

A critical assessment of the bubble model for positronium annihilation in liquids

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1999 J. Phys.: Condens. Matter 11 1463

(<http://iopscience.iop.org/0953-8984/11/6/010>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.214

The article was downloaded on 15/05/2010 at 06:58

Please note that [terms and conditions apply](#).

A critical assessment of the bubble model for positronium annihilation in liquids

Debarshi Gangopadhyay[†], Bichitra Nandi Ganguly[†], Tapas Mukherjee[‡] and Binayak Dutta-Roy[†]

[†] Institute of Nuclear Physics, 1/AF Bidhannagar, Calcutta 700 064, India

[‡] Physics Department, Bhairab Ganguly College, Calcutta 700 056, India

Received 21 October 1998

Abstract. The bubble model conventionally used to fit the observed characteristics of the pick-off component of ortho-positronium decay in liquids is subjected, in the present study, to a critical assessment. It is demonstrated that in its usual form (namely that of a bubble with a sharp boundary) the model is untenable, when confronted conjointly with experimental data on the lifetime and angular correlation of the decay gammas. A modified version of the model that is relatively free from such shortcomings is presented.

1. Introduction

The bubble model was proposed by Ferrell [1] to account for the observed increase in the lifetime of ortho-positronium (o-Ps) for the pick-off [2] process in liquid helium. This is a process whereby the positron in o-Ps ‘senses’ electrons with opposite spin in surrounding atoms and annihilates through the two-photon mode. The underlying physical mechanism for the formation of the bubble around the positronium was conceived to be through the repulsive interaction due to electron exchange between o-Ps and atoms of the surrounding liquid leading to self-trapping, somewhat akin to what is thought to occur in the polaron theory for electrons in condensed matter. In the initial version of the model, for the sake of simplicity, the self-trapping potential confining the positronium in the bubble was taken to be an infinitely repulsive spherical well of radius R (to fix the notation). The resultant quantal zero-point energy of the positronium of mass $2m$ (m being the electron mass) is accordingly given by

$$E = \frac{\pi^2 \hbar^2}{4mR^2}$$

which in turn exerts a pressure of $-\partial E/\partial R$, to be balanced by the forces of compression introduced through the rather crude notion of surface tension (σ) of the fluid. Thus the total energy $E_{tot} = E + 4\pi R^2\sigma$ (the second term representing the contractile surface energy) is minimized. We shall refer to this relation—namely, $\partial E_{tot}/\partial R = 0$ —as the balance condition. Therefore, given the surface tension of the liquid, the balance condition enables one to determine the radius R of the bubble and, thus, within the confines of the model, the wavefunction

$$\psi_{Ps} = \frac{1}{\sqrt{2\pi R}} \sin\left(\frac{\pi r}{R}\right)$$

of the positronium inside the cavity, r being the radial distance of the centre of mass of Ps from the centre of the bubble. Ferrell went on to assume that, inside the bubble, helium was present as a saturated vapour, providing the electrons for pick-off annihilation.

A variant of this scheme was put forward by Tao [3] and also by Eldrup *et al* [4] employing again the infinite spherical well, but describing the pick-off electrons as forming a layer of thickness ΔR uniformly coating the inner surface of the bubble. These authors next assumed that the annihilation rate of o-Ps inside the electron layer is 2 ns^{-1} (the spin-averaged annihilation rate of p-Ps and o-Ps) and accordingly the pick-off rate is then obtained as

$$\lambda_p = 2 \left\{ \frac{\Delta R}{R} - \frac{1}{2\pi} \sin \left(2\pi \frac{\Delta R}{R} \right) \right\} \text{ns}^{-1} \quad (1)$$

where the expression in braces is simply the probability for the Ps atom to be inside the electron layer, to wit,

$$\int_{R-\Delta R}^R |\psi_{\text{Ps}}(r)|^2 d^3r.$$

This model has gained considerable popularity not so much for the soundness of the underlying ideas but rather because of the simplicity of the final expressions. Indeed algebraic relationships for lifetime and angular correlation have been found to roughly fit the general trend of mainly the lifetime data.

