

## Possibilities for Bose condensation of positronium

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In this paper we consider the possibilities for producing a dense gas of  $N \cong 10^5$  triplet positronium (Ps) atoms in vacuum contained in small cavity  $V \cong 10^{-13}$  cm<sup>3</sup>. We then consider the scenario where this dense gas of polarized Ps atoms may cool through a weakly interacting Bose transition. The rates for thermalization with the wall, equilibrium in the bulk, and loss of polarization by exchange collisions are calculated. A method for observing the transition is also discussed.

### I. INTRODUCTION

Bose-Einstein condensation, one of the most striking phenomena in statistical physics, occurs when a macroscopic fraction of an ensemble of particles obeying Bose statistics collapses into a single state at low temperatures.<sup>1,2</sup> For noninteracting nonrelativistic particles with mass  $m$  at a density  $n$  in three dimensions (3D) the critical temperature  $T_c$  below which the lowest-energy state is macroscopically occupied is determined by

$$n\lambda_D^3 = 2.65, \quad (1)$$

where the de Broglie wavelength is given by

$$\frac{\lambda_D}{2\pi} \equiv k_D^{-1} = (2mT)^{-1/2}. \quad (2)$$

At the temperature determined by Eq. (1) the chemical potential  $\mu$  goes through zero. As the temperature is lowered below  $T_c$  the number of particles in the zero-momentum state  $\langle n_0 \rangle$  develops a macroscopic value:

$$\langle n_0 \rangle / n = 1 - (T/T_c)^{3/2}. \quad (3)$$

The massive Bose particles we have available to us in the laboratory are usually atoms made up of even numbers of fermions bound with some typical atomic ( $\cong 10$  eV) energy  $E_A$ . This binding energy is usually large compared to the energy (temperature) scales of interest. Because of this the component particles interact with a short-range force. At low energies, i.e., temperatures  $k_B T \ll E_A$ , the interactions may be characterized by an  $S$ -wave scattering length  $a$ . If  $g \equiv na^3 \ll 1$  the system is close to ideal and for  $T < T_c$  perturbation theory in  $g$  describes the ground state of the system and its excitation spectrum. Near  $T_c$  there are fluctuation corrections arising from critical phenomena, which even for a weakly interacting system, are nontrivial and interesting. These occur in a critical region of width<sup>3</sup>

$$\frac{T - T_c}{T_c} \leq g^{1/3}. \quad (4)$$

They lead to a shift in  $T_c$  from the value given in Eq. (1) and a change in the classical exponent ( $\frac{3}{2}$ ) appearing in Eq. (3). They also modify the dynamical behavior of the

system, for example, the dynamic structure factor  $S(k, \omega)$  (Ref. 4) shows critical behavior.

It is generally believed that the superfluid properties of He<sup>4</sup> below the  $\lambda$  point arise from a nonideal Bose condensation.<sup>2</sup> While much progress has been made on microscopic theories of the  $\lambda$  transition, many details are not amenable to calculation since the system consists of a *strongly interacting* set of bosons. Aside from liquid He<sup>4</sup>, the other possible systems that we might study are not only metastable, but are comprised of a relatively small number of particles. Rather than detracting from their utility, such systems are actually of interest for the light they might shed on nonequilibrium processes and the effects of small numbers of particles on thermodynamics.

Of the alternative Bose systems to He<sup>4</sup>, spin-polarized atomic hydrogen H $\downarrow$  has received the most attention.<sup>5</sup> It is known that a gas of polarized atomic hydrogen will remain a gas, i.e., the molecule does not form and it will not freeze into a solid, down to zero temperature. At densities  $n \cong 10^{18}$  cm<sup>-3</sup> the system is weakly interacting and will Bose condense at temperatures of roughly  $10^{-2}$  K. Although a gas of H $\downarrow$  is a good approximation to an ideal Bose system, workers have been unable to achieve high enough densities or low enough temperatures to observe its Bose condensation. The gases formed from atomic vapors of Na and Cs, which are also *candidates* for Bose condensation,<sup>6</sup> have been successfully cooled to microdegree temperatures by laser fields but no results on Bose condensation have been reported.

