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Positron lifetime distributions in fluids

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Experimental studies of positron and positronium decay in dense fluids suggest that these particles are capable of forming self-trapped states in some fluids in a broad region of temperature and density surrounding the liquid-vapor critical point. A central question in understanding the phenomena is whether the light particle (lp) actively creates a static potential well in the fluid in which it localizes, or randomly visits favorable fluctuations. The fact that the experimental measurements yield a single, well-defined decay rate for each decay mode suggests that the environment of the lp is static. Earlier *mean-field* theories could not shed much light on this question. However, two recent applications of quantum Monte Carlo methods show that substantial fluctuations occur in the lp environment. In this paper, the distribution of fluctuations is described. It is shown that the apparent conflict is resolved if the time scale for fluctuations in the environment is much shorter than the duration of each measurement, and estimates for the different relaxation processes are provided.

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INTRODUCTION

It has been known for some time that an excess light particle (electron, positron, or positronium atom) which has thermalized in a fluid can become localized [1]. In contrast with the usual Anderson picture, localization occurs here because the light particle can influence its local environment, resulting in the name self-trapping [2]. The classic example of self-trapping is provided by the behavior of a positron, or positronium atom, in dense He gas at about 6 K [3]. The annihilation rate of each particle depends strongly on the density ρ [4]. At low densities, the decay rates of e^+ and orthopositronium (o-Ps) increase linearly with ρ simply because of the increased availability of electrons. (Because of the long vacuum lifetime of orthopositronium, its positron can annihilate via the pickoff process with a nearby atomic electron [4].) At a critical value, the annihilation rate of the positron increases almost discontinuously while, at a different value, that of the positronium suddenly levels off. Each of these strong departures from linearity is brought about by the sudden change in the number of available electrons which occurs upon localization. For the positron this results from the formation of a droplet of the fluid in its vicinity due to the attractive interaction with the fluid resulting from polarization. In contrast, the fermionic repulsion between the positronium and fluid atoms results in the formation of a bubble. It has been conjectured that localization occurs when the light particle finds an energetically favorable fluctuation in the fluid which it then stabilizes.

The earliest, and crudest, theoretical models for these classic experiments simply assumed that the droplet, or bubble, had a constant atomic density. When averaged over an ensemble this resulted in a spherical potential well whose ground state is occupied by the localized light particle [5]. Considerable improvements in this picture, e.g., introducing a continuously varying local density, led to a description which adequately represented the experimental measurements [6,7]. It was shown that a suitable free-energy density functional could be constructed in terms of $\psi(\mathbf{r})$, the local light-particle wave function, and $\rho(\mathbf{r})$, the local fluid density. Minimizing the free energy resulted in two coupled equations for $\psi(\mathbf{r})$ and $\rho(\mathbf{r})$ which define the localized state. Since $\psi(\mathbf{r})$ and $\rho(\mathbf{r})$ represent average quantities which result in nonuniform fluid structures with diameters of about 20 Å, this model is essentially mesoscopic. Thus it was possible to explain the helium experiments [3] in terms of a model based on a single dominant mesoscopic state.

Density-functional theory (DFT) supports the concept that the self-trapped state is dominant over a specific region of thermodynamic states (defined, say, by the average density and temperature of the fluid), and suddenly disappears when the region boundary is traversed. Outside of this preferred region, the coupled equations which self-consistently determine the localized state only admit to extended solutions. Thus the model is consistent with the experimental observation that the trapped state forms suddenly. In many ways, the transition from extended to localized state is reminiscent of a phase transition and it has been pointed out elsewhere that the mesoscopic amplitude ψ may be regarded as an order parameter [8].

Following these classic experiments [3], positron and positronium decay were also studied in other noble gases (e.g., argon [9] and xenon [10]) above their critical-point temperature. At these higher temperatures, the variation of decay rate with density on a given isotherm was markedly different than for the earlier work on helium. Although some gases still exhibited the general characteristics of droplet or bubble formation, the variation of decay rate was smooth and did not display a sharp transition.

