CALCULATION OF POSITRON ANNIHILATION RATES IN METALS USING DIFFERENT ENHANCEMENT FACTORS

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Abstract

Positron annihilation rates in metals were calculated using the enhancement factors of Boronski and Nieminen (1986), Sterne and Kaiser (1991), and the local density approximation by solving the Kohn-Sham type of equation self-consistently for electron-positron densities in metals using an interactive scheme. The experimental and calculated positron annihilation rates in metals exhibit the same trend. The results obtained revealed that there is no significant difference in the positron annihilation rates calculated using the enhancement factors of Boronski and Nieminen (1986) and that of Sterne and Kaiser (1991). Furthermore, the results revealed that the positron annihilation rates calculated using the enhancement factor of the local density approximation were in better agreement with experimental values than the ones calculated using the enhancement factors of the two other models or approximations. This shows that the use of local density approximation is better in the prediction of positron annihilation rates in metals; and that the enhancement factor of the local density approximation explains electron-positron correlation in metals better than other models or approximations. Explanations for the observed discrepancies between the experimental and calculated positron annihilation rates in metals are given.

Keywords: *Metals, positrons, annihilation rates, enhancement factors*

1. Introduction

Positron annihilation spectroscopic studies are very important tools for studying electronic structure of solids, defects and defect properties (Rubaszek et al., 2001; Eldrup, 1995; Eldrup and Singh 1997). If a positron is injected into a metallic sample with energy of a few keV, it penetrates to the interior of the sample. There, it thermalises in about $3x10^{-12}$second via collisions with conduction electrons and will become part of the electronic system. Some time later, the positron will annihilate with an electron producing two 0.511MeV annihilation gamma photons. These relatively high-energy gamma photons provide information about the annihilation process in the sample with negligible attenuation or scattering. Positron annihilation studies in metals provide some information about the electronic structure of the metals (Rodda et al., 1963). In order to understand and interpret positron annihilation experiments properly, an understanding of the electron-positron interaction is very important (Sorman, 1996). The electron-positron interaction affects positron annihilation rates in metals. Positron annihilation rates in solids depend on the density of the electrons sampled by the positron (Boronski and Nieminen, 1986).

In calculating positron annihilation rates in metals, the enhancement factor plays an important role. The enhancement factor is defined as a ratio of the electron-positron annihilation rate to its independent particle model counterpart. The electron-positron enhancement factors together with charge distribution are two ingredients that are indispensable for the interpretation of the positron annihilation data (Rubaszek et al.,

2001). The enhancement factor takes into account the electron-positron interaction and it is a crucial ingredient when calculating positron annihilation rates (Barbiellini et al., 1997). The enhancement factor describes how the positron distorts the electron wave function. According to Nieminen (1983), there is no satisfactory general theory available for the enhancement factors, which would take into account the true nature of the electron wave functions. The enhancement factor affects positron annihilation rates as well as electron-positron correlation energy in metals. The enhancement factor is an electron-state-dependent function of the electron density and describes the local enhancement of the electron-positron interaction.

Calculation of positron annihilation rates in solids has being a subject of interest to many researchers. According to Ferrel (1958), positron annihilation rates with conduction electrons are better done on the basis of the Sommerfeld free electron theory. Positron annihilation rate is proportional to the electron density at the site of the positron. The Sommerfeld independent particle model gave annihilation rates that were not in good agreement with experimental values. The discrepancy between the independent particle annihilation rates and experimental values were due to the neglect of the strong electron-positron correlation that enhances the effective electron density at the site of the positron (Kahana, 1963). Also, the average electron density is used in calculating the positron annihilation rates rather than the actual density of electrons at the position of the positron. The actual electron density at the positron is much greater than the average because of the strong Coulomb attraction which the positron exerts on the electrons (Ferrell, 1958).

Brandt and Reinheimer (1971) gave an interpolation expression for calculating positron annihilation rates in solids based on the random phase approximation. The interpolation expression gave annihilation rates that were in agreement with experimental values for some metals. Boronski and Nieminen (1986) applied the two component density functional theory to calculated positron annihilation rates for positrons trapped in vacancies in Al, Cd, Mg, Hg, Li, and Na. The results they got were generally lower than experimental values. Sterne and Kaiser (1991) calculated positron lifetimes (inverse of annihilation rates) in some solids. The results obtained were in good agreement with experimental values for most of the metals they investigated. Puska (1991), performed *ab-initio* calculation of positron annihilation rates in solids. The results he got were in good agreement with experimental values.