The exciton gas produced by pumping an insulator like Cu<sub>2</sub>O with a short laser pulse is also a promising candidate.<sup>7</sup> This system is in many ways very analogous to the positronium system we will discuss here. Densities of  $10^{18}$  cm<sup>-3</sup> with a predicted transition temperature of 10 K have been achieved. Interpretation of the results, which are mostly in the form of luminescence spectra, have recently been shown to be consistent with an almost ideal Bose condensation of excitons with weak interaction effects.<sup>8</sup> The details of, the interaction between the excitons, their interactions with the confining potential, and the effects of other low-lying excitations in the solid all depend on specifics of the host Cu<sub>2</sub>O materials. In particular the band structure, phonon spectra, and the electron phonon coupling constants will surely play an essential role in the quantitative aspects of the phase boundary and

the kinetics of the Bose condensation.

In this paper we show that a rather ideal but somewhat more exotic system might, in fact, be a very good candidate for observing a weakly interacting Bose condensation with very well-defined interactions between the bosons. We refer to a dense gas of positronium (Ps) atoms, in *vacuum*. Recent investigations of the interactions of positrons ( $e^+$ ) and (Ps) with solids have led to extraordinary improvements in the kinds of low-energy experiments we can do with the positron.<sup>9,10</sup> All of the ingredients necessary to achieve Bose condensation of Ps atoms are currently available. We envision a scenario where roughly  $N \cong 10^5$  Ps atoms are trapped in a volume  $V \cong 10^{-13}$  cm<sup>3</sup> and allowed to cool through the Bose transition temperature of 20–30 K in a time of the order of nanoseconds.

## II. SINGLE POSITRONIUM PHYSICS

Ps is comprised of an  $e^+e^-$  bound in a hydrogenic orbit. Its mass,  $2m_e$ , is extremely light compared to H, an important ingredient for achieving reasonable Bose condensation temperatures. [See Eqs. (1) and (2).] Its binding energy is half that of H. The ground state of Ps is a spin singlet separated from an excited triplet by an energy  $\Delta E_{ST} \cong 10$  K. Ps annihilates itself, i.e., becomes high-energy  $\gamma$  rays.<sup>11</sup>

The annihilation characteristics of the Ps atom are dependent on which of the two ground states it is in. The ground-state singlet ( $s$ ) is short lived with a lifetime  $\tau_s = 1.25 \times 10^{-10}$  sec. For a Ps atom at rest the decay occurs with the emission of two 0.5-MeV  $\gamma$  rays which come out precisely in opposite directions due to momentum conservation. If the Ps atom is moving with momentum  $p$ , the  $\gamma$  rays come out with a small angle  $\Theta \cong p/mc$  relative to each other. The nearby triplet state ( $t$ ) is prohibited by selection rules from decay into the two  $\gamma$ -ray channels. It instead decays by three  $\gamma$  rays with a spread of energies and a much longer lifetime of  $\tau_t = 1.42 \times 10^{-7}$  sec. A magnetic field mixes triplet with singlet thus rapidly quenching the triplet.

Like hydrogen, the Ps<sub>2</sub> molecule exists in an overall singlet state.<sup>12</sup> It has a binding energy  $E_M \cong 0.4$  eV. Two low-energy Ps atoms scatter from one another with a cross section  $\sigma \cong 4\pi a^2$  determined by how close the bound state is to the continuum.<sup>13</sup> Specifically,  $a_s = (2mE_M)^{-1/2} \cong 3$  Å, and  $\sigma_s \cong 10^{-14}$  cm<sup>2</sup>. On the other hand, the nonsinglet channel has a scattering length more like a Bohr radius, i.e.,  $a_t \cong 1$  Å and  $\sigma_t \cong 10^{-15}$  cm<sup>2</sup>. Of course, the experiment we are considering here would, for the first time, be sensitive to such cross sections and a measurement of them should be possible.

## III. $e^+$ AND Ps AT SOLID SURFACES

In order to make the case for Bose condensation we need to understand at least qualitatively how a mildly energetic ( $\approx 10$  keV) beam of  $e^+$  interacts with a semi-infinite slab of some simple solid.<sup>9,10</sup> The interaction process is sketched in Fig. 1. The incident beam enters the solid and begins to make electronic excitations in the

