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Positron annihilation in small molecules

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Abstract

Positron annihilation in room temperature samples of the molecular gases N_2, O_2, CO, N_2O and CH_4 has been studied in the density range below 10 amagat using the positron lifetime technique. Careful analyses of the density dependence of the free positron annihilation rates have been performed that have allowed the annihilation parameter, $\langle Z_{\rm eff}\rangle$, to be extracted. We compare our results to those in the literature, and give recommended $\langle Z_{\rm eff}\rangle$ values from experiment. We have also synthesized the existing data for H_2 and derived a recommended value for the annihilation parameter for this molecule.

(Some figures may appear in colour only in the online journal)

1. Introduction

The interactions and fates of positrons, e^+, in various media continue to be of interest and application. Positron annihilation has found widespread use in condensed matter and materials science (see e.g., [1–3]) and is of importance in the astrophysical environment [4] where the 511 keV radiation characteristic of positron–electron annihilation is a prominent feature of the γ-ray map of the Galaxy.

These disciplines are underpinned, to some extent, by fundamental aspects of the behaviour of positrons in gaseous systems. Understanding the role of positron and positronium (the positron–electron bound state) scattering cross sections is, for example, key to explaining the manner in which positrons slow down in various media. Once stopped in whatever medium the positron will eventually annihilate, and the cross section, σ_e, for this process, which governs the annihilation probability of the positron in collisions with a single atom or molecule, is given by

$$\sigma_e = \pi r_0^2 c \langle Z_{\rm eff}\rangle / v. \qquad (1)$$

Here r_0 is the classical radius of the electron, c is the speed of light, with v the positron speed. The parameter $\langle Z_{\rm eff}\rangle$ is interpreted as the effective number of electrons available to the positron for annihilation. The brackets denote an ensemble average over the positron energy distribution, which here will be characteristic of room temperature ($T \sim 293$ K). The free positron annihilation rate, $\langle \lambda_e\rangle = n\sigma_e v$, is given in terms of the n, the number density of gas atoms or molecules, by

$$\langle \lambda_e\rangle = \pi r_0^2 cn \langle Z_{\rm eff}\rangle = 0.201\rho \langle Z_{\rm eff}\rangle \ (\mu{\rm s}^{-1}), \qquad (2)$$

where now the gas density ρ has been recast in units of amagat (1 amagat $= 2.69 \times 10^{25}{\rm m}^{-3}$). $\langle Z_{\rm eff}\rangle$, or more precisely the related speed-dependent quantity $Z_{\rm eff}(v)$, can be precisely defined once the positron–atom(molecule) wave function is known or can be calculated (see e.g., [5], chapter 6). Thus, this parameter is a natural touching point for theory and experiment.

For many years an outstanding problem in positron–molecule annihilation physics was the failure of even sophisticated theories (see e.g., [6, 7]) to reproduce the experimental value of $\langle Z_{\rm eff}\rangle \sim 16$ for the hydrogen molecule. This situation has been resolved recently by Mitroy and co-workers [8] using a confined variational method. We have taken inspiration from this advance and anticipate that further detailed theoretical studies of positron annihilation in small molecules will soon be available, and this has motivated the present study.

Here we report room temperature values for $\langle Z_{\rm eff}\rangle$ for a number of small molecules. Values of this parameter for the species studied here can already be found in the literature, though they are often quoted without uncertainties, may be based upon investigations performed over twenty five years ago, and/or be derived from unpublished data. Various compendia of these data can be found (e.g., [5, 9–11]). Our aim is to provide accurate values of $\langle Z_{\rm eff}\rangle$, with associated uncertainties, which can be used as benchmarks for future investigations from both theory and experiment.

