PHYSICAL REVIEW A VOLUME 14, NUMBER 4 OCTOBER 1976

Self-trapping of ortho-positronium in dense fluid helium*

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(Received 8 September 1975; revised manuscript received 14 June 1976)

Previous theoretical work by the author, on electron self-trapping, is extended to positronium in helium. Results are obtained for several values of the positronium-helium *s*-wave scattering length (a), in order to determine the validity of published analyses of data on the dependence of the ortho-positronium annihilation rate on the average density and temperature of its fluid helium host. It is concluded that the recently reported value of ${}^1Z_{\text{eff}} = 0.129 \pm 0.006$ is not "contaminated" by self-trapping effects or models, while the value reported for $a = 0.77 \pm 0.04$ Å results from an invalid analysis. It is suggested that a value for a in the neighborhood of 0.8 Å may be reasonably inferred from available experimental data and the calculations presented here, but that the precision of this determination is not very high.

INTRODUCTION

There exists a body of experimental data concerning the dependence of the annihilation rate of ortho-positronium (o-Ps) on the average density and temperature of its fluid-helium host. The qualitative interpretation of the data is well known and consists in the implication that for low helium density and/or high temperatures (ρ and T as independent variables) positronium is predominantly "free" and able to sample the average helium density; in contrast, for high density and/or low temperatures positronium is stably self-trapped in a region in which the local helium density is low. Stable self-trapping arises due to Ps-helium repulsion and is formally identical to electron self-trapping in fluid helium.

Quantitative analysis of the experimental data requires knowledge of (i) the annihilation rate to be expected if the o-Ps samples a helium density ρ and of (ii) the Ps-helium interaction which in principle allows a determination of the possible states of the Ps–fluid-helium system and their occupation at a given temperature.

Recent experiments and analyses of those data,[1] based on a model of the occupied system states at a given helium density and temperature, have yielded values of three important parameters: λ_{vac}, the annihilation rate of o-Ps in vacuum; ${}^1Z_{\text{eff}}$, the number of electrons per helium gas atom as seen in the singlet state relative to the positron spin of the o-Ps atom; and a, the Ps-helium s-wave scattering length. The results were compared with alternative determinations and with theoretical calculations. However, certain checks on the internal consistency among the parameters obtained and the method of determination were not considered; it is the purpose of the present work to present calculations, based on the author's previous work,[2] which yield a context in which self-consistency checks may be examined. The reader will be often referred to Refs. 1 and 2 for detailed arguments which will not be repeated here. The conclusion reached is that although the parameters obtained in analysis[1] of the data are quite reasonable, the model used to obtain the Ps-helium scattering length is not valid.

GENERAL CONSIDERATIONS

The dependence of the annihilation rate λ of o-Ps on the average helium density ρ and temperature exhibits the following experimental features: (i) At low helium densities, λ is temperature independent and increases linearly with the average helium density. (ii) As the helium density is further increased, λ departs from the linear behavior, at lower densities for lower temperatures, and tends towards the vacuum annihilation rate. The trend reversal is rather abrupt, with helium density, for low temperatures, but becomes increasingly gentle as the temperature is increased.

The qualitative explanation of these trends can be based on theoretical calculation.[2] At low helium densities the stable state for Ps in fluid helium is the free state, sampling an essentially uniform helium density. However, as the helium density is increased, a self-trapped state eventually becomes the stable state of the system. The change in the stable state, from free to self-trapped, indeed takes place at lower densities for lower temperatures, and the calculated energy difference between the stable self-trapped state and the lowest-energy free state (consistent with a uniform helium density) varies more rapidly, with density, for lower temperatures. The calculations are identical to those which have been reported for electrons in helium[2]; the only changes consist in using the Ps mass (twice that for electrons) and the s-wave scattering length appropriate to Ps-helium. It should be noted that in Ref. 2 the potential energies were related to helium density by using the s-wave scattering length in the Wigner-Seitz (WS) formalism,

$$V_{WS}(n) = \hbar^2 k_0^2/2m; \quad \tan k_0(r_s - a) = k_0 r_s , \tag{1}$$

where m is the particle mass, r_s the radius of the sphere containing one helium atom if the number density is n, and a the s-wave helium-particle scattering length. The low-density limits of these expressions yield the optical potential relationship used in Ref. 1,

$$V_0 = (\hbar^2/2m)4\pi n a . \tag{2}$$

However, for the densities of interest in this work V_{WS} is between 35% and 100% larger than V_0 (for given n and a), so that the effect of using V_0 or V_{WS} must be examined.

As previously mentioned, for quantitative analyses of o-Ps annihilation rates in helium, as functions of density and temperature, there are three parameters of interest, λ_{vac}, ${}^1Z_{eff}$, and a. In the free state,[1]

$$\lambda = \lambda_{vac} + (4\pi r_0^2 c/M)\, {}^1Z_{eff}\, \rho ; \tag{3}$$

thus a linear dependence of λ on ρ is expected, and obtained, with zero density intercept equal to λ_{vac}. Since λ_{vac} is well known (0.72×10^7 sec^{-1}), the decay data has been used to obtain ${}^1Z_{eff}$. To assure that this determination is not "contaminated" by self-trapping effects, one should check that the data used lies in the region in which the s-wave scattering length predicts the absence of stable self-trapped states (this may be of special importance at high temperatures, when the effects due to self-trapping are slowly varying with density).

