The Positronium state in quartz

B. Barbiellini\textsuperscript{1} and P.M. Platzman\textsuperscript{2}

\textsuperscript{1} Department of Physics, Northeastern University, Boston, Massachusetts 02115, USA
\textsuperscript{2} Bell Laboratories, Alcatel-Lucent, 600 Mountain Avenue, Murray Hill, New Jersey 07974, USA

The positronium state in quartz is described by a linear superposition of two states: the first describing the free positron in the crystal and the second corresponding to a positronium Bloch wavefunction in the lattice. The condition for positronium formation in the electron gas is deduced by using variational calculations of the positron binding energy to the electron system. The self annihilation parameter $\kappa$ introduced in positron lifetime experiments can be properly justified by using the mixed state. A variational method to calculate $\kappa$ is proposed.

\textbf{PACS numbers:} 36.10.Dr, 71.60.+z, 78.70.Bj, 82.30.Gg

Positronium (Ps), first observed in 1951 \cite{1}, is a unique probe atom to test bound state QED \cite{2}. Ps is formed in two spin states, 75% as ortho-Ps with parallel spins (triplet state) and 25% as para-Ps with antiparallel spins (singlet state). The energy difference between these spin states (hyperfine splitting) is about 8.4 \times 10^{-4} \text{ eV}. Para-Ps annihilates in vacuum mainly into photons of 511 keV. However, ortho-Ps annihilates in vacuum into three photons in order to conserve the spin. The calculated lifetime in vacuum for para-Ps is $\tau_p = 124$ ps while the corresponding value for ortho-Ps is $\tau_o = 142$ ns, which is in good agreement with recent experimental results \cite{3}.

Near surfaces the positron wave function overlaps with electrons outside the Ps. Therefore, the annihilation with electrons having an antiparallel spin decreases the ortho-Ps lifetime. This process is called pick-off annihilation and results in two photons. For instance, a quartz surface binds a ortho-Ps atom with an attractive van der Waals interaction \cite{4} and the lifetime is then reduced to about 10 ns \cite{5}.

Quartz is also one of the the rare materials, where Ps is delocalized in a Bloch state leading to peaks with negligible widths at reciprocal lattice vectors in the angular correlation of annihilation radiation of the positron annihilation (ACAR) spectrum. Brandt et al \cite{6} by studying the ACAR in quartz saw a spectacular fine structure. Greenberger et al. \cite{7} demonstrated that this structure was the manifestation of Ps formation in the form of a Bloch state. With the help of high resolution two dimensional ACAR \cite{8} it has been possible to clearly extract the square modulus of the Ps Bloch function at the $\Gamma$ state in momentum space.

Moreover, Saito and Hyodo \cite{9} showed the existence of two well defined spin states ortho- and para-Ps by measuring two different lifetime components and by introducing a self annihilation parameter $\kappa$. Inside the quartz crystal, the ortho-Ps lifetime becomes as low as 270 ps \cite{10} and the quantum mechanical interpretation of the Ps state inside the material becomes a controversial topic \cite{11}. In fact, in some alkali metals the positron lifetime is above 400 ps (i.e. much larger than the ortho-Ps lifetime in quartz) and the positron-electron pair correlation function is very similar to the case of Ps in vacuum. However, one does not observe any signatures of real Ps formation in alkali metals such as singlet-triplet lifetime splittings or Ps spikes in the ACAR spectra or 3-photon annihilation \cite{11}.

In the quartz crystal, the fine structure in momentum space also suggests a certain decoupling between the Ps and the electronic gas surrounding it. However, how could this condition be met inside the quartz crystal where the Ps sees a considerable amount of electron density? Moreover, how could the self annihilation parameter $\kappa$ be different from zero without violating the quantum mechanical principle of identical particles?