Roellig [5] improved upon Ferrell's model by emphasizing that the notion of an infinitely repulsive trapping potential was physically most unsatisfactory, and that it would be far more reasonable to posit a well of finite height in its place and to ascribe the pick-off annihilation of the positron in the positronium to its quantum mechanical leakage outside the bubble and into the surrounding liquid. The Schrödinger equation is readily solved to obtain the wavefunction for the ground-state centre-of-mass motion of the positronium in a spherical well of height U_0 and range R , which takes the forms $(\sin kr)/r$ and $[\exp(-\kappa r)]/r$ inside and outside the well respectively. Here

$$k = \sqrt{\frac{4mE}{\hbar^2}} \quad \text{and} \quad \kappa = \sqrt{\frac{4m(U_0 - E)}{\hbar^2}}$$

where E is the energy eigenvalue determined by the matching condition $\kappa = -k \cot(kR)$ for the wavefunction at the well boundary. This equation has to be solved numerically. The balance condition

$$\frac{\partial}{\partial R} (E + 4\pi R^2 \sigma) = 0 \quad (2)$$

provides, given the surface tension σ of the liquid, a relationship between U_0 and R . Thus, but for one independent parameter, the model stands determined. Therefore, with two pieces of experimental information (lifetime and angular correlation data) the model can be tested.

Since pick-off annihilation involves the positron in the positronium and the electrons in the surrounding medium, the rate for the process is governed by the product of the probability for finding the positronium in the liquid and the density of available electrons in the surrounding liquid. This factor is given by the overlap integral

$$\mathcal{P} = 4\pi \int_R^\infty |\psi_{\text{Ps}}(r)|^2 r^2 dr \quad (3)$$

where the integration is over the region containing the liquid (namely, outside the bubble). The pick-off rate is thus obtained as

$$\lambda_{\text{pick-off}} \equiv \lambda_p = 4\pi r_0^2 c \rho Z_{ef} \mathcal{P} \quad (4)$$

where $r_0 = e^2/mc^2$ is the classical electron radius, c is the velocity of light, ρ is the number density of molecules in the liquid, Z_{eff} is the effective number of electrons available per molecule for pick-off annihilation by the positron in o-Ps.

Apart from the influence of the bubble on the annihilation rate (and hence the lifetime $\tau_p = 1/\lambda_p$), the bodily motion of the positronium confined to move in the bubble implies, by virtue of momentum conservation, a nontrivial angular correlation (namely, not back to back) for the two photons emitted through the annihilation of para-positronium (p-Ps) in the bubble. In order to determine the angular correlation curve $N(\theta)$ and the experimentally quoted quantity $\theta_{1/2}$ (which is the full width at half-maximum (fwhm), given by $N(\frac{1}{2}\theta_{1/2}) = \frac{1}{2}N(0)$), all one needs is the momentum distribution of the positronium inside the bubble, which is given by the square modulus of the Fourier transform of the wavefunction, namely

$$P(p) = 4\pi p^2 |\tilde{\psi}_{Ps}(p)|^2 \quad (5a)$$

with

$$\tilde{\psi}_{Ps}(p) = \left(\frac{1}{2\pi\hbar}\right)^{3/2} \int \psi_{Ps}(r) \exp\left(-i\frac{\vec{p}\cdot\vec{r}}{\hbar}\right) d^3\vec{r} \quad (5b)$$

and the angular distribution curve is given by

$$N(\theta) = \int_{p=mc\theta}^{\infty} \frac{1}{p'} P(p') dp' \quad (5c)$$

from which $\theta_{1/2}$ is readily calculated.

Having thus set forth the basics of the model, we go on in the next section to point out its inadequacies through a critical analysis. This is followed by a proposed modification of the model and we close with a section devoted to concluding remarks.

