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Positronium Bose-Einstein condensation in liquid ⁴He bubbles

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A hollow spherical bubble containing thousands of spin-aligned triplet positronium (Ps) atoms in superfluid liquid 4 He would be stable against breakup into smaller bubbles, and the Ps would form a Bose-Einstein condensate (BEC) with a number density of $\sim 10^{20}$ cm⁻³ and a BEC critical temperature $T_c \approx 300$ K. Estimates suggest that one could make such bubbles in the laboratory containing 10^5 Ps atoms using presently known methods.

I. INTRODUCTION

Positronium (Ps) is the hydrogen-like bound state of an electron and its positron antiparticle. The ground state of Ps is split into (1) a singlet with a mean lifetime for decay into two 511 keV photons of 125 ps; and (2) a triplet state with a 142 ns mean lifetime for decay into three photons with total energy of 1,022 keV [1, 2]. Hollow bubbles in liquid helium containing single positronium atoms were discovered in 1957 when R. A. Ferrell's bubble model [3] explained the long lifetimes, nearly the same as the vacuum triplet Ps lifetime, that had been observed for positrons annihilating in liquid ⁴He [4, 5]. Analogous bubbles containing single electrons were elucidated in 1961 [6] and single Ps cavities [7] and electron cavities [8] were discovered in He vapor at low temperatures. Bubble states of single alkali and alkaline earth atoms in liquid helium have recently been demonstrated theoretically [9]. The existence of these different kinds of bubbles is attributed to the Pauli exclusion principle [10, 11], whereby the filled He 2S electron shell strongly repels both free electrons and electrons bound in Ps or other atoms. At a pressure of 1 atm a bubble containing a single electron in liquid helium has a bubble inner radius of (1.72±0.02) nm and an effective mass of about 200 ⁴He masses [12, 13] that is nearly the same as the mass of the ~270 helium atoms comprising the innermost layer of the bubble. A Ps bubble has a nearly identical radius measured to be (1.73±0.13) nm [14, 15], and thus will likely have nearly the same effective mass as an electron bubble. The similar physics of electron and positronium scattering at low energies [16, 17] and the similarity of single-electron and single-Ps atom bubbles suggest the possibility of forming bubbles in liquid He containing many Ps atoms. The existence of multi-electron bubbles [18, 19, 20, 21] in which the electrons form a twodimensional gas on the inner surface of the bubble means that that these structures might be prepared as needed and filled with Ps [22]. Analogous bubbles filled with H1 and with dimensions on the order of 100 µm and densities of 10¹⁹ cm⁻³ have been produced in liquid He [23, 24].

If formed from partially spin-polarized positrons, the minority spin Ps atoms in a many-Ps bubble would decay via collisions with other Ps atoms via spin exchange [25] followed by two-photon electron-positron annihilations. Calculations suggest that this would result in the remaining

Ps forming a single-component triplet Ps Bose-Einstein condensate (BEC) [26, 27, 28] with a critical temperature of about 300 K. Such a route to a Ps BEC would have the unique advantage of self-assembled containment for the BEC, unlike for the case of the even more exotic proposed muonic hydrogen BEC [29]. The time for Ps to cool to less than 100 K in liquid He is likely to be much less than the 125 ps singlet Ps lifetime [14] and would be orders of magnitude faster than in containers made of ordinary materials [30, 31, 32]. The Ps-wall interactions [33] would be very well understood and one might produce BEC's extended in one dimension that would be suitable for observing stimulated annihilation [34, 35]. Ps BEC bubbles could also be manipulated in interesting ways using acoustic cavitation [36]. For example a bubble of 1 μ m radius and containing 10⁸ Ps atoms could in principle be compressed in a few ps to a 100 nm radius by an imploding spherical acoustic wave to produce a neutral pair plasma [37, 38, 39, 40, 41, 42] with an electron density equal to that of metallic sodium.