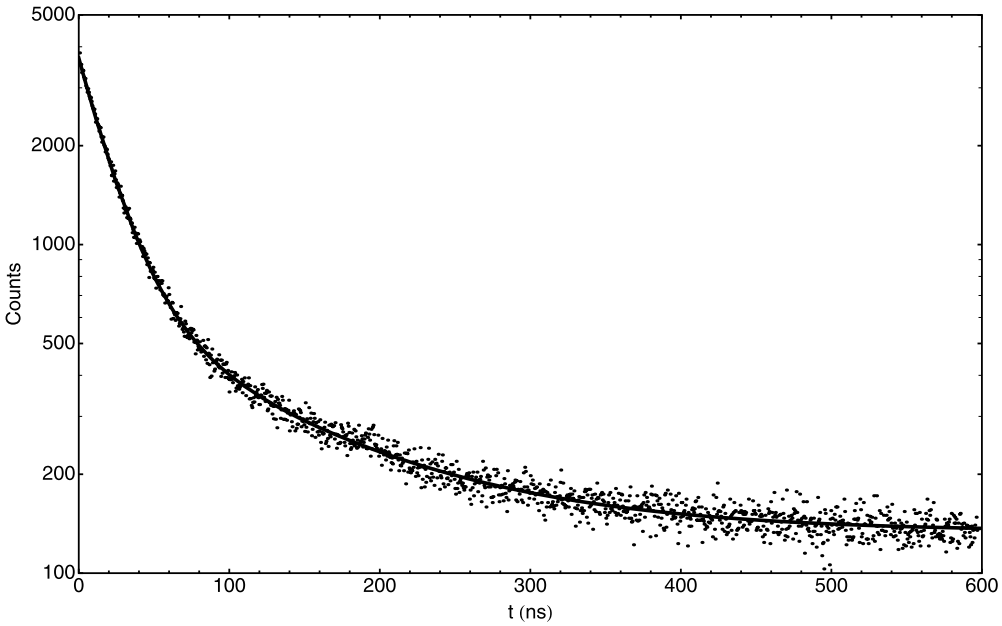

Figure 1. Example of a lifetime spectrum for 5.56 amagat CO, with the prompt peak removed. The solid line is a fit to a double exponential plus background, with a free positron decay rate of $46.0 \pm 0.3\ \mu s^{-1}$.

The remainder of the paper is organized as follows. The next section gives a brief description of the apparatus, the data analysis methods used to extract values of $\langle \lambda_e \rangle$ and details of the gas samples employed. Section 3 presents the data, a discussion of the manner in which $\langle Z_{\rm eff} \rangle$ was extracted and a comparison with existing values: this section also contains an analysis of literature values for H_2. For all species we present recommended values for $\langle Z_{\rm eff} \rangle$. This section also contains a discussion of the effect of impurities on the measured value of $\langle Z_{\rm eff} \rangle$. Section 4 contains the main conclusions of our study.

2. Experimental details

A positron lifetime spectrometer was used for this study which was based around a conventional time-to-amplitude-converter/multi-channel analyzer system (see e.g., [5, 12]). The positrons were derived from a 100 kBq ^{22}Na source sandwiched between two kapton foils of 10 μm thickness that were mounted in a purpose-built holder in the centre of a gas chamber. The delayed coincidence between the nuclear gamma quantum, with an energy of 1.274 MeV (start signal), and one of the photons emitted in the annihilation of the positron (stop signal) was measured. The gamma rays were registered by two plastic scintillator–photomultiplier tube arrangements located on either side of the chamber.

The gas chamber consisted of a stainless steel cylinder, of inner diameter 35 and 210 mm in length, into which the sample gases could be admitted at pressures up to 10 atmospheres. The gas pressure was recorded using a Druck PDCR 4010 pressure transducer. The temperature of the room was kept stable at around 293 K, to about a degree or so. Gas densities were computed from the temperatures and pressures using accepted first order virial coefficient corrections [13].

