{AP}}(r_s,0) = 1 + 1.23r_s - 0.0742r_s^2 + r_s^3/6 \tag{3}$$

On the basis of the PHNC approach [10] the following formula [12] have been presented

$$g_{\mathrm{PHNC}}(r_s,0) = 1 + 1.23r_s - 0.1375r_s^2 + r_s^3/6 \tag{4}$$

One should notice that among the mentioned only the PHNC approach yields correct momentum dependence on the e-p enhancement factors.

In a real material where are natural variations of the electron density from atom to atom we have for the annihilation rate λ the following expression within the LDA approximation

$$\lambda = 16\pi \int d\boldsymbol{r}\, n(\boldsymbol{r})\, g(r_s(\boldsymbol{r}),0)\, |\phi(\boldsymbol{r})|^2 \tag{5}$$

where $\phi(\boldsymbol{r})$ is the positron wave function and $n(\boldsymbol{r})$ is the local value of the electron density.

The comparison of lifetimes found with the above integral (5) to experimental data for many elements shows that the results of calculations are most close to the experiment if the BN formula (2) is used. The worst agreement gives the formula (3) of Arponen and Pajanne. For the elements with more complicated electronic configuration, e.g. where d- or f-electrons play an important role, the results of all of the approaches clearly differ from the experimental values.

In order to improve the agreement of theoretical calculations with the experimental data Barbiellini *et al*. [2], basing on earlier works of Perdew [7] on the electron systems, proposed the generalized gradient approximation (GGA) for the positron-electron problem. In their approach the local e-p enhancement factor depends both on the electron density and the absolute value of the local gradient of this density. It is made to obey the formula

$$g_{\mathrm{GGA}}(r_s(\boldsymbol{r}_p,0) = 1 + (g_{\mathrm{AP}}(r_s(\boldsymbol{r}_p,0) - 1)e^{-\alpha\varepsilon} \tag{6}$$

where

$$\varepsilon = \left| \frac{\nabla n(\boldsymbol{r})}{n(\boldsymbol{r}) q_{\mathrm{TF}}(\boldsymbol{r})} \right|^2 \tag{7}$$

where $q_{\mathrm{TF}}(\boldsymbol{r})^{-1}$ is the local Thomas-Fermi screening length and α is a phenomenological constant chosen equal to 0.22 in order to assure the best fit to the experimental positron lifetimes in metals.

Thus, the above expression for $g_{\mathrm{GGA}}(r_s)$ was used in (5) according to Ref. [2]. Barbiellini *et al*. [2] used the formula (3) in (6) because they wanted this to be consistent with correlation energy calculations of Arponen and Pajane [1]. These correlation energies are necessary for calculations of positron wave function $\phi(r)$ in a real metal.

In fact, this GGA correction slightly improved the desired agreement of lifetimes for some elements. However, as has been recently shown by Campillo *et al*. [5] there still remain divergencies, and especially for low electron density systems, the GGA based on the formula (3) does not give the reasonable lifetimes.

Our remedy to improve this defficiency of the GGA is to use the formula (4) instead of (3) within the GGA framework. We base on the fact that the formula (4) applied within the LDA gives better agreement with the experiment than the formula (3) used in the same type of approximation. Moreover, in 1998 Boroński and Stachowiak [4] presented results of calculations for e-p correlation energies in the electron gas based on the PHNC approach, thus consistent with the formula (4).

Then, our expression for the GGA reads

$$g_{\mathrm{GGA}}(r_s(\boldsymbol{r}_p,0) = 1 + (g_{\mathrm{PHNC}}(r_s(\boldsymbol{r}_p,0) - 1)e^{-\alpha\varepsilon} \tag{8}$$

where the new value for α is equal to 0.10. The corresponding formula for the correlation energy necessary for the construction of the e-p correlation potential is the following

$$E_{\mathrm{GGA}}^{\mathrm{corr}}(r_s) = E_{\mathrm{LDA}}^{\mathrm{corr}}(r_s)e^{-\alpha\varepsilon/3} \tag{9}$$

where $E_{\mathrm{LDA}}^{\mathrm{corr}}(r_s)$ bases now on the interpolations presented in [4].