It is expected that stable self-trapping will be indicated by data which yield λ smaller than that given by Eq. (3), and that such data may be analyzed to obtain a value for the s-wave scattering length. This is not a simple procedure, since first one must decide how to relate a to the density (using V_0, V_{WS}, or some other procedure) and then, in principle, one should take arbitrary configurations of the helium atoms, calculate the states for Ps, calculate the annihilation rate in such states, and perform thermal averages over all important configurations. The model which was used in Ref. 1 assumes that only one state has appreciable occupancy and that that state is one described by Ps in the lowest state consistent with an otherwise empty cavity in the fluid, with the radius of the cavity determined by equilibrium considerations. This model must be carefully examined to substantiate its appropriateness.

RESULTS

There exist various theoretical estimates of the Ps-helium s-wave scattering length, that of Fraser,[3] 1.11 Å, Fraser and Kraidy,[4] 0.91 Å, and most recently of Drachman and Houston,[5] 0.73 Å. Using the methods I presented previously[2] calculations were carried out for the locus, in density-temperature space, of the conditions needed to change the stability of the Ps-helium system from the free state to a self-trapped one. The results are reported in Fig. 1; the solid lines are obtained using the indicated value of a in the Wigner-Seitz approximation, Eq. (1), to relate a and the helium density. Results are also shown (dot-dash lines) for two values of the scattering length and the optical potential relationship of Eq. (2). In order to indicate the variation of the relative binding energy of the stable self-trapped state to the free state as density and temperature change, dashed curves are also shown (WS approximation, three values of a) for conditions in which the stable self-trapped state is $2kT$ below the energy of the free state. Finally, symbols are used to denote the conditions in which λ has been measured by various experimental groups.[1,6-8] As shown in the previous work,[2] the empty-cavity model as a description of the stable self-trapped state is asymptotically correct in the limit of very strong binding relative to the free state, but it is quite inadequate when this relative binding energy is less than approximately $6kT$. The assumption that the stable state is the only one with appreciable occupancy in thermal

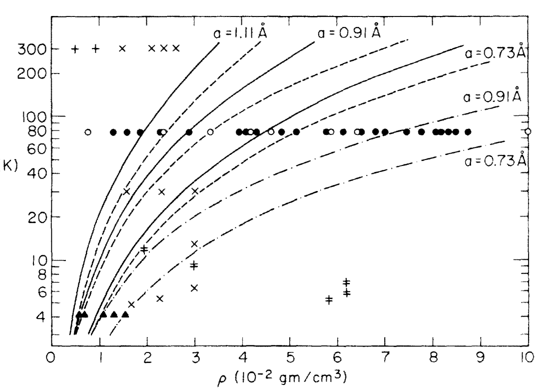

FIG. 1. Solid lines indicate calculated conditions necessary to change the stable state of the Ps-helium system from free to self-trapped, for the three values of the s-wave scattering length noted, in the Wigner-Seitz approximation. Dot-dash lines, same as above but in the optical-potential approximation. Dashed lines, conditions in which the stable self-trapped state is $2kT$ below the free state, WS approximation, three scattering lengths. Symbols indicate conditions at which λ has been measured experimentally: ×, Ref. 1; +, Ref. 6; ●, ▲, Ref. 7; ○, ‡, Ref. 8.

equilibrium is clearly wrong when the relative binding energy is of order kT, since under those conditions even the free state may have appreciable occupancy as well as many self-trapped configurations other than the stable one (see, for example, Fig. 2 of Ref. 2); relative occupancy also depends on relative entropies, and this facet has not yet been thoroughly explored.

DISCUSSION

In view of the results presented in Fig. 1, let us examine the experimental data and conclusions drawn from its analyses. In the range indicated, for any value of the s-wave scattering length and either method (V_0 or V_{WS}) of relating it to density, the room-temperature measurements[1,6] are in the region where the free state is the stable state. Thus we conclude that the derived value for ${}^1Z_{eff}$ has only statistical errors and is model independent. Figures 2 and 3 summarize the results of experimental measurements of λ in helium, from various sources (the symbols are the same as those those in Fig. 1). As can be seen in Fig. 2, the room-temperature data points lie on a good straight line, as has been noted previously,[1,6] and are the source of ${}^1Z_{eff}$ in those reports. The two lower density points at[1] 30 K also lie on that line, but not the highest density point at 30 K. The data in Fig. 3, closed[7] and open[8] circles (with clearly larger error bars) at 77 K and triangles[7] for 4.2 K, along with the straight line[1] from Fig. 2, seem qualitatively as expected but quantitatively anomalous. The closed symbols are above the straight line for low densities, and the 77-K data[7] are systematically above that of Ref. 8. The indications of self-trapping are evident at high densities. Given the anomalies of Fig. 3 there are relatively few points from which a scattering length can be inferred by using Figs. 1 and 2. Also, Fig. 1 and Ref. 2 may be used to examine the validity of the analyses of the data points at 4.8, 5.4, and 6.4 K carried out in Ref. 1; this examination follows.