In all cases the lifetime spectra contained a large prompt peak due to positron annihilations in the source holder and at the chamber wall. After this peak, events from positrons stopped in the gas could be fitted by two overlapping exponentials with a constant background (see figure 1 which shows a typical example). For most of the gases studied here, the faster of these two components was that due to the free positrons (the other was due to the annihilation of ortho-positronium atoms), such that $\langle \lambda_e \rangle$ could be extracted at each density. The exception is O_2. This molecule has a lone pair of electrons and is known [14, 15] to efficiently convert triplet ortho-positronium to the short-lived (125 ps) singlet, para-positronium. This process is much more rapid than annihilation of free positrons, such that the latter forms the long-lived component in the annihilation spectra of this molecule.

Table 1. The purity of the gas samples used in the present study, together with the recommended values for the positron annihilation parameter $\langle Z_{\rm eff} \rangle$. In all cases the impurities as stated by the suppliers were carbon dioxide, nitrogen and unspecified hydrocarbons.

Gas	Stated purity	Recommended $\langle Z_{\rm eff} \rangle$
N_2	99.998%	30.8 ± 0.2
O_2	99.999%	26.5 ± 0.1
CO	99.5%	39.7 ± 1.0
N_2O	99.5%	68.2 ± 1.0
CH_4	99.995%	140.0 ± 0.8
H_2	various	16.0 ± 0.2

The gas samples in the present study were obtained from commercial suppliers and were used without further purification. Table 1 presents brief details for each gas. Whilst it is not anticipated that impurities will play a major role in this experiment, there are certain molecular species (e.g., the alkanes) which have very high values of $\langle Z_{\rm eff} \rangle$ (see e.g., [16] for a review), such that even the presence of small quantities may falsely enhance the measured $\langle Z_{\rm eff} \rangle$ over the true value. The hydrocarbon impurities present in our gas samples were listed as unspecified by the manufacturers. Samples of CO, N_2O and