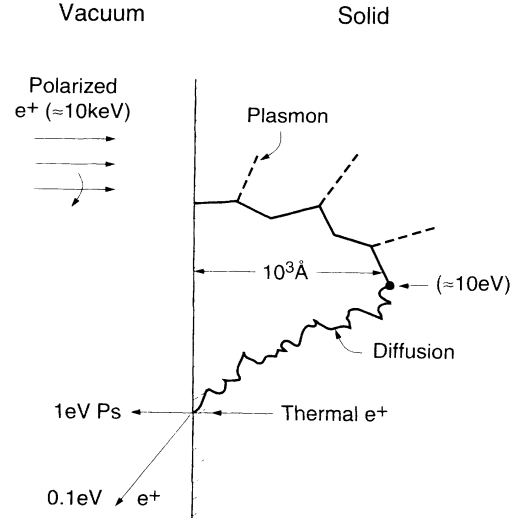


FIG. 1. Schematic of the interaction of a 10-keV beam with a typical solid like Si.

material, plasmons, inner-shell ionizations, etc. The energetic  $e^+$  reaches an energy of 10 eV or so in a short time  $\tau < 10^{-13}$  sec, and does so in a distance of roughly 1000 Å for a 5-keV beam.<sup>14</sup>

At this point in its history the  $e^+$  begins to slow down to thermal energies by electron-hole pair excitations and phonon emission. In this regime, its motion in the solid may be described as diffusive. Calculations and experiments convincingly demonstrate that diffusion coefficients  $D_p$  for our superthermal  $e^+$  are roughly 1 cm<sup>2</sup>/sec.<sup>9,15</sup> Since annihilation times in the solid  $\tau_A$  are, in most materials, a fraction of a nanosecond a typical  $e^+$  diffuses a distance  $l_D \cong \sqrt{D_p \cdot \tau_A} \cong 2 \times 10^3$  Å. This simple estimate suggests that some large fraction (50%) of our initial 5-keV  $e^+$  beam returns to the surface.

Since the work function for  $e^+$  and Ps is negative in many materials (Ni, Al, . . .),<sup>9,10</sup> emission of  $e^+$  and Ps occurs with high probability. Typically the energy of the  $e^+$  is nonthermal, i.e., directed perpendicular to the surface and greater than 1 eV while the Ps energy distribution is similarly nonthermal but most often greater than 1 eV. This unique surface interaction characteristic permits the generation of very bright pulsed  $e^+$  beams. [See Canter in Ref. 10.]

## IV. DENSE POSITRONIUM

The “initial” condition for the Ps Bose condensation experiment is sketched in Fig. 2. A bunched 5-nsec brightness-enhanced microbeam (diameter  $1\mu$ ) consisting of  $N \geq 10^6$   $e^+$  at an energy of 5 keV is incident at time  $t=0$  (5-nsec uncertainty) on the surface of a Si target which has, at the point of entry, a small cavity etched into it. We choose Si rather than a metal for several reasons to be discussed shortly.

The geometry we envision will typically be a cylindrical cavity of  $1\mu$  diameter with a height of  $d=1000$  Å. These dimensions are rather easily achieved with conventional lithography techniques. The  $e^+$ , as discussed, stop in the Si at a depth of about 1000 Å. About one-quarter

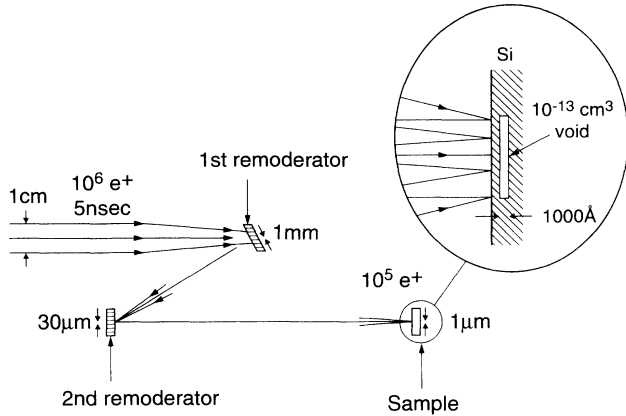


FIG. 2. Sketch of a pulsed brightness-enhanced  $e^+$  microbeam incident on a Si target with a void etched in it. The dimensions are typical dimensions for the proposed experiments.

of the initial  $e^+$  will be reemitted as electron volt Ps into the cavity. The singlet Ps rapidly decays leaving us with a hot gas of bosons, triplet Ps atoms, at a density  $n \equiv N/V \approx 10^{18} \text{ cm}^{-3}$ . We would now like to understand how and if this hot gas of Ps atoms will cool to a Bose condensed state before they disappear in roughly a few hundred nanoseconds (several times a triplet lifetime). If this indeed happens we would like to know how we would observe such a condensed state.