2. Inadequacy of the existing bubble model

Using the formalism presented above, there have been [6–8] extensive calculations using the finite-well potential for a variety of liquids and comparison has been made with experimental data. While there is reasonable agreement, at first sight, between the model and observations, nevertheless, as was pointed out [11, 12] by the present authors, there exists a systematic discrepancy, in that if one determines the parameters of the model (namely R and U_0) using the balance condition (equation (2)) and a fit to the lifetime data, and if one uses these values to calculate the fwhm $\theta_{1/2}$ of the angular correlation of the decay gammas, then $\theta_{1/2}$ is systematically underestimated as compared to experiment. Even though these deviations are at most of the order of 20%, their signs are the same in all cases, as can be seen from table 1. The experimental data on positronium annihilation, as well as values of the surface tension, density and Z_{eff} , have been taken from the compilation given by Nakanishi and Jean [8]. We have shown results for only some of these liquids in the main body of the paper, the rest being relegated to the appendix in order to avoid a break in the text caused by a long table.

To further expose the problems involving the bubble model with sharp boundaries, another strategy is found to be very revealing. The model parameters are determined on the one hand by fitting the lifetime data and on the other by fitting the angular correlation, and then a comparison of the two sets is made. We observe from table 1 that the depth of the potential U_0 obtained by using the former protocol can be as much as a factor of two smaller than that arrived at through the latter methodology, while the range parameter (R) could be 10 to 15% smaller. This inability to provide a consistent set of parameters to fit the two measurements clearly underlines the need to modify the model, even more convincingly than the indications referred to in the previous paragraph. An insensitivity is discernible from table 1 in the energy value

Table 1. The bubble model with sharp boundaries. The first four columns for each liquid represent entries with potential parameters fixed by fitting the observed λ_p as input. The last three columns pertain to results using the observed $\theta_{1/2}$ as input. The entries under $\delta\theta_{1/2}$ are percentage deviations of the predicted values of $\theta_{1/2}$ from the observed values with λ_p as input.

Liquid	R (Å)	E (eV)	U_0 (eV)	$\delta\theta_{1/2}$ (%)	R (Å)	E (eV)	U_0 (eV)
<i>n</i> -pentane	4.85	0.424	0.747	-17%	5.36	0.435	1.44
<i>n</i> -hexane	4.66	0.456	0.790	-15%	5.08	0.465	1.33
<i>n</i> -heptane	4.55	0.480	0.838	-15%	4.97	0.490	1.44
<i>n</i> -decane	4.33	0.523	0.894	-17%	4.81	0.423	1.72
Iso-octane	4.66	0.464	0.835	-18%	5.53	0.493	4.78
Benzene	4.11	0.575	0.962	-17%	4.53	0.586	1.69
Water	3.13	0.922	1.36	-19%	3.47	0.928	2.14

(E) of the positronium in the cavity as determined from the two methods of fitting, which, while giving potentials with widely differing depths (U_0), nevertheless yield similar values for the energy E . This is because of the fact that as R increases the kinetic energy (due to the zero-point motion dictated by the uncertainty principle) decreases. Correspondingly, one sees that the potential energy in the latter case is greater. Thus the total energies in the two situations are very close to each other.

Another serious lacuna as regards the bubble model with sharp boundaries becomes apparent when one calculates the lifetime $\tau_p = 1/\lambda_p$ based on parameters determined by fitting $\theta_{1/2}$. Thus for example in the case of pentane one predicts $\tau_p = 13.9$ ns, while the experimental value is only 4.25 ns. This discrepancy of a factor of more than three clearly demonstrates the unsatisfactory nature of the model. It is also to be remarked that λ_p is extremely sensitive to the parameters of the model as compared to $\theta_{1/2}$. This is because $\theta_{1/2}$ is determined essentially by the radius R of the bubble, while λ_p depends on the overlap integral involving the square modulus of the positronium wavefunction on the one hand and the density distribution of the available electrons of the medium on the other.

A further indication of the inadequacy of the bubble model with sharp boundaries is revealed when one examines the data pertaining to pick-off annihilation in ordinary water (H_2O) and heavy water (D_2O). The best fits to the angular correlation data in the two cases are given by potential depths (U_0) which differ from each other by about 20%. This, however, should not be the case, as the depth of the potential is essentially provided by the exchange interaction between the electron in the positronium and the electrons in the surrounding molecules of the liquid. But the wavefunctions of electrons in the molecules of H_2O and D_2O must be essentially identical. Therefore, one expects the values of U_0 in the two cases to be almost the same or close (within experimental errors). Thus, when looked upon critically, the bubble model with sharp boundaries appears to be sorely in need of modification.

