

# Path Integral Monte Carlo Simulations of Positronium in Argon

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## Introduction

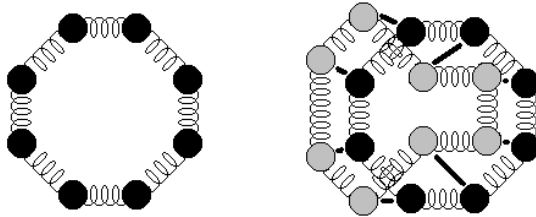
Positrons are a useful and non-destructive tool for probing materials; their current uses range from detecting changes in weapons materials [1] to determining whether a diamond is natural or synthetic [2]. Because the positron is the antiparticle of the electron, there are no positrons present in ordinary matter, and it is possible to closely determine what happens to positrons sent into a material. Positron spectroscopy can be used to resolve small defects that other probes, such as electrons, cannot distinguish. For example, if stainless steel is strained less than 10% of the way to failure, this early fatigue damage cannot be detected by standard nondestructive evaluation techniques, but a 210 ps positron lifetime signal increases in intensity from 15% to over 70% due to the increased number of defects [3]. Details about the solid structure are determined from the gamma rays released when the positron annihilates with an electron in the solid.

There are many unanswered questions about the behavior of positrons in solids, and, correspondingly, experimentalists sometimes use inaccurate models to interpret their data. This is where my thesis comes in. Because the problem of a positron in a crystal is too complex for an analytic solution, we have turned to a computational technique known as path-integral Monte Carlo (PIMC) to help determine the positron's behavior. We have developed code to model positronium (Ps), the bound state of a positron with an electron, in an external potential. This is unlike previous computer models of positrons in solids in that the positron and electron are simulated as having independent degrees of freedom, using their exact Coulombic propagator. This allows us to measure characteristics of the Ps atom that could not previously be studied.

## Methods

In my thesis, I remind the reader that the partition function needed to calculate thermal averages for a quantum particle is mathematically equivalent to the partition function for a classical ring polymer. In other words, we can represent the positron and electron as two closed chains of “beads,” as illustrated in Figure 1, where each bead represents the probability of finding the particle at that location. The springs between each pair of beads have spring constant  $\frac{mP}{\beta^2 \hbar^2}$ , where  $m$  is the mass of the quantum particle,  $P$  is the number of beads (typically in the thousands), and  $\beta$  is the inverse temperature.

In addition to the harmonic potential due to the springs, the potential felt by each bead is determined by the external potential and the Coulombic interaction. The



**Figure 1:** Models of quantum particles treated using PIMC. Left: single positron. Right: positron and electron. Thick black lines represent Coulomb interactions.

external potential felt by a bead at position  $\mathbf{r}$  is  $\frac{V(\mathbf{r})}{P}$ , where  $V(\mathbf{r})$  is the potential that the quantum particle would feel at that location due to the surrounding atoms. To determine the external potential for the positron, we use a technique called density functional theory (DFT). The external pseudopotential felt by the electron is difficult to calculate, and we use a phenomenological form based on experimental data.

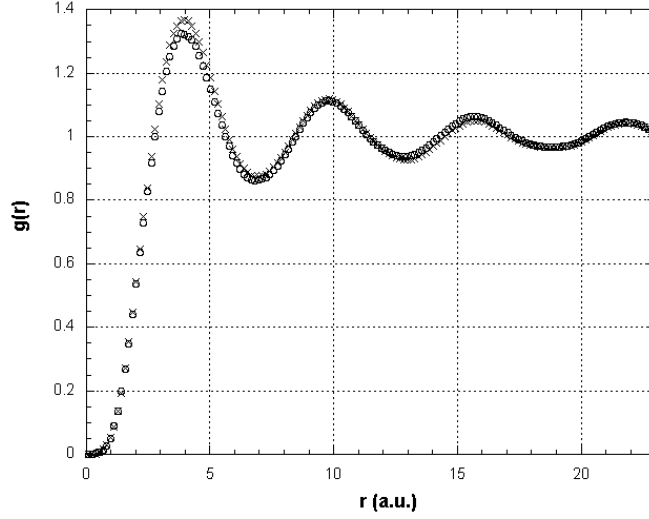
The simulation is performed using the standard Metropolis algorithm [4]: we choose states with probability proportional to their thermal likelihood, and then weight them equally as we calculate thermodynamic averages. We use this algorithm to move small sections of each chain for about a million passes, calculating quantities such as the probability density function  $P(r)$  (which gives the probability of finding the electron and positron a distance  $r$  apart), the pair correlation function  $g(r)$  (which gives the probability of finding a solid atom located a distance  $r$  from the positron), and the Ps energy and lifetime.