In this work, positron annihilation rates in metals will be calculated using enhancement factors of Boronski and Nieminen (1986), Sterne and Kaiser (1991) and the enhancement factor of the local density approximation. The calculated positron annihilation rates in metals will be compared with experimental values. The variation of the calculated and experimental positron annihilation rates in metals with the electron gas parameter will be investigated.

2. Theoretical considerations and calculations

In the two-component density functional theory of Boronski and Niemani (1986) the ground state energy functional of a system of electrons and positrons in an external potential is given as

$$E[n_-, n_+] = F[n_-] + F[n_+] + \int dr V_{ext}(r)[n_-(r) - n_+(r)] - \int dr \int dr' \frac{n_-(r)n_+(r)}{|r-r'|} + E_c^{e-p}[n_-, n_+] \qquad (1)$$

where F[n-] and F[n+] are the respective one-component density functional for electrons and positrons, and F[n] is

$$F[n] = T[n] + \frac{1}{2}\int dr \int dr' \frac{n(r)n(r')}{|r-r'|} + E_{xc}[n] \quad (2)$$

T[n] is the kinetic energy of non-interacting electrons and positrons; $E_{xc}[n]$ is the exchange-correlation energy functional. E_c^{e-p} is the electron-positron pair correlation energy functional. The one-particle Schrodinger equation for electrons and positrons are

$$-\frac{1}{2}\nabla^2\psi_i^-(r) + \left[\frac{\delta E_{xc}[n_-]}{\delta n_-(r)} - \phi_c + \frac{\delta E_c^{e-p}[n_+, n_-]}{\delta n_-(r)}\right]\psi_i^-(r) = \varepsilon_i^-\psi_i^-(r) \quad (3)$$

$$-\frac{1}{2}\nabla^2\psi_i^+(r) + \left[\frac{\delta E_{xc}[n_+]}{\delta n_-(r)} - \phi_c + \frac{\delta E_c^{e-p}[n_+, n_-]}{\delta n_+(r)}\right]\psi_i^+(r) = \varepsilon_i^+\psi_i^+(r) \quad (4)$$

where ϕ_c is the Coulomb potential given as

$$\phi_c = \int dr' \frac{n_0(r') - n_-(r')^+ n_+(r')}{|r-r'|} \quad (5)$$

$n_0(r)$ is the positively charged background arising from the external potential. The electron and positron densities are obtained by summing over all the occupied states.

$$n_-(r) = \sum_{E_i \le E_F} |\psi_i^-(r)|^2, \quad n_+(r) = \sum_{i=1}^{N_0} |\psi_i^+|^2 \quad (6)$$

where $\psi_i^+(r)$ is the wave function of positrons, $\psi_i^-(r)$ is the wave function of the electrons, n_(r) is the density of the electrons, $n_+(r)$ is the density of the positrons. N_0 is the total number of electrons, E_f is the Fermi energy. Equations (1) to (6) are solved self-consistently for the electron and positron densities for different metals using an iterative scheme.

According to many body calculations, positron annihilation rates in solids is given as (Boronski and Nieminen, 1986, Eldrup, 1995.

$$\lambda = \pi r_0^2 c \int n_+(r) n_-(r) g(r_s, 0) dr \quad (7)$$

where r_0 is classical electron radius, c is the speed of light in vacuum, $n_+(r)$ is the positron density, n_(r) is the density of the electron and $g(r_s,0)$ is the enhancement factor. The enhancement factor describes the electron-positron interaction. Calculation of positron annihilation rates in metals using eqn. (7) is made possible with the aid of parameterized expression for calculating the enhancement factor. Arponen (1978) gave the expression for the enhancement factor as (eqn. 8).

$$g(r_s,0) = 1 + 1.23r_s + \text{higher order terms} \quad (8)$$

where r_s is electron gas parameter. The higher order terms contains several terms and a contribution coming from ring summation (Rubaszek et al., 2001). The enhancement factors are based on many body theories for calculating positron annihilation rates in solids. Boronski and Niemanien (1986) based on the many body calculations of Arponen and Pajanne (1979) gave the interpolation expression as (eqn. 9).

$$g_{BN}(r_s,0) = 1 + 1.23r_s + 0.8295r_s^{3/2} - 1.26r_s^2 + 0.3286r_s^{5/2} + \frac{r_s^3}{6} \quad (9)$$