3. A modified version of the bubble model

Apart from the shortcomings of the bubble model with sharp boundaries revealed through the detailed comparison with data as discussed above, it should also be emphasized that even from a purely theoretical point of view, despite the popularity of the bubble model, the picture of a sharp boundary is at variance with the general notion of a liquid–vapour interface [11], and even more so for such microscopic dimensions as can be seen in the numerical study reported by Rusanov and Brodskaya [12] in their molecular dynamical simulation of a small liquid

drop. Indeed, critical remarks to that effect can also be found in the positronium literature. To quote from a paper by Roellig [5] addressing the pick-off process in liquid helium: ‘the agreement between the calculated values and the experimental data is surprising, for the bubble may not possess a definite radius, and there very well may be a transition region between the cavity which has a helium atom density of zero and the bulk density of the liquid’. It is, therefore, surprising that despite recognition of this shortcoming there have been no efforts towards rectification, as far as we are aware, except for some recent attempts by the present authors [9, 10].

Here it may also be appropriate, in support of our present attempts, to recall some remarks made in a paper [3] by Tao: ‘which type of more complicated potential well is the best or better one in the calculation of the pick-off rate of o-Ps in liquids . . .?’

Recognizing that the cavity appearing around the positronium should have a density profile $\rho(r)$ for the surrounding liquid more in the nature of a smooth (exponentially varying) liquid–vapour interface rather than having a sharp and abrupt drop from zero to the bulk density (ρ_0), we adopt a natural choice:

$$\rho(r) = \rho_0[1 - \exp(-r/R)] \quad (6)$$

with the parameter R now characterizing the ‘size’ of the cavity. Furthermore, since the repulsive exchange interaction between the Ps atom and the host molecules is short ranged, it is not too unreasonable (in the spirit of such hybrid ‘micro–macro’ models) to take the self-trapping potential to have the same radial dependence as that of the density, to wit,

$$U(r) = U_0[1 - \exp(-r/R)]. \quad (7)$$

The normalized ground-state wavefunction is readily found in terms of standard (Bessel) functions to be

$$\psi_{\text{Ps}}(r) = \left(\frac{k}{2\pi s}\right)^{1/2} \left(\frac{1}{r}\right) J_{2kR}(2bR e^{-r/2R}) \quad (8a)$$

where the effective asymptotic wavenumber appearing above is given by

$$k = \sqrt{\frac{4m}{\hbar^2}(U_0 - E)} \quad \text{and} \quad b = \sqrt{\frac{4m}{\hbar^2}U_0}.$$

The quantity s occurring in the wavefunction serves to normalize it and is given by the series

$$s = \sum_{n=0}^{\infty} \{[J_{2kR+n+1}(2bR)]^2 + [J_{2kR+n}(2bR)]^2\} \quad (8b)$$

where $J_\nu(z)$ is the Bessel function of order ν and argument z . The ground-state energy E is obtained from the eigenvalue condition

$$J_{2kR}(2bR) = 0. \quad (8c)$$

The overlap integral, needed for the lifetime determination—see equation (4)—is readily evaluated using standard mathematical tables [13] to yield

$$\mathcal{P} = 1 - \frac{2kR}{s} [J_{2kR+1}(2bR)]^2. \quad (9)$$

On the other hand, the Fourier transform of the wavefunction necessary for the calculation of the angular correlation (see equations (5a), (5b) and (5c)) is also analytically available in the form of a rapidly convergent series.

With this modified version of the bubble model (with smoothed boundaries), we have followed the same procedure as was adopted in the previous case and the results are displayed in table 2, following the same scheme as was used in table 1.

Table 2. The modified bubble model with smooth boundaries.

