Self-consistent field method and non-self-consistent field method for calculating the positron lifetime*

Zhang Jie(张 杰)†, Liu Jian-Dang(刘建党), Chen Xiang-Lei(陈祥磊), and Ye Bang-Jiao(叶邦角)

Department of Modern Physics, University of Science and Technology of China, Hefei 230026, *China*

(Received 24 January 2010; revised manuscript received 11 May 2010)

Many methods are used to calculate the positron lifetime, these methods could be divided into two main types. The first method is atomic superposition approximation method and the second one is the so called energy band calculation method. They are also known as the non-self-consistent field method and self-consistent field method respectively. In this paper, we first introduce the two basic methods and then, we take Si as an example and give our calculation results, these results coincide with our latest experimental results, finally, we discuss the advantages and disadvantages of the two methods.

Keywords: positron lifetime, atomic superposition approzimetion method, pseudopotential method

PACC: 7870B

1. Introduction

Positron was rapidly applied to material science since it was discovered in the 1930s, and positron annihilation in physics is called positron annihilation spectroscopy (PAS). PAS includes a variety of experimental techniques and it is an important method for probing the electron and atomic structure of solids.[1−4] As in the case of other methods, the theory underlying positron annihilation has developed from some models describing the positron–solid interaction to 'first principles' methods predicting the annihilation characteristics for different environments and conditions.[5] The description of the state of a thermalized positron in a crystal requires the solution of the Schrödinger equation, from this point of view, it is very important to obtain the potential of the positron.

Different approaches dealing with the potential of positron form the so called different calculation methods. As mentioned above, for calculating positron lifetime, there are two main ways. The first one is the atomic superposition approximation method (ATSUP).[6−8] Essentially, it is a non-self-consistent process, so we can also call it non-self-consistent method (NSCF). The second one is energy band calculation method.[5] Because it is a self-consistent process, we can call it self-consistent method (SCF). There are many methods in the internation at present, such as the linear muffintin orbital (LMTO) method,[9−12] full-potential linearised augmented plane-wave (FLAPW) method,[13] pseudopotential method[14,15] and so on. In fact, in theory, all the methods which could be used to calculate the electron structure of condensation can also be used to calculate the positron lifetime.

In our nation, the ATSUP method has been developed several years ago,[16,17] so, here we only introduce the method briefly. In this paper, we give the details of the pseudopotential method since it is the first time in our nation to use this approach to calculate the positron lifetime. As an example, here we only give the calculation results of silicon, these results coincide with our latest experimental results. Because the calculation process of positron defect lifetime is similar as the process for calculating positron bulk lifetime, here we only give the positron bulk lifetime of silicon (of course, we have used the pseudopotential method to calculate positron lifetime of many materials, these results will be given in later papers).

2. Calculation methods

The positron state of a thermalized positron in a crystal requires the solution of the following Schrodinger equation:[18,19]

$$\left[-\frac{1}{2}\nabla^2 - V_{\rm H}(r) + V_{\rm nuc}(r) + u(n_{\rm e})\right]\psi_+(r) = \varepsilon_+\psi_+(r), \quad (1)$$

*Project supported by the National Natural Science Foundation of China (Grant No. 10675115) and the Important Direction Project of the Chinese Academy of Sciences.

†Corresponding author. E-mail: zjszm@mail.ustc.edu.cn

© 2010 Chinese Physical Society and IOP Publishing Ltd

where $V_{\rm H}(r)$ is the Hartree potential of positron, $V_{\rm nuc}(r)$ is the nucleus potential, $u(n_{\rm e})$ is the positron–electron exchange–correlation potential. There are three frames to deal with the exchange–correlation potential at present—local density approximation (LDA),[20] weighted density approximation (WDA),[21,22] generalised gradient approximation (GGA).[23] In this study the LDA scheme is adopted to calculate the exchange–correlation potential. The other potential could be calculated in the following two methods:

2.1. ATSUP method (NSCF method)

In this method, the electron density and the crystalline Coulomb potential are constructed in a non-self-consistent process:

$$n_{-}(r) = \sum_i n_{-}^{\rm at}(r - R_i), \tag{2}$$

where $n_{-}^{\rm at}$ is the free-atom electron density, R_i is the occupied atomic site. The crystal Coulomb potential $V_{\rm C}(r)$ is expressed as:

$$V_{\rm C}(r) = \sum_i V^{\rm at}(r - R_i). \tag{3}$$

2.2. Pseudopotential method (belongs to SCF method)

In this method, only the outer electrons are considered, the inner electrons together with the nucleus are considered as atomic core (From this point of view, this method only considers the positron annihilation with valence electron but ignores the positron annihilation with inner electrons. For most solids, this is right,[24] meanwhile, we can predict that the calculation results of positron annihilation rate are a little small and so the positron lifetime is a little bigger than the experimental one). The valence electron density is obtained from a self-consistent process. After the electron density is obtained, the total potential of the positron $V_t(r)$ can be obtained:

$$\begin{aligned} V_{\rm t}(r) &= -V_{\rm H}(r) + V_{\rm nuc}(r) + u(n_{\rm e}) \\ &= \sum_n \sum_i v(r - \mathbb{R}_n - T_i) - \int \frac{\rho_{\rm e}(r')}{|r - r'|} \cdot {\rm d}^3 r \\ &\quad + u_{\rm LDA}(n_{\rm e}), \end{aligned} \tag{4}$$

where $\mathbb{R}_n$ denotes the set of all Bravais lattice vectors and T_i is a non-primitive vector of a two-atom basis. $v(r) = Ze^2/r$, here Z is the charge number of the cation.

From the above two methods, the potential of the positron can be obtained, and then the Schrödinger equation can be solved. After we get the positron wave function, the positron density distribution could be calculated through the following equation:

$$n_{\rm p}(r) = \sum_i^{N_{\rm p}} |\psi_i^{\rm p}(r)|^2, \tag{5}$$

where $N_{\rm p}$ is the number of positrons, $\psi_i^{\rm p}(r)$ is the wave function of positrons. When positron density is known, then the positron lifetime could be obtained.[5]

3. Calculation results

The single crystal silicon has the diamond structure, its crystal structure is given in Fig. 1.

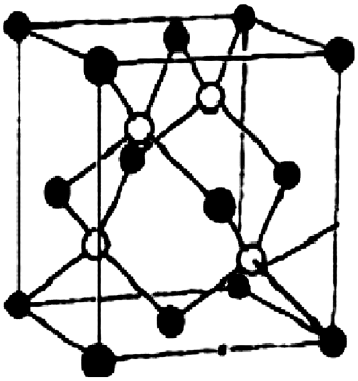


Fig. 1. Unit cell of diamond structure.

Some parameters used in our calculation are given in Table 1. a_0 is the lattice constant of silicon and ε_∞ is the high frequency dielectric constant.

Table 1. Lattice constant and high frequency dielectric constant of silicon.

crystal	a_0/Å	ε_∞
Si	5.43088[25]	12.0[26]

In practical calculation, we first divide the unit cell into $50 \times 50 \times 50$ grid points. Although the calculation results are good, the calculation time is long. Thus we divide the unit cell into $24 \times 24 \times 24$ grid points, the calculation results are also fine, and the time is significantly reduced. The calculation results of positron bulk lifetime are shown in Tables 2 and 3.

Table 2. The calculated positron bulk lifetime of silicon (the grid points: $50 \times 50 \times 50$).

crystal	ATSUP (NSCF)	pseudopotential (SCF)	our latest experimental result
Si	219ps	221ps	220ps

Table 3. The calculated positron bulk lifetime of silicon (the grid points: $24 \times 24 \times 24$).

crystal	ATSUP (NSCF)	pseudopotential (SCF)	our latest experimental result
Si	217ps	226ps	220ps

Form Tables 1 and 2, we can see that the calculation results within two schemes are coincide with the experimental ones. Figure 2 gives the positron density distribution within the pseudopotential frame. The positron wavefunction has already normalized.

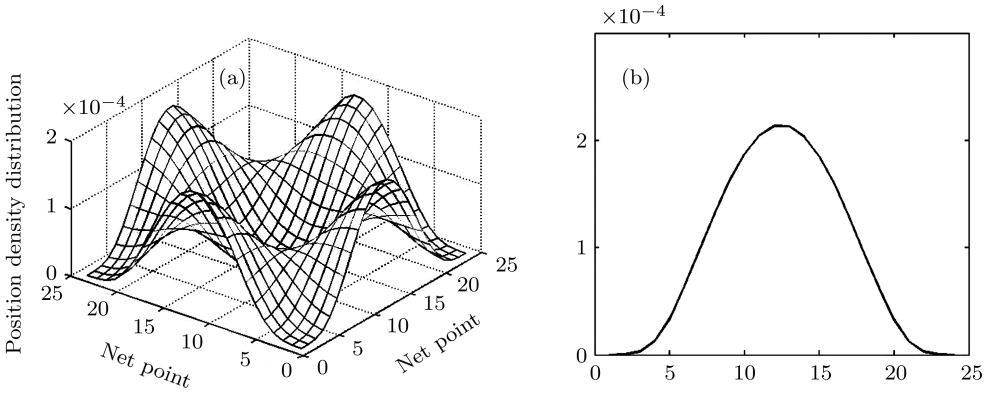


Fig. 2. Positron density distribution. (a) The positron density distribution in the (001) atomic plane; (b) The positron density distribution in the ⟨001⟩ direction.