In Ref. 1 the decay rate observed at 4.8, 5.4, and 6.4 K was analyzed using the single-occupancy, empty-cavity model, in the V_0 approximation, to obtain $a = 0.77 \pm 0.04$. This analysis, when coupled with the present results, quoted in Fig. 1, is unfortunately invalid. If the optical model (V_0) were appropriate and $a = 0.77$ Å then, according to Fig. 1, the single-occupancy, empty-cavity model would not be appropriate for these three points because their binding energy would not be sufficiently large, most notably at 4.8 K and the density in question the stable state is barely bound relative to the free state. These remarks become more evident if the total energy of the states used in the

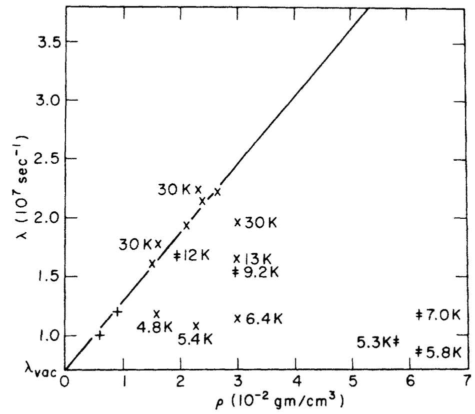

FIG. 2. Measurements of the o-Ps annihilation rate λ. Symbols are the same as in Fig. 1; the measurements are at the temperature noted or for 300 K when unlabeled. The straight-line fit (Ref. 1) through the 300-K data represents annihilation from the free state, while decay rates below the line are interpreted as evidence for self-trapping due to the accompanying lowered local density.

calculation[1] is estimated; in all three cases the pressure-volume work needed to construct the empty cavity is somewhat larger than the energy gained in binding the positronium.[9] Reinterpreting the calculation of Ref. 1 by using V_{WS} instead of V_0 changes the result to $a = 0.5$ Å, but the objections to this reinterpretation are identical to those of the previous one. Furthermore, according to Fig. 1 either of the interpretations would have to come into conflict with the experimental evidence of self-trapping at 30 K, $\rho = 3 \times 10^{-2}$ g/cm^3, and that at 4.2 K (Fig. 3).[10] From the experimental data as quoted in Figs. 2 and 3 and our results as shown in Fig. 1 one can come to the conclusion that if the WS model is appropriate then indeed $a \sim 0.8$ Å, but the limits of ± 0.1 Å are not unreasonable and may

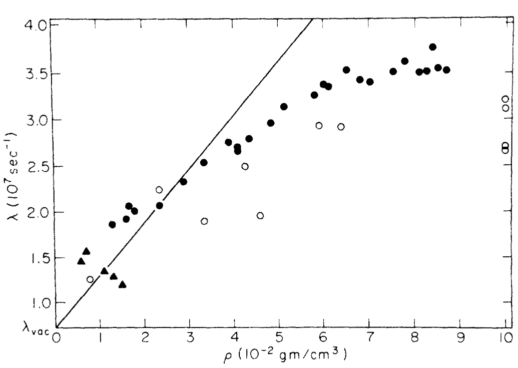

FIG. 3. Measurements of the o-Ps annihilation rate λ. Symbols as in Fig. 1. Open and closed circles correspond to 77 K, triangles to 4.2 K. The straight line is that of Fig. 2.

be rather too narrow, since the data available in the transition region, where the stability is changing from free to self-trapped Ps, is quite limited.

A detailed analysis of the data in the style used by Ref. 1 is really quite difficult even in concept, since thermal fluctuations, that is, occupancy of states other than the stable one, are difficult to take into account, and they give rise to uncertainties in the precision with which the parameter a may be determined. It is hoped that more data will be obtained in the region of ρ-T space for which the stability change to self-trapped Ps is expected. Similar data are starting to become available for materials other than helium,[11] as are angular correlation data from the $2-\gamma$ decay of para-positronium.[12]

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*Work supported by the Materials Research Center, University of North Carolina, under National Science Foundation Grant No. DMR 7203024.

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[9]In the notation of Ref. 1, $\frac{4}{3}\pi R^3 p + E_z - V_0$ may be found for the three points in question and ranges from approximately $+1kT$ to $+25kT$, where the zero of energy is the free state. Clearly, arbitrary states of the Ps-helium system must have higher energy than that of the stable state, which may be estimated from Fig. 1. Relative entropy will also bear on occupation; this facet thas not yet been thoroughly explored.

[10]With either interpretation, these points would lie above the appropriate curve in Fig. 1 and thus would be expected to correspond to free-state stability, in conflict with their measured λ.

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[12]Measurements at 77 K in helium by C. V. Briscoe and A. T. Stewart (private communication).