Liquid	R (Å)	E (eV)	U_0 (eV)	$\delta\theta_{1/2}$ (%)	R (Å)	E (eV)	U_0 (eV)
<i>n</i> -pentane	5.29	1.22	2.23	0.3%	5.29	1.22	2.22
<i>n</i> -hexane	5.10	1.32	2.39	3.0%	5.19	1.33	2.39
<i>n</i> -heptane	4.97	1.38	2.52	3.0%	5.07	1.40	2.53
<i>n</i> -decane	4.76	1.51	2.74	0.9%	4.82	1.51	2.75
Iso-octane	5.06	1.34	2.44	-2.0%	5.09	1.32	2.48
Benzene	4.53	1.66	3.00	2.0%	4.64	1.68	3.03
Water	3.55	2.66	4.68	4.5%	3.55	2.69	4.57

In the first place one observes that the deviation $\delta\theta_{1/2}$ has under the modification been reduced from as much as 20% to less than five per cent. What is even more dramatic is the fact that the values of the model parameters U_0 and R , the height and range of the potential respectively, determined from the lifetime data and the same obtained by fitting the results of the angular correlation experiments are now almost identical, in stark contrast to what was the case for the bubble with sharp boundaries. Furthermore, for the sake of comparison it may be mentioned that fitting the model to the observed value of $\theta_{1/2}$ for pentane leads to a prediction of 3.40 ns for the pick-off lifetime, which is 20% off as compared to the factor-of-three discrepancy with the sharp-boundary model. Even this discrepancy is because of the extremely sensitive dependence involved, as described earlier.

Furthermore, using the modified model, the depths of the potential (U_0) for H₂O and D₂O differ from each other by only about three per cent in contrast to the twenty per cent discrepancy obtained when we used the original version.

Another interesting observation that deserves more attention is the relationship between the depth of the potential and the number density of available electrons, as is evident from a perusal of table 3.

Table 3. The depth of the potential and number density of the electrons: the value of $(\rho Z_{eff}/U_0) \times 10^3$ for different liquids.

<i>n</i> -hexane	Iso-octane	Propanol	Benzene	Toluene	Mesitylene	Water
30	31	31	28	28	30	24

The contention that the depth of the potential should be proportional to the number density of available electrons in the surrounding molecules is physically very plausible, as the self-trapping of the positronium is supposed to arise from the electron exchange interaction between it and the atoms of the medium. That this relationship is approximately obeyed is rather satisfying and indeed suggests that U_0 may serve [14] as a fundamental important property of a given liquid and can be related to surface tension at a given temperature similar to the parachor [15].

A point of criticism of this modified model could be that the solution involves Bessel functions which, though standard, forces one to take recourse to numerical calculations. One could argue that the Eldrup model despite its shortcomings provides convenient relationships between the parameters of the model and observables such as $\theta_{1/2}$ and λ_p . In order to compensate for this 'drawback' we provide below some fits to simple algebraic forms that we have made for our modified version of the model:

The radius parameter (R) of the exponential potential is approximately related to the

angular correlation ($\theta_{1/2}$) through

$$R = \frac{13.5}{\theta_{1/2}} - 0.64. \quad (10)$$

Again the energy (E) of the positronium in the bubble can be approximately related to the radius parameter (R) and to $\theta_{1/2}$ through

$$E = 0.24\theta_{1/2}^2 \quad (11a)$$

$$E = \frac{43.6}{(R + 0.64)^2}. \quad (11b)$$

In all of these formulae, E is to be expressed in eV, R in Å and $\theta_{1/2}$ in milliradians.

The pick-off lifetime, $\tau_p = 1/\lambda_p$ in ns, is approximately related to the radius parameter through

$$R = 2.15 + 0.713\tau_p. \quad (12)$$

4. Concluding remarks and future outlook

We have argued that the bubble model for pick-off annihilation of o-Ps in liquids, in the form that is used, is rather unsatisfactory. In the first place, if the parameters of the model are chosen to fit the lifetime data, the predicted fwhm for the angular correlation shows systematic departures from their observed values. This lacuna of the model can be brought into sharper relief through the fact that the depth of the potential that confines the positronium in the bubble when determined from the lifetime data departs by as much as a factor of two from its value fixed from the observed angular correlation.

We ascribe these serious contradictions between the model and observation to the rather unphysical assumption of a sharp bubble boundary. Accordingly, we have rectified this feature through a modified model and have then proceeded to show that this is largely free from these inconsistencies.