4. Discussion

From our practical calculation, we find the calculation results of the pseudopotential method are a little larger than the experimental ones (see Tables 2 and 3), this is in our expectation. As mentioned above this method ignores the positron annihilation with the core electrons. As a result, the calculation of the positron annihilation rate is a little small and the positron lifetime is a little bigger than the experiment ones. In a word, from these results, we can see that the two methods for calculating positron bulk are powerful and we can also use these methods to predict the positron bulk lifetime. Besides, in order to obtain a good calculation result and reduce the calculation time, in general, we can divide the unit cell into $35 \times 35 \times 35$ grid points.

5. Conclusion

Two methods for positron calculation are introduced, the first one is a non-self-consistent field method and the second one is a self-consistent field method. In this study, the pseudopotential method is used. We take silicon as an example and give the calculation results, which coincide well with our latest experimental ones. For general solids, it is good enough to divide the unit cell into $35 \times 35 \times 35$ grid points. In this circumstance, a good calculation result can be obtained and the calculation time is short.

References

[1] Dannefaer S, Mascher P and Kerr D 1986 *Phys. Rev. Lett.* **56** 2195

[2] Saarinen K, Laine T, Kuisma S, Nissila J, Hautojarvi P, Dobrzynski L, Baranowski J M, Pakula K, Stepniewski R, Wojdak M, Wysmolek A, Suski T, Leszczynski M, Grze-

gory I and Porowski S 1997 *Phys. Rev. Lett* **79** 3030
[3] Gebauer J, Krause R R, Domke C, Ebert P and Urban K 1997 *Phys. Rev. Lett.* **78** 3334
[4] Krause R, Saarinen K, Hautojarvi P, Polity A, Gartner G and Corbel C 1990 *Phys. Rev. Lett.* **65** 3329
[5] Puska M J and Nieminen R M 1994 *Rev. Mod. Phys.* **66** 841
[6] Puska M J and Nieminen R M 1983 *J. Phys.* F: *Met. Phys.* **13** 333
[7] Campillo J M, Ogando E and Plazaola F 2007 *J. Phys.*: *Condens. Matter* **19** 176222
[8] Makkonen I, Hakala M and Puska M J 2006 *Phys. Rev.* B **73** 035103
[9] Alatalo M, Barbiellini B, Hakala M, Kauppinen H, Korhonen T, Puska M J, Saarinen K, Hautojarvi P and Nieminen R M 1996 *Phys. Rev.* B **54** 2397
[10] Puska M J, Makinen S, Manninen M and Nieminen R M 1989 *Phys. Rev.* B **39** 7666
[11] Boev O V, Puska M J and Nieminen R M 1987 *Phys. Rev.* B **36** 7786
[12] Ghosh V J, Alatalo M, Asoka K P, Nielsen B, Lynn K G, Kruseman A C and Mijnarends P E 2000 *Phys. Rev.* B **61** 10092
[13] Kontrym S G, Samsel C M, Biasini M and Kubo Y 2004 *Phys. Rev.* B **70** 125103
[14] Benosman N, Amrane N, Mecabih S and Aourag H 2000 *Mater. Chem. Phys.* **61** 1727
[15] Amrane N 2009 *Mater. Chem. Phys.* **114** 283
[16] Chen X L, Xi C Y, Ye B J and Weng H M 2007 *Acta Phys. Sin.* **56** 6695 (in Chinese)
[17] Chen X L, Kong W, Weng H M and Ye B J 2008 *Acta Phys. Sin.* **57** 3271 (in Chinese)
[18] Tang Z, Hasegawa M, Nagai Y, Saito M and Kawazoe Y 2002 *Phys. Rev.* B **65** 045108
[19] Sterne P A and Kaiser J H 1991 *Phys. Rev.* B **43** 13892
[20] Daniuk S, Sob M and Rubaszek A 1991 *Phys. Rev.* B **43** 2580
[21] Rubaszek A, Szotek Z and Temmerman W M 1998 *Phys. Rev.* B **58** 11285
[22] Rubaszek A, Szotek Z and Temmerman W M 2000 *Phys. Rev.* B **61** 10100
[23] Barbiellini B, Puska M J, Torsti T and Nieminen R M 1995 *Phys. Rev.* B **11** 7341
[24] Puska M J and Corbel C 1988 *Phys. Rev.* B **38** 9874
[25] Hubbard C R, Swanson H E and Mauer F A 1975 *J. Appl. Crystallogr.* **8** 45
[26] Puska M J, Makinen S, Manninen M and Nieminen R M 1989 *Phys. Rev.* B **39** 7666