II. BUBBLE PARAMETERS

The radii of Ps bubbles in liquid ⁴He can be calculated following the method of Ferrell [3] who found the radius of a single Ps bubble by minimizing the total energy. The latter was taken to be the sum of the zero point energy of the Ps atom confined by the infinite potential walls of a hollow sphere of radius r, $E_0 = \pi^2 \hbar^2 / 4 m_e r^2$, plus the bubble surface energy $E_S = 4\pi r^2 \sigma$, where σ is the surface tension, $\sigma = 0.95 \times 10^{-4} \text{ J m}^{-2}$ at 4.2 K and $3.1 \times 10^{-4} \text{ J m}^{-2}$ at 2.0 K [43, 44]. Including the contribution of the hydrostatic pressure p, the total energy is

$$E_{Total} = \pi^2 \hbar^2 / 4m_e r^2 + 4\pi r^2 \sigma + \frac{4}{3}\pi r^3 p. \tag{1}$$

At 1 atm pressure this equation predicts that the equilibrium single Ps bubble radii are r = 1.51 nm at 2.0 K and r = 1.73 nm at 4.2 K; with zero pressure, the equilibrium radii are 1.67 nm at 2.0 K and 2.24 nm at 4.2 K.

The size of a multi-Ps BEC bubble in liquid 4 He may be calculated in a similar manner except that the zero point energy is replaced by E_0 , the weak scattering approximation of the ground state energy of a BEC within a spherical

potential well of infinite height, radius r, and volume V containing N identical Bose particles of mass m_{Ps} characterized by a positive s-wave scattering length a [45]:

$$E_0 = \frac{2\pi\hbar^2 aN^2}{m_{p_s}V} = \frac{3\hbar^2 aN^2}{2m_{p_s}r^3}.$$
 (2)

This expression should be valid provided a/λ and na^3 are both much smaller than 1 [46], where $\lambda = \sqrt{2\pi\hbar^2/m_{Ps}kT}$ is the Ps thermal de Broglie wavelength at a temperature of 2 K and n is the Ps number density $n<10^{21}$ cm⁻³. Under these conditions both a/λ and na^3 are less than 0.003. The radius for a triplet Ps BEC bubble is then found by minimizing the total energy

$$E_{total} = \frac{3\hbar^2 a N^2}{2m_{p_s} r^3} + 4\pi r^2 \sigma + \frac{4}{3}\pi r^3 (p - p_{vap}), \tag{3}$$

and the pressure term now includes a correction for the vapor pressure of the Ps gas [47],

$$p_{vap} = \frac{kT}{\lambda^3} g_{5/2}(1) = (m_{Ps} / 2\pi\hbar^2)^{3/2} (kT)^{5/2} g_{5/2}(1).$$
 (4)

Here $g_{5/2}(1) = 1.34149...$ and the vapor pressure will be < 0.13 atm for Ps temperatures less than 100 K.

Using the triplet Ps - triplet Ps scattering length a = 3.00 $a_{\rm Bohr}$ [48] and the surface tension $\sigma = 3.1 \times 10^{-4}$ J m⁻² at 2.0 K, we calculate in Fig. 1 the bubble radius r and the number density n of triplet m=1 Ps atoms as a function of the total number of Ps atoms N for various hydrostatic pressures, neglecting the Ps vapor pressure. For slightly negative pressures the bubbles become unstable at high values of N. For $N > 10^5$ and at a positive hydrostatic pressure of 1 atm the Ps number density is nearly constant, $n = 1.3 \times 10^{20}$ cm⁻³, for which the BEC critical temperature would be $T_c \approx 370$ K. We thus see that even if the Ps does not immediately thermalize to below 100 K it is still going to Bose-Einstein condense with the fraction of the atoms in the ground state of the bubble given by

$$f_{Condensed} = 1 - (T/T_c)^{3/2} > 0.8.$$
 (5)