In this paper, we solve the problem of the quantum mechanical interpretation of the Ps state in a solid by using a mixing of the state $\Psi_1$ representing the positron freely moving in an electron system and the state $\Psi_2$ of a Ps attached to this system. This new state is characterized by a mixing angle $\theta$, which can be calculated variationally. An interesting wavefunction ansatz $\Psi_2$ for the Ps bound state to metals has been given by Bergersen \cite{12}. Unfortunately, the annihilation rates computed from this ansatz are much lower than the corresponding experimental values. However, we will see that $\Psi_2$ may become stable when the electron density is less than the typical metallic values.

We will start by studying the problem of a positron in a homogeneous electron gas. This system is completely characterized by the electron gas parameter

$$ r_s = \left( \frac{3}{4\pi n} \right)^{1/3}, $$

where $n$ is the electron density. For instance, the Fermi momentum in atomic units is $p_F = 1.92/r_s$ and the Thomas Fermi screening wave number is $q_{TF} = 1.56/\sqrt{r_s}$ \cite{13}. Variational Quantum Monte Carlo (QMC) calculations can be performed by using simulation cells containing $N = 226$ electrons and one positron \cite{14, 15}. The variational wave function in the standard Jastrow-Slater form is given by

$$ \Psi_1 = D_\uparrow D_\downarrow J \varphi_+, $$

where $D_\uparrow$ is a Slater determinant formed by plane waves for spin up electrons, $D_\downarrow$ is a similar Slater determinant.
for spin down electrons, $\varphi_+$ is the positron wave function and $J$ is a Jastrow factor used by Boronski [14]. The wave function $\varphi_+$ is constant since the positron is in the lowest energy state. The Jastrow factor can be factorized as

$$J = J_{ee} J_{ep} J_{ee},$$

where $J_{ee}, J_{ep}$ and $J_{ee}$ contain the electron-electron, electron-positron and electron-electron-positron correlations respectively. The factor $J$ has 12 variational parameters, which have been optimized using the stochastic gradient approximation (SGA) [15, 16]. The positron-electron binding energy $E_B$ shown in Fig. 1 is the negative of the electron-positron correlation energy given by

$$E_c = \langle \Psi_1 | H | \Psi_1 \rangle - \langle \Psi_0 | H | \Psi_0 \rangle,$$

where $H$ is the Hamiltonian of the system and $\Psi_0$ is the state of the system non interacting with the positron. Boronski [14] by performing variational QMC calculations for $r_s$ ranging from 2 to 10 has observed an anomalous behavior of the annihilation rates at $r_s = 6$. He suggested that this effect is an indication of Ps formation.

To check this hypothesis, we have considered the trial wavefunction proposed by Bergersen [12]

$$\Psi_2 = A [ \Psi_{N-1} \phi],$$

where $A$ is the antisymmetrization operator enforcing the Pauli principle, $\Psi_{N-1}$ is the wavefunction of the system without an electron and $\phi(r_1, r_j)$ is the Ps wavefunction containing variational parameters. Similar wavefunctions have been used to describe excitons in solids [17]. Unlike the wavefunction $\Psi_1$ used by Boronski, Bergersen’s original ansatz neglects the role of the electron-electron correlation in the calculation of binding energy $E_B$. This energy can parametrized by

$$E_B = E_{PS} \frac{r_s^\eta}{r_s^\eta + 12.5},$$

where $E_{PS}$ is the Ps binding energy in vacuum and $\eta = 2.2$. We have verified as shown in Fig. 1 that for $r_s > 6$ the positron binding energy $E_B$ given by Bergersen is larger than $E_B$ obtained from QMC calculations using $\Psi_1$ [14]. Thus, this observation indicates that Ps becomes stable for $r_s > 6$. Nevertheless, a more accurate study introducing a Jastrow factor in Bergersen’s ansatz may change the critical value of $r_s$.