Attempts were made to apply DFT to these gases as well, but the results were not so satisfying [9–12]. In general, at these higher temperatures, the fits to experimental data were superior for the case of positrons rather than for positronium. Even at the high temperatures considered, DFT still predicted a dominant, localized state in a particular density range. In common with the case for helium, the predictions still changed suddenly with the onset of localization. However, the variation of the *experimental* decay rates with density were all remarkably smooth and did not exhibit the sudden changes of slope described earlier. As before, they suggested that atomic clustering could occur in the neighborhood of the localized positron and that a cavity surrounded the positronium atom. However, the theoretical curves in the case of positronium were essentially independent of density (flat) in the trapping region [12], whereas the experimental curves increased monotonically.

Sharma and McNutt developed an alternative model based on the idea that an "intelligent" lp selects locations in the fluids where local-density fluctuations favor its existence [13]. Their semiphenomenological approach has been successfully applied to the analysis of *o*-Ps decay in a number of fluids at low density [9,14]. Its failure to correctly model data in the critical region was typically interpreted as confirmation that the lp is localized.

COMPARISON OF MODELS

DFT results in the computation of a single, optimal, localized state for the lp-fluid system. Consequently it is a mean-field theory and provides no information concerning fluctuations in the important physical quantities, such as the local density and the annihilation rate. Because its sole output is a single system state, it is easy to become seduced into believing that this state has a physical reality, i.e., that the lp truly sees a static local density which is successfully modeled by DFT. This picture is further supported by the ease with which experimentalists are able to assign specific annihilation rates to each decay mode. It can be argued that, if fluctuations in the lp environment are large, then there should be experimental ramifications such as the observation of a *continuous* distribution of lifetimes. This type of behavior has been observed in polymers, where the positron becomes localized in a rigid pore, or region of free volume [15,16]. The continuous distribution in pore sizes, or free volumes, results in a corresponding distribution of experimental lifetimes. It is surprising that this behavior is not observed in fluids. The question of whether or not this is a consequence of the absence of large local fluctuations has never been conclusively resolved.

The ideal physical system which DFT tries to emulate is the adiabatic model, in which the lp degrees of freedom are treated via quantum mechanics and the fluid degrees of freedom are treated classically. The lp interacts with a fluid atom located at $\mathbf{R}_j$ via a pairwise interaction potential, say, $w(\mathbf{x}-\mathbf{R}_j)$. In the past decade it has become possible to model this system directly, without invoking the further approximations asserted by DFT. The method used is referred to as quantum Monte Carlo (QMC), and is based on the discretized imaginary-time path-integral representation of the lp [17]. In QMC the quantum lp is represented by a fictitious classical ring polymer containing p sites with nearest-neighbor harmonic interactions characterized by a temperature-dependent force constant given by $4mp/\hbar^2\beta^2$.

Each site α of the polymer interacts with every fluid atom through the scaled down potential $w(\mathbf{x}_\alpha-\mathbf{R}_j)/p$. Thus, in the canonical ensemble, the probability density of the lp-fluid configuration space is given by

$$(1/Z)\exp[-\beta U(\underline{\mathbf{R}})-\beta\Phi(\underline{\mathbf{x}},\underline{\mathbf{R}})]\ , \tag{1}$$

where

$$\Phi(\underline{\mathbf{x}},\underline{\mathbf{R}})\equiv\sum_{1<\alpha<p}[(2mp/\beta^2\hbar^2)|\mathbf{x}_{\alpha+1}-\mathbf{x}_\alpha|^2 +(1/p)W(\mathbf{x}_\alpha,\underline{\mathbf{R}})] \tag{2}$$

and $U(\underline{\mathbf{R}})$ is the interatomic potential energy of the atoms with positions $\{\mathbf{R}_j\}=\underline{\mathbf{R}}$. In the limit $p\rightarrow\infty$, the equivalence is exact. During the last decade, QMC has been extensively applied to the behavior of an excess thermalized electron [18]. In some treatments, the effects of induced polarization of the fluid atoms have been considered explicitly [19].