Sterne and Kaiser (1991) gave the interpolation expression for calculating enhancement factor as

$$g_{SK}(r_s,0) = 1 + 0.1512r_s + 2.414r_s^{3/2} - 2.01r_s^2 + 0.4466r_s^{5/2} + 0.1667r_s^3 \quad (10)$$

In the local density approximation the enhancement factor is given by the interpolation expression (LiMing et al., 1997) as

$$g_{LDA}(r_s,0) = 1 + 1.23r_s - 0.0742r_s^2 + \frac{r_s^3}{6} \quad (11)$$

In this work, the enhancement factor will be calculated using the interpolation expressions given in eqns. (9), (10) and (11) while positron annihilation rates in metals will be calculated using eqn. (7). The calculated positron annihilation rates will be compared with experimental values.

3. Results and discussion

The variation of the enhancement factor of Boronski and Niemine (NB), Sterne and Kaiser (SK) and the local density approximation (LDA) with electron gas parameter is shown in Fig. 1. Figure 1 reveals that the enhancement factors calculated using the above-mentioned models or approximations increases exponentially with increase in the electron gas parameter. The enhancement factor of Boronski and Niemanien (1986) has the highest value while the enhancement factors of Sterne and Kaiser (1991) and the enhancement factor of the local density approximation has nearly the same value for each of the metals. Figure 1 suggests that electron-positron correlation in metals is low for metals in the high-density region and high for metals in the low-density region. This may be due to the free electrons present in the metals in the low-density region.

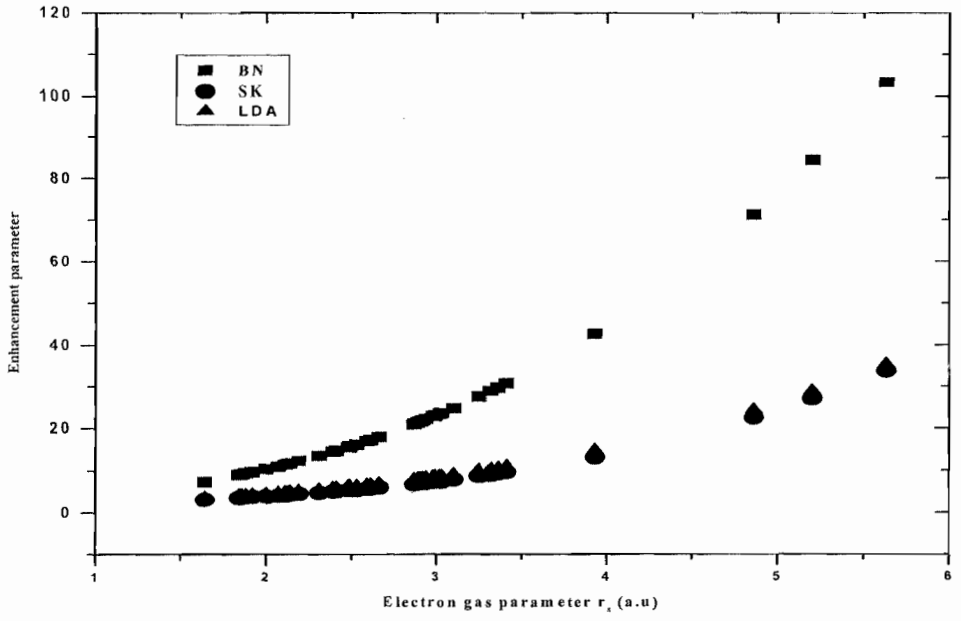

Fig. 1: Variation of calculated parameterized enhancement factors due to Boronski and Nieminen (BN), Sterne and Kaiser (SK) and local density approximation (LDA) with electron gas parameter.