We have gone on to present some simple approximate algebraic relationships between the parameters of the model and experimentally observed quantities.

Apart from the relevance of the foregoing discussions in providing a model for positronium annihilation in liquids and thus delineating a setting for the understanding of the physical basis of these observations, these considerations have important bearings on some issues in positronium chemistry. Indeed Goldanskii and Shantarovich [16] have pointed out that when suitable acceptor (Ac) molecules are present in the solvent, the positronium enclosed in a bubble could form a Ps–Ac complex. There have been some attempts [17] to develop a model for the dynamics of such a process and to study the stability of such a complex. This, however, has been done within the ambit of the bubble model with sharp boundaries. Needless to say, in view of the results set forth in this paper, these investigations cry out to be revised.

Another aspect of the problem that needs further scrutiny stems from the fact that the largest discrepancies for the bubble model (and even its modified incarnation) occur for polar liquids (with high surface tension) such as water. Indeed some authors have expressed the view that this very description may be inappropriate in such situations. This may indicate a further direction of development of the underlying model, involving the notion of curvature-dependent surface tension [18], which was implemented in a way by Byakov and Petuchov [19]. This question has also been addressed by the present authors elsewhere [20]. Of course, there are also issues such as the need to have a hybrid model which combines the bubble model with the free-volume model [21] for positronium annihilation in liquids. And, of course, a

point of criticism of the bubble model is that it injects into a problem, basically microscopic in character, notions such as surface tension which belong to the category of macroscopic ideas. Furthermore, the estimation of Z_{eff} is indeed questionable, and the contributions of inductive effects due to polarizability and electric dipole moments are not properly reflected in such models. However, overcoming these limitations would lead to a situation where the entire simplicity of the model would be impaired. These and other problems are being looked into by our group and lie outside the strict purview of the present study.

Appendix

Table A1. The bubble model with (top half) sharp boundaries and (bottom half) smooth boundaries.

Liquid	R (Å)	E (eV)	U_0 (eV)	$\delta\theta_{1/2}$ (%)	R (Å)	E (eV)	U_0 (eV)
Methanol	4.45	0.510	0.921	-9.5	4.69	0.515	1.23
Ethanol	4.44	0.508	0.903	-13	4.77	0.515	1.37
Propanol	4.36	0.523	0.920	-13.4	4.72	0.532	1.46
Butanol	4.30	0.541	0.954	-13.9	4.66	0.550	1.56
Octanol	4.18	0.564	0.967	-15.4	4.63	0.577	1.84
Cyclohexane	4.74	1.55	2.85	-4	4.80	1.58	2.83
Methylcyclohexane	4.36	0.523	0.915	-16.9	4.83	0.536	1.78
Toluene	4.14	0.572	0.972	-17	4.58	0.585	1.81
Ethyl benzene	4.08	0.581	0.957	-13.3	4.39	0.587	1.40
o-xylene	4.06	0.591	0.992	-15	4.51	0.577	1.54
m-xylene	4.11	0.578	0.980	-14	4.45	0.586	1.50
p-xylene	4.13	0.573	0.972	-14	4.48	0.582	1.52
Mesitylene	4.18	0.569	0.996	-13	4.51	0.577	1.54
Hexafluorobenzene	4.69	0.521	1.283	-3	4.77	0.524	1.47
Diethyl ether	4.75	0.442	0.780	-16.5	5.25	0.453	1.50
Acetone	4.29	0.531	0.894	-16.5	4.72	0.541	1.57
Carbon disulphide	3.60	0.520	0.788	-26	4.14	0.611	1.19
Heavy water	3.17	0.913	1.379	-18	3.38	0.915	1.78
Methanol	4.84	1.47	2.69	8.6	4.82	1.49	2.58
Ethanol	4.86	1.46	2.69	5.9	4.78	1.47	2.57
Propanol	4.80	1.51	2.78	5.4	4.76	1.52	2.69
Butanol	4.74	1.56	2.88	5.0	4.64	1.57	2.77
Octanol	4.65	1.62	3.01	2.7	4.61	1.63	2.96
Hexane	4.74	1.55	2.85	4.0	4.80	1.58	2.83
Methylcyclohexane	4.81	1.51	2.77	1.4	4.82	1.51	2.76
Toluene	4.59	1.65	3.04	2.6	4.61	1.66	3.01
Ethyl benzene	4.56	1.68	3.08	8.5	4.63	1.74	3.04
o-xylene	4.52	1.70	3.13	5.4	4.61	1.75	3.12
m-xylene	4.57	1.67	3.07	6.4	4.66	1.72	3.05
p-xylene	4.59	1.65	3.04	6.0	4.68	1.70	3.03
Mesitylene	4.61	1.64	3.02	5.9	4.70	1.68	3.01
Hexafluorobenzene	4.85	1.47	2.78	8.3	4.93	1.52	2.73
Diethyl ether	5.22	1.27	2.35	1.7	5.27	1.28	2.35
Acetone	4.77	1.53	2.81	3.9	4.83	1.55	2.80
Carbon disulphide	4.49	1.78	3.18	10.5	4.51	1.93	3.21
Heavy water	3.59	2.64	4.7	3.0	3.66	2.75	4.75