The mean spatial extent of the polymer indicates the degree of localization of the lp. As expected, in the absence of the lp-atom interaction, it is on the order of the thermal wavelength. A localized lp is characterized by a strongly clumped polymer. Except for nearest neighbors, the mean distance between an arbitrary pair of polymer sites is nearly constant and independent of their relative position on the chain. An extended state is characterized by a unimodal distribution of site-site separations which is strongly peaked around symmetrically opposed sites. The fluid structure in the neighborhood of the lp is effectively investigated by constructing the site-atom and polymer center-atom radial distribution functions. Except for a small region near an atomic center where the repulsive interaction is strongest, at high temperatures these functions are nearly unity, indicating little or no lp-atom correlation. As the temperature is lowered, the range of correlation grows, suggesting that the lp is influencing its surroundings. It is greatest in the vicinity of the critical point, where the large compressibility of the fluid results in a strong response to the external perturbation produced by the lp [20].

In QMC, the mean values of physical observables can be expressed in terms of the appropriate polymer-atom correlation functions. For example, using appropriate units, the mean annihilation rate of a thermalized positron is given by

$$\langle\lambda\rangle=\rho\int d\mathbf{r}\,f_{\mathrm{el}}(\mathbf{r})g_{\mathrm{FP}}(\mathbf{r})\ , \tag{3}$$

where $f_{\mathrm{el}}(r)$ is the electron density a distance r from an atom, ρ is the average fluid density, and $g_{\mathrm{FP}}(r)$ is the polymer site-atom radial distribution function [21]. Similarly, the variance of λ can be expressed in terms of both two- and three-point correlations [21]. The same correlations are required for evaluating the pickoff decay rate of *o*-Ps. However, here the internal state of the *o*-Ps atom must also be known because it is a composite particle and the path integral only represents its center of mass [22].

In contrast with DFT, which suggests that localized states occur suddenly, and only exist within definite regions of the fluid parameters, QMC demonstrates that, at finite temperature, the degree of localization varies continuously with density. Recently, in two separate studies, QMC has been applied both to the problem of an excess positron [23] and an excess orthopositronium atom [22], which have thermalized in a fluid. The fluid was modeled by the Lennard-Jones (6,12) pair potential. Because experiments on positron and positronium lifetimes in xenon have disclosed an unusually large positron annihilation rate [10], the values of the parameters were chosen to represent xenon. In each study, specific isotherms were selected for their experimental interest. QMC was used to theoretically investigate the mean decay rate as a function of density on the isotherms chosen for each particle and compared with the observations.

These calculations showed improved agreement with the experimental measurements. They were especially good in modeling the region of transition away from linear density dependence, where major difficulties are encountered with DFT. Although the predictions must still be termed as qualitative due to the lack of reliable lp-atom interaction potentials, for the positron this situation is rapidly changing and *ab initio* calculations of effective positron-atom functions are finally becoming available [24].

ANALYSIS OF FLUCTUATIONS

In addition to determining the average annihilation rates at a specified density and temperature, the path-integral Monte Carlo computations were used to simultaneously generate the complete decay-rate equilibrium distribution. This is not possible with the earlier models since they are mean-field theories and, for the first time, provides the opportunity to answer questions concerning the constancy of the lp's environment.