Once a PIMC code for Ps was written and debugged, we chose to study a highly idealized environment: spherical pores of realistic sizes. This work led to a publication, Ref. [5]. Proceeding to the task of modeling a more realistic solid, we chose to simulate solid argon (Ar) because of the presence in the literature of both experimental and theoretical results for this solid [6-12]. Also, since finding a good pseudopotential felt by the electron is one of the most challenging aspects of this method, we decided to employ our new simulation method in a system in which this potential has already been studied in detail.

## Results

There are a number of disputed facts about Ar that we have addressed. For instance, some researchers have attributed the longest lifetime of Ps in Ar to the self-trapping of Ps in voids [9]. Others, however, point out that the long lifetime is observed even at temperatures where there should be no voids [10]. We studied face-centered cubic (fcc) Ar and Ar with a monovacancy (for which we removed a single atom from the middle of each block of eight unit cells). We found no dramatic change in the lifetime or in the pair correlation function (Figure 2), indicating that the Ps atom does not fall into

the available void. Rather, due to the polarization potential of our model, it remains close to the Ar atoms. This is a significant result because it contradicts the standard assumption about the behavior of Ps near a vacancy.



**Figure 2:** Pair correlation function  $g(r)$  for Ps in Ar. Circles: Ps in solid Ar. Crosses: Ps in Argon with a monovacancy.

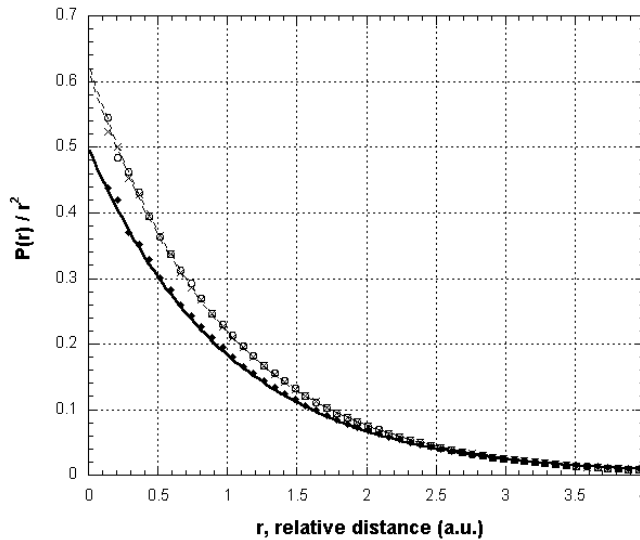
When Ps is placed in a solid, the change in the probability density where the relative coordinate between the particles is zero is described by  $\kappa$ , the “internal contact density” of Ps:

$$\kappa = \frac{|\psi(0)|^2}{|\psi_{\text{free}}(0)|^2}. \quad (1)$$

We can write the total annihilation rate of either state as

$$\Gamma = \kappa\Gamma_0 + \Gamma_{\text{p.o.}}, \quad (2)$$

where  $\Gamma_0$  is the self-annihilation rate of free Ps and  $\Gamma_{\text{p.o.}}$  is the rate of “pick-off” annihilation due to other electrons in the solid. We found that the Ps wavefunction is “squeezed” inside solid Ar, as seen in Figure 3, which shows  $P(r)/r^2 = 4\pi|\psi(r)|^2$ . (You can note in this figure the excellent agreement of our numerical estimate of  $|\psi_{\text{free}}(0)|^2$  with theory.) From the curve fit parameters we calculate the internal contact density of Ar to be  $\kappa_{\text{Ar}} = 1.25 \pm 0.02$ . While this makes sense given our potential,  $\kappa \leq 1$  in all the materials in which it has thus far been measured [13, 14]. To date, we have found no experimental data on  $\kappa$  in Ar with which we can compare our own, interesting result. It turns out that the hyperfine energy splitting between o-Ps and p-Ps scales with  $\kappa$  [15], and can be determined in materials using a technique called “magnetic quenching.” Future investigations in our group will hopefully involve simulating Ps in  $\alpha$ -SiO<sub>2</sub>, in which  $\kappa$  has been measured to be about 0.3 [14].



**Figure 3:** Probability density function. Solid line:  $1S$  exact theory. Diamonds: Free Ps simulation. Circles: Ps in solid Ar. Crosses: Ps in Ar with a monovacancy. Lines are curve fits: results for free Ps were fit to  $0.50e^{-1.00r}$ , the results for Ps in solid Ar were fit to  $0.63e^{-1.05r}$ , and the results for Ps in Ar with a monovacancy were fit to  $0.62e^{-1.05r}$ , where all uncertainties are in the last digit.

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