References

- [1] Ferrell R A 1957 *Phys. Rev.* **16** 108
Daniel T B and Stump R 1959 *Phys. Rev.* **115** 1599
Stewart A T and Briscoe C V 1967 *Proc. Wayne State University Conf. on Positron Annihilation* ed A T Stewart and L O Roellig (New York: Academic Press) pp 383–6
- [2] Garwin R L 1957 *Phys. Rev.* **167** 108
Dresden M 1954 *Phys. Rev.* **93** 1413
- [3] Tao S J 1972 *J. Chem. Phys.* **56** 5499
- [4] Eldrup M, Lightbody D and Sherwood J N 1981 *Chem. Phys.* **63** 51
- [5] Roellig L O 1967 *Proc. Wayne State University Conf. on Positron Annihilation* ed A T Stewart and L O Roellig (New York: Academic) p 127
- [6] Mogensen O E and Jacobsen F M 1982 *Chem. Phys.* **73** 223
- [7] Buchikin A P, Goldanskii V I, Tatur A O and Shantarovich V P 1971 *Sov. Phys.–JETP* **33** 615
- [8] Nakanishi H and Jean Y C 1988 *Positron and Positronium Chemistry* ed D M Schrader and Y C Jean (Amsterdam: Elsevier) pp 159–92
- [9] Mukherjee T, Ganguly B N and Dutta-Roy B 1997 *J. Chem. Phys.* **107** 7467
- [10] Mukherjee T, Das S K, Ganguly B N and Dutta-Roy B 1998 *Phys. Rev. B* **57** 13 363
- [11] See, for example, Plischke M and Bergersen B 1989 *Equilibrium Statistical Mechanics* (Englewood Cliffs, NJ: Prentice-Hall) p 122
- [12] Rusanov A I and Brodskaya E N 1977 *J. Colloid Interface Sci.* **62** 542
- [13] *Tables of Integrals, Series and Products* 1965 ed I S Gradshteyn and I M Ryzhik (New York: Academic)
- [14] Levay B and Vertes A 1973 *Radiochem. Radioanal. Lett.* **14** 227
- [15] Glasstone S 1947 *The Elements of Physical Chemistry* (New York: Van Nostrand) p 153
- [16] Goldanskii V I and Shantarovich V P 1974 *Appl. Phys.* **3** 335
- [17] Ryzhkov A I and Shantarovich V P 1992 *Mater. Sci. Forum* **105–110** 1699
- [18] Tolman R C 1949 *J. Chem. Phys.* **17** 333
- [19] Byakov V M and Petuchov V R 1983 *Radiochem. Radioanal. Lett.* **58** 91
- [20] Mukherjee T, Gangopadhyay D, Das S K, Ganguly B N and Dutta-Roy B 1999 *J. Chem. Phys.* at press
- [21] Ferrell R A 1956 *Rev. Mod. Phys.* **28** 308
Brandt W, Berko S and Walker W W 1960 *Phys. Rev.* **120** 1289