Since the bubble energy is positive, one might wonder about the stability against break-up of a large bubble into smaller bubbles. If the pressure is zero, Eq. 3 implies that a bubble containing N Ps atoms has positive energy $E(N) = A \times (N)^{4/5}$ where $A = 5.148 \times 10^{-21}$ J. A bubble with 2N particles has energy

$$E(2N)=A\times(2N)^{4/5}=2^{4/5}\times E(N)=1.74\times E(N),$$
 (6)

A bubble with 2N particles has 13% less energy than the sum of the energy of two separate bubbles with N particles each, and therefore a large bubble is stable to breakup into two smaller ones. The concomitant heating of the Ps due to the

merging of two bubbles of 10⁵ Ps each would be about 3 K. This temperature rise would have little effect on the merged Ps state since the temperature rise is small compared to the

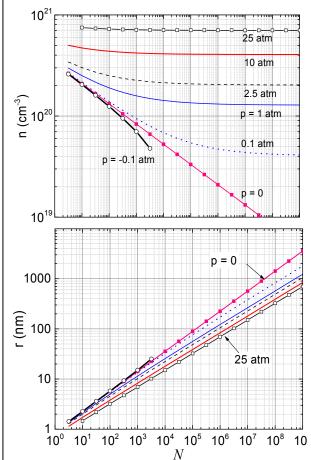


Fig. 1. Calculated bubble radius r and Ps number density n at 2 K as a function of the total number of BEC Ps atoms N for various pressures neglecting the vapor pressure of the Ps gas. Note that the plots are on log-log

BEC critical temperature ~300 K.

III. SIGNATURE OF A Ps BEC BUBBLE

An experimental signature for distinguishing a state consisting of many single Ps bubbles versus the same number of 100% spin polarized Ps atoms in a few large BEC bubbles is that the lifetime of the first is (91 ± 5) ns [4], while the lifetime of the BEC state would be within a few percent of the 142 ns vacuum lifetime of triplet Ps [49]. This is because the decay rate due to collisions of the Ps with the He atoms of the bubble wall would be a negligible factor of $N^{-1/3} \approx 0.1 - 0.01$ times the wall component of the single Ps bubble decay rate $\sim 4 \text{ us}^{-1}$.

A superior signature of a Ps bubble BEC would be to observe the angular correlation of the two-photon annihilations induced by suddenly (~10 ns) applying a 1 T magnetic field transverse to the polarization direction. The annihilation photon pairs from the BEC state Ps would be

essentially perfectly anticollinear compared to the 60 μ rad full width at half maximum expected from the two-photon annihilations of Ps with a thermal distribution of velocities at 2 K. This signature could be acquired using a multi-counter detector for measuring the angular correlation of annihilation radiation [50, 51]. A third signature of the Ps BEC would be the observation of a very narrow resonance (~10 GHz FWHM) using co-propagating two photon 1S-2S spectroscopy [52].