Collisions of the hot Ps atoms with the walls of the cavity and with each other leads to cooling equilibration and annihilation. While not much is known quantitatively it is possible to make some good semiquantitative estimates of all the important processes taking place in the volume and at the walls of our cavity. The dominant and simplest type of collision is the elastic collisions with the walls of the cell at a rate

$$\Gamma_w \approx v_{th}/d. \quad (5)$$

In our case the thermal velocity  $v_{th}$  of Ps at, say, 0.1 eV of energy is roughly  $10^7 \text{ cm/sec}$  while the important dimension  $d \approx 10^{-5} \text{ cm}$  so,  $\Gamma_w \approx 10^{-12} \text{ sec}^{-1}$ . With each elastic collision there is some probability that there will be an inelastic event in which a surface phonon is emitted. We can estimate the probability of such an inelastic event.

Like  $e^+$  a simple one-particle Ps potential describes much of the important physics of the interaction of a 0.1-eV Ps atom with the wall. At large distances from the surface Ps experiences an attractive  $1/z^3$  van der Waals potential.<sup>10,16</sup> At short, angstrom-type, distances the potential is cut off by some complicated many-body couplings to the electrons in the solid. At this point the simplistic one-particle potential becomes repulsive, rising to match the measured negative work function for Ps in the material. In addition, a complex part representing the onset of other channels, disassociation, annihilation, etc., turns on.<sup>10</sup> At roughly 0.1 eV of energy a Ps atom is not quantum mechanically reflected from the long-range

part of the potential nor are there many absorptive channels available.<sup>17</sup> To a good approximation the Ps atom with energy  $E$  acts very much like a fairly well-defined classical particle, simply bouncing specularly off the wall. Occasionally it strikes individual surface atoms and excites a phonon. For an inelastic event with momentum transfer  $k = \sqrt{2mE}/\hbar$  the probability of phonon emission occurring in this process is roughly given by

$$P \sim (k^2 \delta^2) R. \quad (6)$$

Here  $\delta^2$  is the mean surface displacement of a surface atom due to thermally excited phonon modes.<sup>18</sup> The quantity  $R$  is meant to phenomenologically include effects which have been left out of our discussion such as absorptive channels and quantum reflection. It is of order 1 for sub eV Ps atoms.

For most solids with lattice constant  $b$  and a Debye frequency  $\omega_D$ ,

$$\frac{\delta^2}{b^2} \approx 0.05 \left[ \frac{k_B T}{\hbar \omega_D} \right]. \quad (7)$$

At room temperature for Si ( $\omega_D = 1000 \text{ K}$ ) and for Ps atoms at 0.1 eV ( $k = 0.2 \text{ \AA}^{-1}$ ), Eqs. (6) and (7) ( $R \approx 0.5$ ) gives  $P \approx 0.01$ . The energy of a typical phonon  $\omega_\tau$  emitted in the scattering will be  $\hbar \omega_\tau \approx kv_s$  with  $v_s \approx 10^5$  a surface phonon velocity. For our 0.1-eV Ps,  $\omega_\tau \approx 20 \text{ K}$ . This means that roughly every  $10^{-10} \text{ sec}$  the energy drops by 20 K, i.e., our gas will cool to below room temperature in less than 10 nsec. This cooling rate is suitable for our proposed experiment, assuming no other collisions quench the ortho (triplet) Ps.

If the solid wall is metallic, i.e., there are unpaired spins, then exchange of the  $e^-$  in the triplet Ps with  $e^-$  in the metal occurs with high probability. Since the solid has electrons with spins in both directions the triplet is quenched. This is disastrous. If the wall is insulating, i.e., the electron spins are paired then exchange scattering will be energetically forbidden. The only allowed process is a weak relativistic effect which arises because of the time-dependent magnetic fields generated as electrons in the solid interact with the moving Ps atom. While it is very difficult to get a quantitative estimate of this rate we do know that the magnetic term in the Hamiltonian describing the coupling of the Ps atom to the solid is reduced by a factor  $v/c$  from the direct Coulomb term.<sup>19</sup> Here  $v$  is the velocity of the electrons (positrons) in the atom (Ps), i.e.,  $v/c \approx 3 \times 10^{-3}$ . The probability, for a spin-flip (sf) transition to occur would surely be smaller than  $v^2/c^2$ . Thus, we estimate  $\Gamma_w^{sf} \lesssim (v^2/c^2) \Gamma_w \approx 10^{-5} \Gamma_w$ , i.e., too small for us to worry about.