Next, we study the possibility in quartz to have a mixing

$$| \Psi_3 > = \cos(\theta) | \tilde{\Psi}_1 > + \sin(\theta) | \tilde{\Psi}_2 >.$$

The state $\tilde{\Psi}_1$ is the same as $\Psi_1$ except that the Kohn-Sham orbitals for quartz replace the planes waves of the homogeneous electron gas in the Slater determinants. Moreover, the corresponding positron state $\varphi_+$ is given by a periodic function with an energy $I$ with respect to the electron conduction band. A value $I = 1.7$ eV is obtained using the positron and electron chemical potentials from density functional theory [18] and the energy gap of $E_G = 8.9$ eV. Likewise, the state $\tilde{\Psi}_2$ is the same as $\Psi_2$ except that the corresponding $\Psi_{N-1}$ is the quartz groundstate wavefunction and the corresponding $\phi$ is a Ps Bloch wavefunction. The variational parameters of $\Psi_3$ can be optimized by using the SGA [15, 16]. The mixing angle $\theta$ and the self-annihilation parameter $\kappa$ [9] are related via the formula

$$\kappa = \sin^2(\theta).$$

The angle $\theta$ or the parameter $\kappa$ can also be calculated with the SGA. This method can be used to verify the experimental value $\kappa = 0.34$ [9] and the Ps binding energy 4.8 eV for quartz [19].

In conclusion, we have presented a state representation of a Ps entangled with the electronic structure of quartz. This model is suitable to explain the fine structures observed in the momentum density distribution and the singlet-triplet positron lifetime splitting.

We thank R. Saniz and A.P. Mills for useful discussions. This work was supported by the US Department of Energy contract DE-FG02-07ER46352 and benefited from the allocation of computer time at the NERSC and the Northeastern University’s Advanced Scientific Computation Center (NU-ASCC).
[1] M. Deutsch, Phys. Rev. 82, 455 (1951).
[2] S. G. Karshenboim, Int. J. Mod. Phys. A 19, 3879 (2004).
[3] A. Badertscher, P. Crivelli, W. Fetscher, U. Gendotti, S. Gninenko, V. Postoev, A. Rubbia, V. Samoylenko, D. Sillou, Phys. Rev. D 75, 032004 (2007).
[4] R. Saniz, B. Barbiellini, A. Freeman and P.M. Platzman, Phys. Rev. Lett. 99, 096101 (2007); Phys. Rev. Lett. 100, 019902 (2008).
[5] D.B. Cassidy and A.P. Mills Jr, Nature 449, 195 (2007).
[6] W. Brandt, G. Coussot, and R. Paulin, Phys. Rev. Lett. 23, 522 (1969).
[7] A Greenberger, A.P. Mills, A. Thompson, and S. Berko, Phys. Lett. 32A, 72 (1970).
[8] P.E. Bisson, P. Descouts, A. Dupanloup, A.A. Manuel, E. Perreard, M. Peter and R. Sachot, Helv. Phys. Acta 55, 110 (1982).
[9] H. Saito and T. Hyodo, Phys. Rev. Lett. 90, 193401 (2003).
[10] A. Dupasquier, P. De Natale and A. Rolando, Phys Rev B 43, 10036 (1991).
[11] V. L. Telegdi, J. C. Sens, D. D. Yovanovitch and S. D. Warshaw, Phys. Rev. 104, 867 (1956).
[12] B. Bergeren, Phys. Rev. 181, 499 (1969).
[13] B. Barbiellini, Phys. Lett. 134A, 330 (1989).
[14] E. Boronski, Europhys. Lett. 75, 475 (2006).
[15] A. Harju, B. Barbiellini, S. Siljamaki, R.M. Nieminen and G. Ortiz, Phys. Rev. Lett. 79 (1997) 1173 (1997).
[16] D. Nissenbaum, Ph. D. thesis, Northeastern University (2008), [http://hdl.handle.net/2047/d10016466]
[17] B. Barbiellini in New Directions in Antimatter Chemistry and Physics, C. M. Surko and F. A. Gianturco, eds., Kluwer Academic Publishers, The Netherlands, (2001).
[18] Y. Nagashima, Y. Morinaka, T. Kurihara, Y. Nagai, T. Hyodo, T. Shidara, and K. Nakahara, Phys. Rev. B 58, 12 676 (1998).