Histograms representing the decay-rate distribution of e^+ and *o*-Ps at 340 K are displayed in Figs. 1–4 for a range of densities. A few general observations are in order: (i) in each case the *relative* width of the distribution is of order unity; (ii) in each case, the width is greater at lower density; and (iii) at higher density, the width is greater for *o*-Ps. It is clear that these distributions are totally different from those representing macroscopic quantities, where the relative width is of order $f^{-1/2}$, where f is the number of degrees of freedom of the system. It is convenient to characterize them by their relative dispersion $\zeta_\lambda = \sigma_\lambda / \langle \lambda \rangle$, where σ_λ^2 is the variance of the decay rate calculated for each distribution. For *o*-Ps in xenon at 340 K, $0.83 > \zeta_\lambda > 0.44$ for dimensionless density $\rho^* = \rho\sigma^3$ in the range $0.017 < \rho^* < 0.35$ [22]. σ is simply the Lennard-Jones length parameter (the value of r for which the potential vanishes); the value of ρ^* at the critical density is close to 0.35. For e^+ in xenon at $T = 340$ K, $1.2 > \zeta_\lambda > 0.18$ for $0.01 < \rho^* < 0.5$ [23].

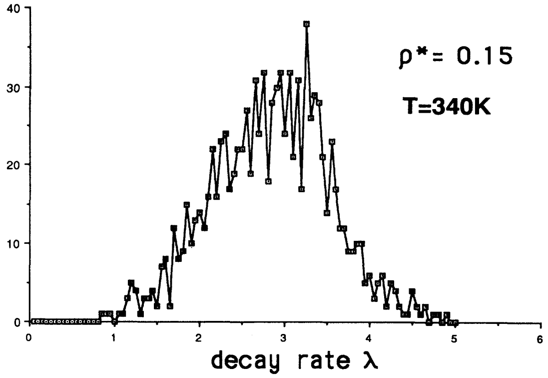

FIG. 2. Annihilation rate histogram for positrons thermalized in xenon at moderate density. Although atomic clustering about the positron is extensive at this density, the fluctuations in λ are still large.

These calculations clearly demonstrate that, in each case, the local environment of the lp fluctuates strongly. The question that needs to be addressed here is how these large variations can be reconciled with the experimental

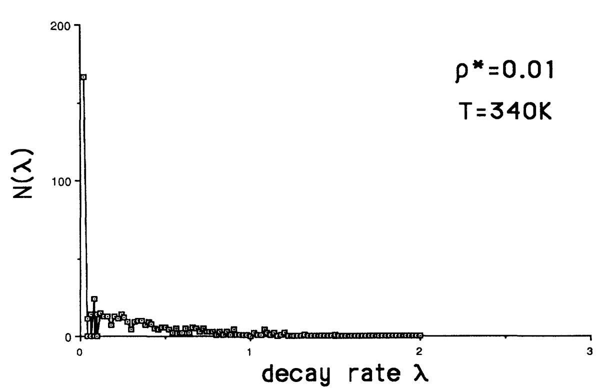

FIG. 1. Annihilation rate histogram for positrons thermalized in xenon at low density. The concentration of values near λ=0 occurs because of the large number of evacuated regions at this density.

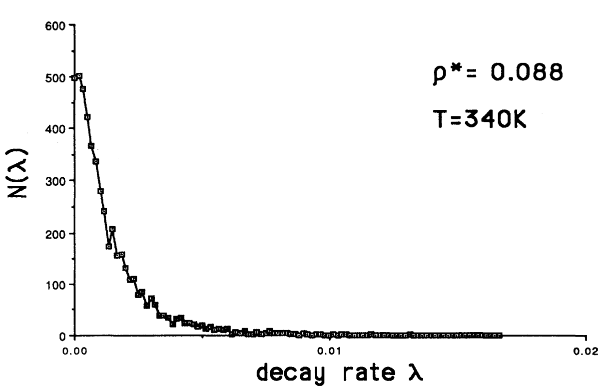

FIG. 3. Annihilation rate histogram for orthopositronium thermalized in xenon at low density. Again we see that *o*-Ps prefers empty regions, and that the variation in λ is large.