IV. PRODUCTION OF PS BEC BUBBLES

We now consider how one might produce a multi-Ps bubble beginning with the trapping [53] and accumulation [54] of 100 ns pulses of 3×10^7 monoenergetic 5 keV partially spin polarized positrons. These pulses would first be focused to a 50 µm spot on a Ni(100) single crystal positron remoderator [55, 56, 57] in vacuum. The positrons will be about 28% polarized along their velocity direction [58]. At the exit side of the Ni crystal approximately 15% of the positrons are reemitted with energies of 1.0 eV and an energy spread of ~ 40 meV [59]. The 4×10^6 remoderated [60] positrons will be accelerated to 5 keV, and implanted into a spot of area 0.5 µm² on a diamond film of thickness 250 nm, as indicated in Fig. 2. The median positron stopping depth will be ~130 nm [61] and the positrons will stop in a broad distribution about the mean. From the measured mobility of positrons in natural diamond at 100 K, $\mu_{+} \approx 240 \text{ cm}^{2}\text{V}^{-1}\text{s}^{-1}$ [62], we find the positron diffusion coefficient from the Einstein-Smoluchowski relation $D = \mu k_B T/e = 2.1 \text{ cm}^2 \text{s}^{-1}$. This implies that thermalized stopped positrons diffuse a mean distance corresponding to half way across the diamond film in their mean lifetime (97.5 ± 1.5) ps in isotopically pure diamond [63]. About 20% of the incoming positrons will be emitted into the liquid He in the form of Ps at the diamond exit surface with energies from 0 to 3 eV [64]. About 14% of these (half of the 28% positron polarization), or 10⁵ pure m=1 triplet Ps atoms, will survive the ensuing spin exchanging collisions. The emitted Ps, indicated by the shaded area in Fig. 2, will immediately form single Ps bubbles which then coalesce into ever larger bubbles [65]. Note that the Ps-He total cross section at 0-1 eV is $\sim 12\pi a_0^{\frac{1}{2}} \approx 1.0 \times 10^{-15}$ cm² [66]. At 1 eV the Ps velocity is $\sim 6 \times 10^7$ cm s⁻¹. The liquid He number density is $n_{\rm He} = 1.88 \times 10^{22}$ cm⁻³, so the Ps mean free path at 1 eV is $(n_{\text{He}}\sigma)^{-1} = \sim 0.5$ nm and the Ps slowing down time to 0.1 eV or so for say 1000 collisions is ~1 ps. Assuming the Ps scatters randomly in the He it will thus have many chances to form a single Ps bubble.

We now have to ask: (1) Is a 100 ns time scale sufficient for the organization of this collection of Ps and He atoms into one or more multi-Ps bubbles? and (2) Is the thermal conductivity of the superfluid He sufficient to remove the heat of the positronium injection and thermalization? The answer to the first question is probably "yes", since the displaced He atoms in forming a large bubble will only have to move ~ 100 nm in 10^{-7} s, which corresponds to an average

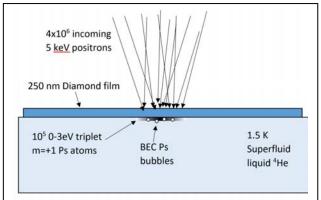


Fig. 2. Geometry of a target for forming BEC positronium bubbles in superfluid He. Energetic positrons stop in a thin diamond film. The positrons thermalize and diffuse to the back surface of the film, and are emitted into the He as positronium which collects into bubbles.

velocity $v \approx 100 \text{ nm} / 10^{-7} \text{ s} = 1 \text{ m/s}$, much less than the speed of sound in Liq He which is 234 m/s at 1.5 K [67].

To answer the second question we need to determine the fate of the energy of 4×10^6 5 keV positrons deposited in the diamond film, $E_{\rm diamond}=3.2\times10^{-9}\,\rm J$, and the energy of 10^5 1-3 eV Ps atoms deposited in the liquid He, $E_{LHe}=2.4\times10^{-14}\,\rm J$. The corresponding heat fluxes for a 100 ns deposition time and area 0.5 μm^2 are $F_{\rm diamond}=6.4\times10^6\,\rm W\cdot cm^{-2}$ and $F_{LHe}=48\,\rm W\cdot cm^{-2}$. The thermal diffusion coefficient in isotopically pure diamond [68] is $10^4\,\rm cm^{-2}s^{-1}$ below 100 K so the implantation energy will be spread out to a radius of 300 μm in 10^{-7} s and the heat flux into the liquid He will be reduced to 30 W cm⁻². In superfluid 4 He the maximum heat flux that can be tolerated between two points that are separated by a distance L at 1.8 K and 2.17 K is [69, 70]

$$\dot{q}_{\lambda} = 5.5 \,\mathrm{W} \cdot \mathrm{cm}^{-2} \times (1 \,\mathrm{cm}/L)^{0.294} \,.$$
 (7)

For L = 250 nm, $\dot{q}_{\lambda} = 124 \, \mathrm{W \cdot cm^{-2}}$, which should be sufficient to carry away the energy of both the Ps atoms and the stopping positrons.