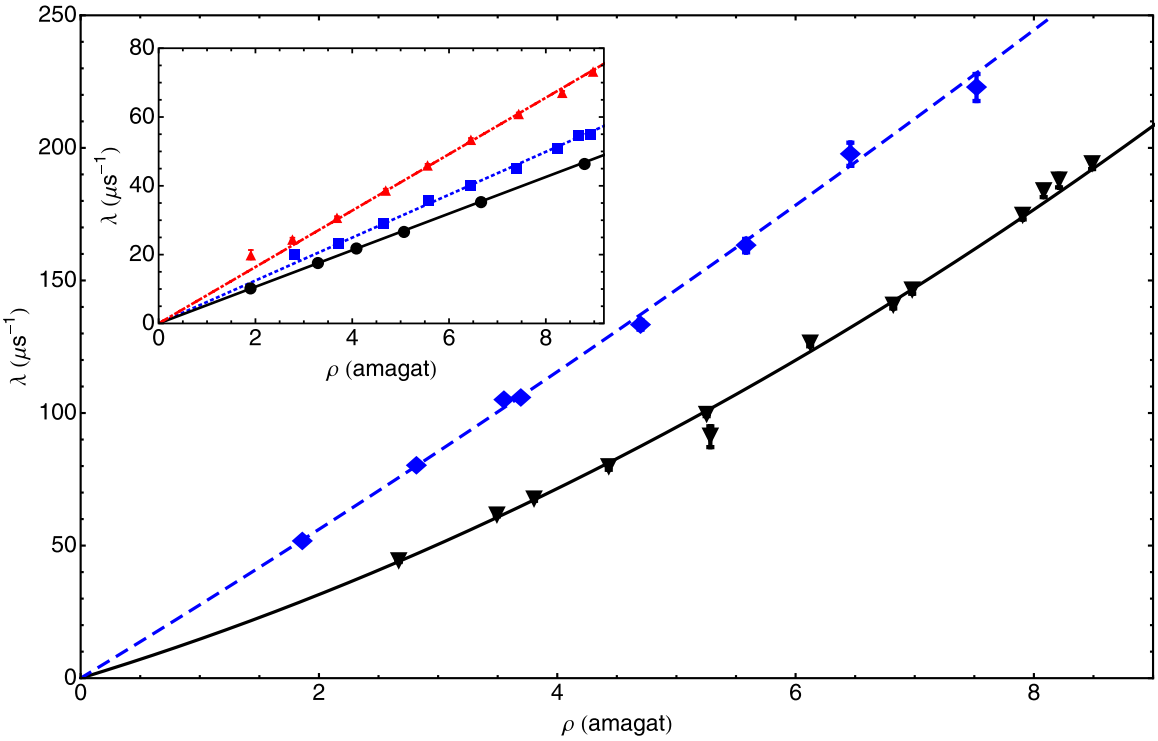

Figure 2. The behaviour of the free positron annihilation rate versus gas density at $T = 293$ K for CH_4 (♦) and N_2O(▼) with the inset showing CO (▲), N_2 (■) and O_2 (●). The lines are the fits as described in the text.

CH_4 were subject to a residual gas analysis by leaking small quantities into a high vacuum chamber. No consistent traces of hydrocarbon impurity were found in any of the gases at the impurity level stated in table 1.

3. Results and discussion

In order to extract vales for $\langle Z_{\rm eff}\rangle$ that are as reliable as possible, our study has concentrated on the low density range to avoid phenomena such as positron annihilation in clusters and/or saturation effects, which are well documented at high densities (see e.g., [11, 17, 18] for synopses). We have also been guided by the recent isolation of three-body effects in positron–molecule annihilation in a selection of species at room temperature [19, 20]. It appears that such processes may occur in the range of densities used in this study and proceed via a reaction of the type, $e^+ + M + M \rightarrow 2\gamma + M^+ + M$ (where M is a molecule), a process which can take place alongside the usual two-body reaction, $e^+ + M \rightarrow 2\gamma + M^+$. It is the latter which must be used to determine $\langle Z_{\rm eff}\rangle$, but if the three-body process is conflated, or an average over a range of densities is taken, then the derived value of $\langle Z_{\rm eff}\rangle$ may be in error.

We note, following [20], that assuming both the two- and three-body reactions can occur, then the positron annihilation rate (see equation (2)) can be rewritten to include an extra term as,

$$\langle \lambda_e \rangle = 0.201 \langle Z_{\rm eff}\rangle \rho + \langle b \rangle \rho^2. \tag{3}$$

Fitting $\langle \lambda_e \rangle$ yields the parameter $\langle Z_{\rm eff}\rangle$, characteristic of the two-body e^+–M interaction, and the so-called three-body coefficient, $\langle b \rangle$, which has been the object of separate studies [12, 19, 20]. The behaviour of $\langle \lambda_e \rangle$ versus gas density is shown for all five samples in figure 2, with lines fitted according to either equation (2) or (3). Equation (3) was used for CH_4 and N_2O, but for the other three gases (shown in the inset of the figure) equation (2) proved adequate; i.e., fitting with equation (3) yielded a three-body correction consistent with zero. In the remainder of this section we will give a brief general discussion of the potential influence on positron annihilation of impurities in the gas samples, before describing our results for each gas in turn with reference to previous experimental work, and to theory where available.

3.1. Role of impurities

The discovery that certain species of molecules, and in particular the higher alkanes [16], can have very high values of $\langle Z_{\rm eff}\rangle$ means that a quantitative discussion of their possible influence is necessary. Given that the overall annihilation rate due to a mixture of gases is the sum of those for the individual species, it is straightforward to show that if there are i gases present in a sample, then the measured annihilation parameter, $\langle Z_{\rm eff}\rangle^m$, can be found from

$$\rho \langle Z_{\rm eff}\rangle^m = \sum_i \rho_i \langle Z_{\rm eff}\rangle^i. \tag{4}$$