Figure 2 shows the variation of calculated and experimental positron annihilation rates with the electron gas parameter for different metals. As shown in the Fig. 2 and Table 1, there is no much difference in the annihilation rates calculated using the parameterized enhancement factor due to Boronski and Nieminen, (1986) and the enhancement factor of Sterne and Kaiser, (1991) for all the metals investigated. Although, the enhancement factor of Sterne and Kaiser (1991) gave annihilation rates that are slightly

higher than the ones obtained using the enhancement factor of Boronski and Nieminen (1986). The annihilation rates calculated using the above enhancement factors agreed with experimental values for few elements in the high-density region ($r_s \leq$ 3a.u). In the low-density region, ($4 \leq r_s \leq 3$), the annihilation rates calculated with the above enhancement factors are close to but generally lower than the experimental values. The discrepancy between the experimental annihilation rates and the ones calculated using the enhancement rates of Boronski and Nieminen (1986) was because the enhancement factor was based on the calculation of Arponen and Pajanne (1979). The enhancement factor of Boronski and Nieminen (1986) over estimated the electron-positron correlation of metals. In the calculation of Arponen and Pajanne (1979), the inhomogeneities of the lattice and electron-positron correlation were neglected (Osiele, 2001). Also, the discrepancy between the observed annihilation rates and the ones calculated using the enhancement factor of Sterne and Kaiser (1991) was due to the inexact account of electron-electron correlation.

Table1: Positron annihilation rates in metals calculated using the three enhancement factors and experimental values. The experimental values were taken from Welch and Lynn, (1976). BN is annihilation rates in metals calculated using the parameterized enhancement factor of Boronski and Nieminen, (1986); SK is positron annihilation rates in metals calculated using the parameterized enhancement factor of Sterne and Kaiser (1991), LDA is the positron annihilation rates in metals calculated using the parameterized enhancement factor of the local density approximation.

Metal	r_s (a.u)	Annihilation rates ($\times 10^9 s^{-1}$)			
		BN	SK	LDA	EXPT
Li	3.25	2.98	3.01	3.47	3.38
Na	3.93	2.57	2.60	2.93	2.94
K	4.86	2.34	2.35	2.55	2.51
Rb	5.20	2.29	2.31	2.50	2.41
Cs	5.63	2.25	2.27	2.37	2.39
Be	1.88	6.68	6.64	7.51	6.67
Mg	2.66	3.75	3.79	4.39	4.42
Ag	2.39	4.38	4.41	5.09	7.52
Au	2.39	4.38	4.41	5.05	9.09
Cd	2.59	3.89	3.93	4.55	5.38
Al	2.07	5.58	5.58	6.37	6.13
Ga	2.19	5.05	5.07	5.81	5.10
In	2.41	4.33	4.36	5.03	5.04
Sn	3.02	3.21	3.25	3.76	4.99
Pb	2.90	3.36	3.40	3.94	4.57
Sb	2.53	4.02	4.06	4.69	3.78
Y	2.61	3.85	3.89	4.50	4.01
Bi	2.49	4.12	4.15	4.80	3.98
Sc	3.32	2.92	2.96	3.40	4.35
Ti	1.92	6.41	6.38	7.24	6.80
V	1.64	8.81	8.67	9.67	.....
Cr	1.86	6.52	6.77	7.65	.....
Mn	2.14	5.26	5.27	6.03	.....
Fe	1.85	6.89	6.84	7.73	9.43
Co	2.07	5.58	5.58	6.37	8.45
Ni	2.07	5.58	5.58	6.37	9.56
Cu	2.12	5.34	5.35	6.12	8.20
Zn	2.31	4.62	4.65	5.35	5.63
Zr	2.11	5.39	5.39	6.17	6.13
Nb	2.13	5.30	5.31	6.08	.....
Mo	1.84	6.96	6.91	7.80	8.20
Ag	2.89	3.38	3.42	3.96	7.52
La	3.10	3.12	3.16	3.65	4.03
Ta	2.48	4.14	4.18	4.43	8.93
Pt	2.00	5.94	5.92	6.74	8.62
Au	2.39	4.38	4.41	5.09	9.09
Hg	3.36	2.89	2.93	3.39	5.17
La	3.10	3.12	3.16	3.65	4.03
Ce	3.03	3.20	3.24	3.75	4.17
Pr	3.02	3.21	3.25	3.76	4.27
Nd	3.02	3.21	3.25	3.76	4.27
Sm	2.98	3.26	3.30	3.82	4.13
Eu	3.41	2.85	2.89	3.31	3.57
Gd	2.99	3.25	3.29	3.80	4.08
Tb	2.92	3.34	3.38	3.91	4.10

Dy	2.94	3.34	3.38	3.91	4.24
Er	2.91	3.35	3.40	3.92	4.20
Tm	2.89	3.38	3.42	3.96	4.13
Yb	2.99	3.25	3.29	3.80	3.75
Lu	2.87	3.41	3.45	3.99	4.11

From Fig. 2 and Table 1, positron annihilation rates calculated using the enhancement factor of the local density approximation are in good agreement with experimental values for some metals like Mg, Zr, and Yb. In the low-density region, positron annihilation rates calculated using the enhancement factor of the local density approximation is in very satisfactory agreement with experimental values. The success of the local density approximation can be attributed to the fact that in the local density approximation, correlation effect and crystal structure were put into consideration (Barbiellini et al., 1996). The success of enhancement factor according to the local density approximation in calculating positron annihilation rates in metals supports it success in calculating other metallic properties (Puska, 1991).