The diffusion coefficient for single Ps atom bubbles in liquid He may be found from the fluctuation-dissipation relation. In particular the Stokes-Einstein relation for diffusion of spherical particles through a liquid with low Reynolds number says the diffusion coefficient D is related to the viscosity η by

$$D = \frac{k_B T}{6\pi \eta r} {8}$$

For a Ps bubble of radius 2 nm in liquid He-II at 1.6 K where the viscosity is $\eta = 1.3 \times 10^{-6}$ Pa·s [71, 72], the diffusion coefficient is

$$D = 4.5 \times 10^{-5} \text{ cm}^2 \text{s}^{-1}$$
 (9)

From this we determine that the single Ps-bubble diffusion length in pure liquid He is $\lambda = \sqrt{Dt} = 7$ nm for t = 10 ns and 20 nm for t = 100 ns. The mean free path for single Ps bubble-bubble collisions is thus such as to lead to the conclusion that bubble coalescence will be rapid. On the contrary, a 100 nm radius bubble moves only 2 pm in 100 ns in response to the buoyancy force of the liquid He. This implies that there should be ample time for the coalescence of many single Ps bubbles into one or a few many Ps bubbles.

V. UTILITY OF Ps BEC BUBBLES

It is interesting to ask if one might scale up the Ps bubble BEC concept to obtain evidence for stimulated annihilation. First we need a means for flipping a triplet Ps BEC into the singlet state in a time shorter than the 125 ps singlet Ps lifetime so that the entire collection of Ps atoms may decay into two photons at about the same time. In principle one could accomplish this by adiabatic rapid passage [73] using a swept frequency pulse of RF that passes through the 203 GHz Ps $1^3S_1 \rightarrow 1^1S_0$ resonance [74].

The exactly on-resonance cross section for the single photon stimulated two-photon annihilation of an individual Ps atom [75] is $\sigma = 10^{-20}$ cm² [34]. However, when the nominal stimulated gain is less than one,

$$G_{\text{Nominal}} \equiv l \langle n \rangle \sigma < 1,$$
 (10)

the effective gain for a photon travelling a distance l through a BEC of average ${}^{1}S_{0}$ Ps density $\langle n \rangle$ will be [34, 76]

$$G_{\text{Below threshold}} = \sqrt{l\langle n\rangle\sigma}$$
 (11)

This amazing prediction of larger than expected gain in the below threshold limit would imply that experimental evidence for stimulated annihilation might not be so difficult to attain, requiring as few as 10⁹ BEC singlet Ps atoms in a suitable geometry.

VI. CONCLUSIONS

The number density of a gas of spin polarized Ps contained within a hollow spherical bubble in liquid helium has been calculated as a function of the number of Ps atoms, N, and applied pressure. The contained Ps gas should be a Bose-Einstein condensate with its temperature not far from that of the liquid He and with a BEC critical temperature greater than 300 K. It appears that bubbles with $N \approx 10^5$ could be created and the Ps momentum distribution measured using current technology. Further developments could lead to experiments demonstrating stimulated annihilation. The many-Ps bubbles should make possible the reproducible production not only of a BEC but also of various states of the

neutral e⁺-e⁻ plasma [77] that might appear upon sudden compression to higher densities.

The above discussion has introduced a well-defined set of many-positronium systems, the Nth member of which consists of N spin-polarized Ps atoms confined within a hollow spherical bubble of radius r(N) [see Fig. 1] in liquid He at a standard temperature and pressure. Born from Ferrell's original concept of the single Ps bubble in liquid He [3], the members of this new endless set may be thought of as cousins of Wheeler's polyelectron series, Ps, Ps $^-$, Ps $^+$, ... [78] which terminates at Ps $_2$ [79]; the spherical He bubble walls make up for the lack of chemical binding that brings the original polyelectron series to an end [80].

VI. ACKNOWLEDGMENTS

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