Here the ρ_i are the partial densities for the individual gas species. Clearly for our samples the main gas is dominant ($> 99.5\%$) such that equation (4) can be rewritten approximately as,

$$\langle Z_{\rm eff}\rangle^m \approx \langle Z_{\rm eff}\rangle^1 + \sum_{i\geqslant 2} f_i \langle Z_{\rm eff}\rangle^i, \tag{5}$$

where $f_i = \rho_i/\rho$ and such that the quantity of interest $\langle Z_{\rm eff}\rangle^1 = \langle Z_{\rm eff}\rangle \approx \langle Z_{\rm eff}\rangle_m - \sum_{i\geqslant 2} f_i \langle Z_{\rm eff}\rangle^i$. From here it is easy to see that if the impurity gases have modest values of $\langle Z_{\rm eff}\rangle$ (i.e., $\sim Z$, the actual number of electrons in the molecule) then, for sub-%-age values of f_i, their effect will be negligible in the sense that any enhancement will be much lower than the measurement or averaging uncertainties (see sections 3.2–3.7).

3.2. Nitrogen

The N_2 data were fit as shown in figure 2 to yield $\langle\lambda_e\rangle = (6.24 \pm 0.04)\rho$, with the corresponding value of $\langle Z_{\rm eff}\rangle$ of 31.0 ± 0.2.

There have been several measurements of $\langle Z_{\rm eff}\rangle$ for N_2, with those at room temperature mainly due to the UCL group, though mostly reported in obscure publications or conference proceedings. The earliest measurement [21] yielded an average value of 28.89 ± 0.11 over the density range 7–28 amagat. Shortly thereafter Coleman *et al* [22] reported that $\langle Z_{\rm eff}\rangle$ fell linearly with density over the range 5–70 amagat with a zero-density limit of 30.6 ± 0.2. This value was used in a study of positron slowing in nitrogen gas [23], which also included the data at 77 K of Sharma and McNutt [24].

The only other experimental work appears to be that of Tao [25] who found a $\langle Z_{\rm eff}\rangle$ value of 28.9 ± 0.4 over the range from about 8 to 45 amagat. With the work of Coleman *et al* [22] finding $\langle\lambda_e\rangle$ falling below the linear extrapolation from low density it may be expected that a value averaged over a wide density range will be lower than the true value. Though the free positron annihilation data presented by Tao [25] are apparently linear with density, this is the most probable cause of the discrepancy between this earlier work and the later studies. There is no information on the purity of the gas samples used in either of the earlier studies ([22, 25]).

At present we view that the results from the current study have corroborated the zero density value of Coleman *et al* [22]. As such we recommend the value of $\langle Z_{\rm eff}\rangle = 30.8 \pm 0.2$ as a standard experimental value for N_2.

The only theoretical work for comparison appears to be that of Darewych and Baille [26], Gianturco and Mukherjee [27] and Lima and co-workers ([28, 29]), with the correction of do Varella *et al* [29] noted. The oldest of these works [26] using a Born–Oppenheimer type of approximation, with the inclusion of a heuristic polarization potential borrowed from electron-N_2 scattering physics, obtained a $\langle Z_{\rm eff}\rangle$ value of around 23 at room temperature. This is in reasonable accord with experiment, but this situation may be misleading. Later, the more sophisticated close-coupling [27] and multi-channel Schwinger [28, 29] works produced a spread of $\langle Z_{\rm eff}\rangle$ values from around 10 to 48. Thus, the situation from theory remains uncertain, but we have now provided a reference value from experiment for the $\langle Z_{\rm eff}\rangle$ of N_2.

3.3. Oxygen

The O_2 data were fit to equation (2) to yield $\langle\lambda_e\rangle = (5.30 \pm 0.02)\rho$, as the three-body correction is consistent with zero. The resulting $\langle Z_{\rm eff}\rangle$ value is 26.4 ± 0.1. The only other value of $\langle Z_{\rm eff}\rangle$ for comparison seems to be that from the unpublished work of Wright [30] (private communication from Laricchia). Analysing that data (which do exhibit a small density dependence in the low density region) yields a $\langle Z_{\rm eff}\rangle$ of 28.0 ± 0.5. The purity of the gas used was stated to be 99.7% [30], though the nature of the impurities was not given. Thus, the two experimental values are somewhat inconsistent, though the present value has lower uncertainties. As a result we have taken a weighted average, with the associated uncertainty, to arrive at a current best value of $\langle Z_{\rm eff}\rangle = 26.5 \pm 0.1$.