Finally and most importantly we must consider the effect of volume Ps-Ps scattering on the system. It is this scattering which equilibrates the gas and which as we shall see can lead, if we are not careful, to disastrous quenching of our long-lived triplet Ps. In the  $S$ -wave scattering approximation, as pointed out earlier,  $\sigma_s \approx 10^{-14} \text{ cm}^2$  and  $\sigma_t \approx 10^{-15} \text{ cm}^2$  so that the two-body rate  $\Gamma_{2B} = \sigma n v$  ( $n \approx 10^{18} \text{ cm}^{-3}$ ,  $v \approx 10^7 \text{ cm/sec}$ ) is  $10^{11} \text{ sec}^{-1}$  for singlet collisions and  $10^{10} \text{ sec}^{-1}$  for nonsinglet, i.e., rapid. In particular, it is rapid enough to enable the

Ps gas to come to equilibrium as it cools. Calculations show that for a system below  $T_c$  it only takes several interparticle scattering times to develop a Bose condensate.<sup>20</sup> The quantitative details of how the system does this will be interesting to observe.<sup>21</sup> However, the rapid two-body rate is dangerous in that electron exchange between two triplet Ps atoms with spins pointing in different directions can be appreciable if the Ps atoms are not prepared correctly.

The collision of low-energy Ps atoms may be classified (since the orbital angular momentum involved, to a very good approximation is always zero) by the total spin  $s$ , the parity  $P$ , and the charge conjugation parity  $C$ ,<sup>22</sup>  $|s, C, P\rangle$ . The charge conjugation parity  $C$  arises from the invariance of the total Hamiltonian to changing all  $e^+$  into  $e^-$ . For a Ps atom in an orbital  $S$  state  $C = (-1)^{s+1}$ . So, for example, the collision of two  $s=1$  Ps atoms has a  $C$  parity of plus one. Such a state cannot in a low-energy collision convert into one  $s=0$  and one  $s=1$  Ps atom.

Of the 16 possible spin states there are five having the quantum numbers  $|2, +, +\rangle$ , three with  $|1, +, -\rangle$ , three with  $|1, -, +\rangle$ , three with  $|1, --\rangle$ , and finally two with  $|0, ++\rangle$ . The 14 states with  $s=2, 1$  do not mix and hence collisions in these channels will not lead to conversion of triplets into singlets. For the two  $s=0$  states the situation is more interesting. These two states are, in terms of Ps quantum numbers,

$$|0, ++\rangle_t = \frac{1}{\sqrt{3}} \{ |1, -1\rangle |1, 1\rangle + |1, 1\rangle |1, -1\rangle - |1, 0\rangle |1, 0\rangle \}, \quad (8)$$

$$|0, +, +\rangle_s = |0, 0\rangle |0, 0\rangle, \quad (9)$$

and they do mix. In Eq. (8),  $|s, m\rangle$  specify the state of the Ps atom which, in turn, is specified by the spin of the  $e^-(\uparrow)$  or  $e^+(\uparrow)$ , for example,

$$|1, 1\rangle = |\uparrow\uparrow\rangle, \quad |1, 0\rangle = \frac{1}{\sqrt{2}} \{ |\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle \}, \text{ etc.}$$

The mixing between the two  $|0, ++\rangle$  states can be described phenomenologically by matrix elements of the Coulomb interaction  $V$ , i.e.,  $V_{tt}$ ,  $V_{st}$ , and  $V_{ss}$ . Since these matrix will not depend much on spin ( $V_{tt} \cong V_{ss} \cong V_{ts}$ ) it follows that the eigenvalues of  $\lambda_{\pm}$ , i.e.,

$$\lambda_{\pm} = \frac{1}{2} [V_{tt} + V_{ss}] \pm \sqrt{1/4(V_{tt} - V_{ss})^2 + V_{ts}^2}, \quad (10)$$

differ greatly. In the adiabatic approximation in which the eigenvalue  $\lambda_{\pm}$  is evaluated with two Ps atoms separated by a center-of-mass distance  $R$ ,  $\lambda_{\pm}(R)$  will be the effective potential. The binding energy  $E_+$  of the two atoms in the  $\lambda_+(R)$  potential corresponds to the binding energy of the  $\text{Ps}_2$  molecule. We do not know whether there is, in fact, a bound state corresponding to the  $\lambda_-(R)$  potential. If there is such a state the difference between its binding energy  $E_-$  and the ground state  $E_+$  will be of the same order of magnitude as  $E_+$  itself. In any case, if two triplet Ps atoms in the  $s=0$  state collide at low energies, they will be initially in superpositions of roughly equal parts of the  $|+\rangle$  and  $|-\rangle$  states. Since

$\lambda_+(R)$  is attractive and  $\lambda_-(R)$  is very different from  $\lambda_+(R)$ , the phases of the two states will be totally scrambled independent of the initial relative kinetic energy of the two atoms and the outgoing Ps atoms will have a probability of order  $\frac{1}{2}$  of being in singlet states.