Table 1. Positron lifetimes for several metals calculated in the LDA-BN, GGA-AP and GGA-PHNC approximations. Lifetimes for the positron in monovacancies in metals are shown at the right bottom of the table. Experimental data are taken from Ref. [5]

	Li	Na	K	Rb	Al	V	W	Fe	Cu	Ag	Au	Pb
Bulk												
Experiment	291	338	397	406	165	124	105	111	120	130	116	204
LDA-BN	301	331	377	386	166	105	121	104	112	130	116	223
GGA-AP	303	342	402	439	160	118	135	114	130	150	133	249
GGA-PHNC	294	338	393	406	163	109	138	112	118	136	120	228
Monovacancy												
Experiment					244	191	195	175	180	208	205	194
LDA-BN					252	197	217	181	185	226	217	328
GGA-AP					227	194	214	177	192	237	226	331
GGA-PHNC					230	186	205	176	169	202	192	317

Calculations and results

The calculations are preliminary and have been performed only for some elements from the periodic table, including, however, several metals of low and high electron density and *d*- and *f*-shells. Only fcc and bcc structures were considered (an easy calculation). We have assumed the value 0.1 for the parameter α in the formula (6). All electrons in the atomic cores were treated at the same footing as valence electrons as concerns the calculation of the e-p enhancement. We have not divided the electrons into any parts of different kind of localization. Only the total density at the given point was important for the calculation of the enhancement.

The electron density and the positron wave function were calculated with the ATSUP code [8]. The appropriate parts of the code, however, corresponding to the calculations of the correlation potential for the positron and for the e-p enhancement were changed according to (9), (4) and (8), respectively.

We accepted the following notation: The calculations performed within the LDA formula (2) will be labelled as LDA-BN. The calculations performed within the GGA formula (6) with (3) and (4) will be labelled as GGA-AP and GGA-PHNC, respectively.

The results are presented in Table 1. The positron lifetimes in monovacancies are also presented in this table. However, since the experimental data for the monovacancies in alkalis are not known, we have not calculated the corresponding lifetimes.

Conclusions

For most of considered metals, this new formula gives reasonable or even the best results. In particular, the lifetimes for the alkalis agree very well with the experimental data. For some metals with more complicated structure this approach or the conventional LDA Boroński-Nieminen formula assure the best agreement with the experiment. The GGA, as defined by Barbiellini works clearly worse. The situation is similar, as concerns the metals with monovacancies. From considered metals, only Pb cannot be described by any of the above simple approaches. The justification (and a recommendation) for the GGA-PHNC is that the PHNC lifetimes for the whole spectrum of electron densities are closer to BN values than the AP ones. This means that for the limiting case of slow variations of the electronic density, when this approximation works like the LDA, the results should be closer to the experimental data, as the BN results do. This is just the reason that for monovacancy lifetimes, LDA-BN is superior to GGA-AP. As is commonly known, the AP values are clearly too high when used within the LDA approximation. This is that only the fitting parameter alpha makes this approximation to work better for more inhomogeneous materials. This parameter is chosen that way (alpha = 0.22) that it fits most of the experiments best if the electronic structure is calculated within the LMTO-ASA self-consistent scheme. The reliable calculations have to take into account the transfer of the electronic charge due to a self-consistent potential; the electron density distribution yielded by ATSUP procedure is only approximate. Of course, the calculations using all the types of the GGA approximation will be more sensitive to this change in the electron density than the ones performed with the LDA. Therefore, the values of lifetimes calculated with the use of GGA-AP and ATSUP are worse than those calculated within the LMTO. Certainly, the same situation should occur in GGA-PHNC case. The new and the best parameter should be found when calculating, say, with the use of FLAPW (or LMTO) band structure method. Then, one can expect better agreement with the experiment. The results presented in this work are only preliminary, anyway, a step towards correcting the GGA has been done and the tests are promising.

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