To the best of our knowledge, there is only one theoretical work on annihilation in O_2 [27]. Here, two values of $\langle Z_{\rm eff}\rangle$ were given using fixed nuclei and body-fixed variational close-coupling methods. These are 65 and 54, respectively, and as such are in marked disharmony with experiment.

3.4. Carbon monoxide

Fits to the CO data yielded $\langle\lambda_e\rangle = (8.20 \pm 0.04)\rho$, resulting in a $\langle Z_{\rm eff}\rangle$ of 40.8 ± 0.2. The only other values of $\langle Z_{\rm eff}\rangle$ for CO appear to be those quoted by Griffith and Heyland [9], who found the parameter to vary between 38.5 and 24 over the density range 0–172 amagat, with no stated uncertainties. These authors note that a previous value of 69.4 [31] was in error due to the use of an impure gas sample.

The gas sample of CO used here was, at 99.5% purity, one of the most impure samples in our study. It is possible that our measured values of $\langle Z_{\rm eff}\rangle$ may be skewed upwards by this, if there were hydrocarbons present that we could not identify by our residual gas analysis. No information on gas purity was given by Griffith and Heyland [9]. Thus, we assign an average value for $\langle Z_{\rm eff}\rangle$ of CO, with a conservative uncertainty, as 39.7 ± 1.0. To the best of our knowledge, there is just one theoretical study of CO [27], which yielded a value around 33, in acceptable accord with experiment.

3.5. Nitrous oxide

The fit of equation (3) to the N_2O data yielded $\langle\lambda_e\rangle = (13.7\pm0.2)\rho+(1.05\pm0.04)\rho^2$, resulting in a $\langle Z_{\rm eff}\rangle$ 68.2 ± 1.0. This is somewhat lower than the value of 78 quoted without uncertainty and unattributed in the compendium given by Heyland *et al* [10], and to the best of our knowledge there is no theory for comparison. Thus we adopt the value derived from this study as a recommended value of $\langle Z_{\rm eff}\rangle$ for N_2O.

3.6. Methane

The fit of equation (3) to the CH_4 data yielded $\langle\lambda_e\rangle = (27.2\pm0.5)\rho+(0.42\pm0.13)\rho^2$ leading to $\langle Z_{\rm eff}\rangle = 135.3\pm2.5$.

There are several other experimental studies with which to compare. Smith and Paul [32] found a value of 139.6 ± 1.0 from a study confined to gas densities below 2 amagat, with no nonlinearities evident in their annihilation rate data, which were plotted versus gas pressure. A study of the behaviour of $\langle Z_{\rm eff}\rangle$ in methane under the influence of an electric field [33] yielded a zero-field value of 142.7 ± 2.0 at a density of around 1 amagat. In a wide-ranging study of the behaviour of positrons and positronium in methane gas, McNutt *et al* [34] found a low density value of 153.7 ± 0.9. Finally, Wright and

co-workers [35] obtained the value of about 142 ± 2 at low density, rising to around 155 by 16 amagat. Impurity details were, in this case, given in all three studies and using this information, equation (5), and appropriate values for $\langle Z_{\rm eff}\rangle$ for the trace gases reveals that they would have had a negligible effect on these measurements.