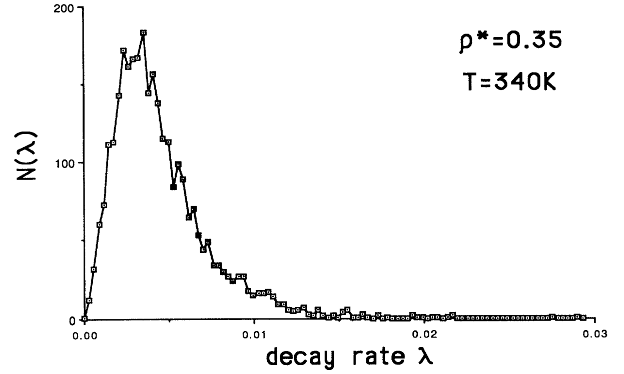

FIG. 4. Annihilation rate histogram for orthopositronium thermalized in xenon at the critical density. The large compressibility of the fluid near the critical point may contribute to the large fluctuations observed.

results which yield a single, easily resolved, annihilation rate for each particle.

For a positron, the annihilation rate is represented by the quantum-mechanical operator [21],

$$\hat{\lambda} = \sum_{1 \le j \le N} f_{\mathrm{el}}(\mathbf{x} - \mathbf{R}_j) \ . \tag{4}$$

It has been pointed out by Percus that the quantity that is actually measured in the laboratory is the mean value of $P(t)$, the probability of survival at time t [8]. Since the annihilation process is Markovian, for a given positron (or positronium atom),

$$P(t) = \exp\left[-\int_0^t \lambda(t')dt' \right] , \tag{5}$$

where here $\lambda(t)$ is the quantum average of the instantaneous annihilation rate experienced by the unstable particle, $\lambda(t) = \langle \psi(t) | \hat{\lambda} | \psi(t) \rangle$, and we have conditioned on the complete initial state of the lp-fluid system at the initial time $t=0$. To determine under what conditions an experiment should yield a single value for λ, let $\langle \lambda \rangle$ represent the mean annihilation rate as above and let $\delta\lambda(t)$ denote its fluctuation at time t. Then

$$P(t) = \exp\left[-\langle \lambda \rangle - \int_0^t \delta\lambda(t')dt' \right] \tag{6}$$

and its average is given by

$$\langle P(t) \rangle = \exp[-\langle \lambda \rangle t] \left\{ 1 + (\tfrac{1}{2}) \int_0^t dt' \int_0^t dt'' c_\lambda(t' - t'') \right\} , \tag{7}$$

where, in (7), we have expanded through second order in $\delta\lambda$ and $c_\lambda(\tau) = \langle \delta\lambda(\tau)\delta\lambda(0) \rangle$ is the decay-rate autocorrelation function. Taking the logarithm of each side, we find

$$\ln\langle P(t) \rangle = -\langle \lambda \rangle t + (\tfrac{1}{2}) \int_0^t dt' \int_0^t dt'' c_\lambda(t' - t'') \ . \tag{8}$$

Thus we need to consider the circumstances where the second term on the right can be neglected. This can occur in two ways. First, if c_λ is very small, only the mean value of $\lambda(t)$ will be detected. However, we have already demonstrated from QMC that $c_\lambda(0)$ is of order $\langle \lambda \rangle^2$ so this doesn't apply. Second, $c_\lambda(\tau)$ may become vanishingly small for times τ much shorter than $t \approx 1/\langle \lambda \rangle$, the time of observation. These issues may be clarified by (i) writing $c_\lambda(\tau) = c_\lambda(0) f(\tau)$; (ii) noting that $c_\lambda(0) = \sigma_\lambda^2 = \langle \lambda \rangle^2 \xi_\lambda^2$; and (iii) transforming the variables of integration in (8) to $\tau = t' - t''$ and $t_c = (t' + t'')/2$. In terms of these quantities (8) takes the form

$$\ln\langle P(t) \rangle = -\langle \lambda \rangle t + \langle \lambda \rangle^2 \xi_\lambda^2 \int_0^t d\tau f(\tau)(t - |\tau|) \ . \tag{9}$$