As shown in Fig. 2, experimental annihilation rates do not follow a regular pattern in the high-density region, but in the low-density region, it does. While calculated positron annihilation rates follow a regular pattern in terms of the electron gas parameter in all the density regions. This is why in the low-density limit; theoretical calculations were in good agreement with experimental values. Also, the basis of the models or approximations that produced the above enhancement factors was the homogeneous electron gas model of solids which can be used to explain the properties and behaviour of simple metals found in the low-density region. The discrepancies between the calculated positron annihilation rates in metals and experimental values may be due to the experimental technique used to determine positron annihilation rates in metals such as sample handling technique, the vacuum condition, the type of detector used and method of data analysis.

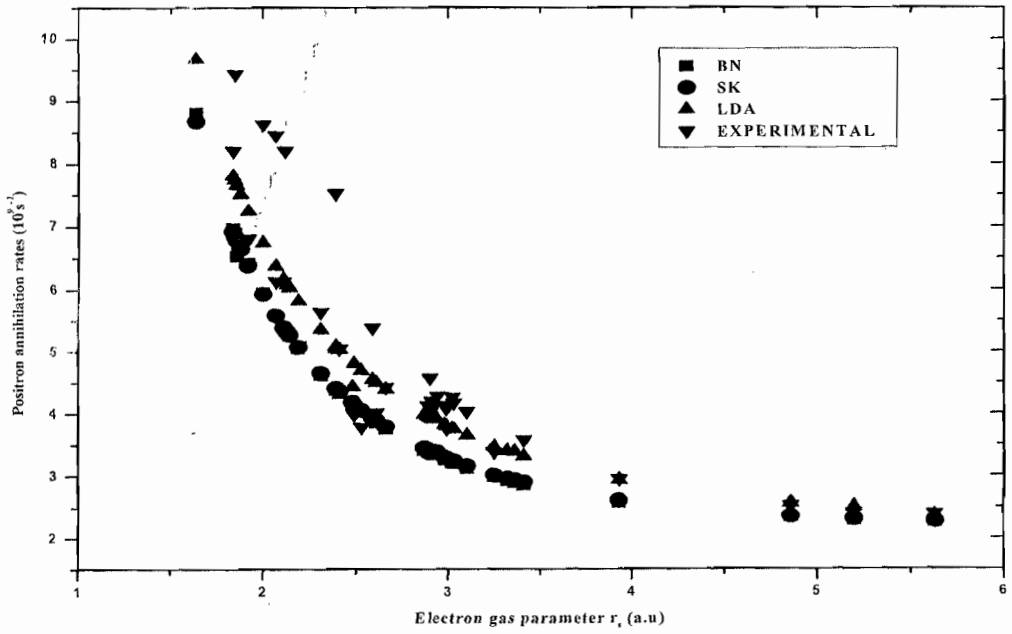

Fig 2: Variation of positron annihilation rates calculated using the parameterized enhancement factors of Boronski and Nieminen (BN), Sterne and Kaiser (SK) and local density approximation (LDA) with electron gas parameter.

4. Conclusion

In this work, positron annihilation rates in metals were calculated using three different enhancement factors. There are no significant difference in the enhancement factors calculated using the parameterized expression of the local density approximation and that of Sterne and Kaiser (1991). Positron annihilation rates in metals calculated using the enhancement factors of Boronski and Nieminen (1986) and that of Sterner and Kaiser (1991) do not vary significantly and they are close to the experimental values only at the low-density region. Positron annihilation rates calculated using the parameterized enhancement factor of the local density approximation is in very good agreement with experimental values mostly in the low-density region. This shows that the local density enhancement factor can be used to predict positron annihilation in metals and it treats electron-positron correlation effects better than the other two models or approximation. The enhancement factor according to local density approximation produced better annihilation rates because in the local density approximation, correlation effect and the inhomogeneity of the real crystal lattice were put into consideration.

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