It is clear that the work of Mao and Paul [33], Smith and Paul [32] and Wright *et al* are in reasonable accord, but that the present work and that of McNutt and co-workers [34] find $\langle Z_{\rm eff}\rangle$ values much different. If, in the present experiment, we had not fitted the annihilation rate using equation (3), an average value of $\langle Z_{\rm eff}\rangle$ of around 142 is implied by our data. This may account for the discrepancy with the result of Wright *et al* [35] who measured over a similar density range to the present work, but not the discord with Paul and co-workers [32, 33]. It is likely that the value of McNutt and co-workers is an overestimate of the room temperature $\langle Z_{\rm eff}\rangle$, as their final value was an average of experiments performed at three temperatures, with two below ambient. Since it is expected that $\langle Z_{\rm eff}\rangle$ will increase as the gas temperature is lowered, such an effect may well have skewed their average value. However, we cannot find fault with the UCL and Toronto work, which included careful studies at low density. Thus, our recommended value of $\langle Z_{\rm eff}\rangle$ is the weighted mean (with associated uncertainty) of all the studies bar McNutt *et al* [34] to arrive at a value of $\langle Z_{\rm eff}\rangle = 140.0 \pm 0.8$ for CH_4.

On the theory side there are two works, from Jain and Thompson [36] and Gianturco *et al* [37]. The polarisation potential model calculation of Jain and Thompson [36] yielded a value for $\langle Z_{\rm eff}\rangle$ of around 100, in very acceptable accord with experiment, given the nature of the calculation. In an exploratory study of a number of polyatomic species, Gianturco and co-workers obtained a value of $\langle Z_{\rm eff}\rangle$ of close to 65 for CH_4.

3.7. Hydrogen

The theoretical situation for molecular hydrogen was summarized recently by Gribakin, Young and Surko [38] in a comprehensive review of positron–molecule annihilation phenomena. Over the years a series of Kohn variational calculations by Armour and co-workers pushed the H_2 $\langle Z_{\rm eff}\rangle$ up from the early theory value of just 2, to around 10 [7, 39, 40]. However, the state-of-the-art work for molecules is that for H_2 by Zhang *et al* [8] who obtained a value of 15.7. The work of Lima and co-workers [28, 29, 41] yielded a value of around 7.3.

There have been three experiments that have derived $\langle Z_{\rm eff}\rangle$ values for H_2 [35, 42, 43] at room temperature. These studies were performed for different reasons (such as exploring the density dependence of the free positron annihilation rate and studying the behaviour of the fraction of positrons that form positronium) and over a range of gas densities and temperatures. McNutt *et al* [43] found an average value of 14.8 ± 0.2, measured over the density range from 10-47 amagat, whilst Wright *et al* [35] fitted their data to find $\langle Z_{\rm eff}\rangle = (16.02 \pm 0.08) - (0.042 \pm 0.003)\rho$ over the region 12–37 amagat. (Note that the highest value measured by Wright *et al* was 15.5 ± 0.2 at a density around 12 amagat.) In a wide-ranging study Laricchia *et al* found an average value of 14.61 ± 0.14 for the range 20–40 amagat. By computing values at the extremes of Laricchia's density range from the fit of Wright *et al*, a mean of 14.76 ± 0.20 ensues, in excellent accord with the value found by the former. Performing a similar analysis for the density range of McNutt *et al* [43] yields an average of 14.82 ± 0.22, in accord with the average value from that experiment.

Thus, we can conclude that all three experiments are in good agreement. We select the final value of $\langle Z_{\rm eff}\rangle$ for H_2 of 16.0 ± 0.2, by assuming the adequacy of the fit of Wright *et al*, though assigning a somewhat larger uncertainty to reflect typical measurement uncertainties of all three experiments.

4. Conclusions

We have presented $\langle Z_{\rm eff}\rangle$ values for the molecular species N_2, O_2, CO, N_2O and CH_4 at a temperature of 293 K. Furthermore, we have conducted a review of the available experimental data for H_2 and from that synthesis have derived a recommended value of $\langle Z_{\rm eff}\rangle$ for this gas. We consider these values to be benchmarks to be used when comparing to future experiments and to theory. We hope that the present study will inspire more theoretical work on low energy positron annihilation as tests of few-body physics.

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