These arguments suggest that we need to perform our experiment starting with a fully polarized  $e^+$  beam. Such beams are commonly available since all radioactive sources generate polarized  $e^+$ 's by a parity nonconserving process which leads to  $e^+$  polarized with their spins in the direction of their momentum. Slowing down inside a solid leaves them about 50% polarized.<sup>23</sup> Starting with polarized  $e^+$ , with spin up, means that for an unpolarized target we will initially have equal mixtures of  $|\uparrow\uparrow\rangle$  and  $|\uparrow\downarrow\rangle$ . Since  $|\uparrow\downarrow\rangle = (1/\sqrt{2})\{ |1, 0\rangle + |0, 0\rangle \}$  we will be left (singlet decays) with an equal mixture of  $|1, 1\rangle$  and  $|1, 0\rangle$ . Collisions between two  $|1, 0\rangle$  states leads, as we have discussed, to spin exchange and singlet Ps generation. Thus, this chain of reasoning suggests that our  $n = 10^{18} \text{ cm}^{-3}$  gas of hot polarized triplet Ps atoms should in a time of the order of 1 nsec completely self-polarize, to a pure  $|1, 1\rangle$  state. It will then happily cool through the Bose transition in tens of nanoseconds.

## V. OBSERVATION OF THE BOSE CONDENSATE

Because of the unique annihilation characteristics of Ps as discussed in Sec. II, we will be able to observe the momentum distribution<sup>24</sup> of Ps atoms directly, by converting a small but significant fraction of triplets to singlets. This can be accomplished by turning on a magnetic field to mix the singlet and triplet states. The Ps will slowly annihilate into two  $\gamma$  rays. The angle between the two  $\gamma$  rays as we have pointed out directly mirrors the momentum of the Ps atom. Thus, an angular correlation profile with enough resolution will indicate the presence of a zero-momentum condensate.

In order to obtain enough counts to measure the complete momentum and time history of the momentum distribution function of the Ps atoms we would use many 5-nsec pulses of positrons. Each  $e^+$  pulse obtained by, for example, emptying a trap where positrons have accumulated, triggers the electronics. One measures coincidences between detectors as a function of time. The angular resolution of the detectors determines the momentum (velocity) resolution. The best angular resolution currently attainable is a  $\Delta\Theta \cong 0.1 \text{ mrad}$ . This implies that we can measure the velocity distribution function with a resolution  $\Delta v \cong c\Delta\Theta \cong 3 \times 10^6 \text{ cm/sec}$  or about two times better than the velocity of a Ps atom at room temperature. With some improvement in  $\Delta\Theta$ , there should be unambiguous evidence of a qualitative peaking of the angular correlation profile as a function of time.

The annihilation of  $\gamma$  rays will do very little damage to the crystal since very little of the energy of these  $\gamma$  rays is deposited in the thin crystal. It is also true the total number of  $\gamma$  rays,  $10^{10}$ – $10^{11}$ , accumulated in an experiment is small compared to the number of atoms in the crystal. In addition, any damage to the crystal by the annihilation radiation is not at all crucial since the crystal acts primarily as a moderator. The Bose condensation occurs in the vacuum, i.e., the hole in the crystal. The Si

crystal must retain its hole and remain a good insulator, both very robust properties.

Observation of a Bose condensate while interesting in itself will not be the only goal of such experiments. We would, for the first time, begin to get direct experimental information about the dense Ps gas, i.e., Ps-Ps collisions and possible Ps molecule formation. Second, all of the problems connected with the time evolution of the condensate, for example, how such a condensate responds to changes in temperature and density, will be fascinating and possible to observe. It also may be possible to look

carefully near the transition for deviations from ideal behavior. Such nonideal behavior will become more apparent as we increase the density 1–2 orders of magnitude. In any event it seems likely that this exotic, but rather ideal, system will be a fascinating one to look at in the near future.

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