For simplicity, assume that $c_\lambda(\tau)$ decays exponentially ($f(\tau) = \exp(-\tau/\tau_\lambda)$) to obtain

$$\ln\langle P(t) \rangle = -\langle \lambda \rangle t + \langle \lambda \rangle^2 \xi_\lambda^2 t \tau_\lambda \times \{ 1 - (\tau_\lambda / t)[1 - \exp(-t/\tau_\lambda)] \} \ . \tag{10}$$

Since $\langle \lambda \rangle t \approx 1$, the condition which must be satisfied if the fluctuations don't contribute measurably to the observations is

$$\xi_\lambda^2 (\tau_\lambda / t) \{ 1 - (\tau_\lambda / t)[1 - \exp(-t/\tau_\lambda)] \} \ll 1 \ . \tag{11}$$

However, as noted above, the path-integral studies yield $\xi_\lambda^2 \approx 1$, so this criterion (11) can be satisfied only if $\tau_\lambda / t \ll 1$. We consider this possibility below.

Because $\lambda(t)$ depends on the atomic positions $\{\mathbf{R}_j,(t)\}$, we assume that its evolution is slaved to the evolution of density fluctuations in the fluid. For simplicity let's assume the relaxation time τ_λ is essentially that of the fluctuation which it has stabilized. For both e^+ and o-Ps, QMC indicates that the diameter of the disturbed region in the fluid, for which g_{FP} is different than unity, is on the order of 20 Å. In the case of e^+, a dense droplet is created in its vicinity [22]. Thus a good estimate of τ_λ for the positron should be the time required for an atom to diffuse 20 Å in the dense fluid. In contrast, a cavity is created around o-Ps which is nearly vacant [21]. An estimate of the relaxation time of this structure is approximately the time for an atom to ballistically traverse the cavity. Consequently, the relaxation time for o-Ps is expected to be much less than that for e^+.

There are a number of reports of experimental measurements of the diffusion constant of atomic xenon. In a recent study, values are reported in the density range of interest [25]. An examination of the data indicates that the diffusion constant of xenon varies slowly with the thermodynamic parameters in a region above the critical density and temperature with a value of approximately $D \approx 2.0 \times 10^{-8}$ m^2 s^{-1}, which yields $\tau_\lambda \approx (20 \text{ Å})^2 / D \approx 0.2$ ns. Now typical values of $\langle \lambda \rangle^{-1} \approx t$ for e^+ annihilation in Xe at 300 K range from about 0.7 to 2.0 ns, yielding $0.1 < \tau_\lambda / t < 0.25$. For the case of positronium, $\langle \lambda \rangle^{-1} \approx t$ is of much longer duration, since the vacuum lifetime of o-Ps is about 140 ns. In addition, the relaxation time is controlled by ballistic motion, which is much more rapid than diffusion. Consequently, $\tau_\lambda/t \ll 1$ for *o*-Ps decay.

CONCLUSIONS

In summary, although light particles show a tendency to localize in dense gases above the critical temperature, recent QMC studies confirm the existence of large statistical fluctuations in their environment [22,23]. This demonstrates that the concerns expressed frequently by Sharma [26] with the view that the localized lp sits in a rather static environment are justified. The Monte Carlo calculations appear to be in conflict with the experimental lifetime measurements, which yield a single, well-defined lifetime for each decay mode. By combining the information which has just become available concerning the size of the relevant statistical fluctuations with "back of the envelope" estimates of the fluctuation relaxation times, we have shown that, for the case of positronium, the conflict is resolved because the time of observation, which is roughly the lifetime, is much longer than the appropriate relaxation time. Consequently, the fluctuations in lifetime occur too rapidly to be observed. However, for the bare positron, the separation of time scales is only marginal. Experimentalists may view this statement as a challenge to detect a measurable lifetime *distribution*. Theorists and computer modelers may wish to improve on the rough estimates given here to more accurately fix the relaxation times.

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