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Measurements of Positron Annihilation Rate in Low-Temperature Gaseous Nitrogen

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The positron annihilation rates have been measured in gaseous nitrogen at 100, 120, 130, 150 and 170 K at densities in the range 3.3–428 amagat. Even at sufficiently low densities at all temperatures, the positron annihilation rates increase with density faster than the linear relationship. This behavior has been discussed in connection with the annihilation of positrons interacting with the cluster of N_2 molecules. The results at much higher densities are in good agreement with the existing data.

KEY WORDS: Positron annihilation rate/ Nitrogen/ Cluster of molecules/

I INTRODUCTION

Positron lifetime measurements in gases have a long history more than thirty years and have provided interesting information on the behavior of slow positrons in gases. In sufficiently low-density gases, the behavior of them may be understood by a picture that they interact with single atoms and molecules. The positron lifetime measurements in such condition give severe tests of positron-gas scattering theories, and are the only method, in practice, giving direct information in the energy range less than ~0.5 eV which is not accessible by the scattering experiments by using low-energy positron beams. On the other hand, in the condition of relatively high densities near the critical temperatures (T_c), it is known that a positron causes local condensation of gas atoms or molecules around itself, due to the dominant attractive nature of the interation.[1–3] This is the self-trapped state of the positron into a cluster. This phenomenon provides information on the positron-gas (-fluid) interaction and on the properties of the fluid itself.

In the present work, we have carried out the positron lifetime measurement in gaseous N_2. N_2 is one of a few gases for which theoretical studies of the positron scattering have been presented[4–6], and the only molecular gas which has a "shoulder" on its lifetime spectra.[7] For N_2, although some precise measurements have been performed at room temperature[8], there is only a limited amount of detailed low-density measurements at low temperatures around T_c.[3,9] Such measurements provide useful information on a positron-N_2 scattering at other than room temperature, that is, at thermal energies other than those corresponding to room temperature. On the positron-induced cluster phenomenon, experimental and theoretical works have been repoted for N_2. The present

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measurement in low-temperature N_2 provides more data for further understanding of this phenomenon.

II EXPERIMENTAL

The chamber (28 mm in internal diameter and 70 mm in active length) was machined from copper (OFHC) rod, and its internal surface was electroplated with gold to enhance the fraction of positrons stopping in the gas. A ^{22}Na positron source ($\sim$7 μCi) deposited on a thick gold foil was wrapped by a thin Mylar film and held near the chamber wall. The chamber wall is about 4 mm thickness and this provides a resisting pressure over 100 atm and sufficient heat conduction. A lead-plated annealed copper ring gasket was used for sealing of the high-pressure gas. As positron lifetime parameters are sensitive to impurities in the sample gas, careful gas handling is required. Gas lines were constructed of stainless-steel tubing (6. 34 mm outer diameter). The system was cleaned with high-purity trichloroethylene before assembly, and then the evacuating and gas flushing procedure was performed for many times. The chamber was held in a sealed copper cylinder immersed in liquid N_2 by a vacuum-insulated gas supply tube. A heater was wound along the chamber. A Pt resistance thermometer and a Au(+0. 07% Fe) –chromel thermocouple were inserted into small holes drilled at the bottom part of the chamber. Temperature of the chamber was controlled by balancing cooling by low-pressure exchange gas with heating by the heater coil supplied from a temperature controller. The stability of the temperature was fairly good. The temperature of the chamber was measured by the Pt thermometer, which has been commercially calibrated. In order to test the manufacturer's calibration and estimate temperature difference over the chamber, we compared the published vapor-pressure curves of liquid N_2[10] and Ar[11] with measured vapor-pressures in the chamber. The uncertainty in temperature determination, arising from the combined effects of temperature difference and of errors in the calibration, is estimated to be less than $\pm$0. 5 K. Commercially supplied high-purity grade N_2 (Minimum purity 99. 9999%; maximum O_2 0. 2 ppm) was used for the sample gas. Gas pressures were measured by using precision Bourdon gauges. The accuracy of the measured pressure is better than 1% for all the data points above 2. 4 atm and better than 2% for the other lower data points. The measured pressures were corrected for barometric fluctuations at low pressures. Density of N_2 gas, D (in amagat; 1 amagat$=2.69\times10^{19}$ cm^{-3}), was evaluated from measured pressure-temperature data by using a equation of state given by Jacobsen et al[10]. The error in D due to all causes is estimated to be less than 3% [for all data at 100, 120, 150 and 170 K, and for those in regions $D\lesssim100$ and $D\gtrsim350$ at 130 K. For data except for the above regions at 130 K it exceeds 3% and especially in the vicinity of the critical point ($150\lesssim D\lesssim300$ at 130 K; $T_c=126.2$ K and $D_c\sim250$ amagat) it becomes over ten %.

Two types of positron lifetime measuring systems were employed in the present measurement. At low densities, the high counting rate system was used, which is similar to one developed by Coleman et al[12], in order to obtain data with good statistical-accuracy. Detectors consisted of large fast plastic scintillators (12 cm diameter $\times$10 cm and 12 cm diameter $\times$ 7. 5 cm for start and stop detectors, respectively) and 5" photomultipliers (RCA 4522). The start and stop rates were approximately 7×10^3 and 2×10^4 sec^{-1}, and the coincidence rate was about 6×10^2 sec^{-1}. The time resolution was 1.2 nsec FWHM for ^{60}Co. A raw spectrum obtained from this system is deformed by random-coincidence events due to the high counting rate. Therefore, it was processed by the "signal restoration method"[13] to deduce the restored ("true") spectrum before the non-linear fitting analysis. The restored spectrum was analyzed by a simple exponential fit program. For all the data except for those at 150 K and those lower than 4.8 amagat at 170 K, the spectrum was fitted into two exponential components (positron and ortho-positronium (o-Ps) components) as variable parameters in the region about 15 nsec far from the peak position ensuring that the prompt and shoulder contributions were excluded. For all the data at 150 K and the lower-density data at 170 K, a fixed value of the ortho-positronium annihilation rate, λ_{o-Ps}, which was calculated by the relation of $\lambda_{o-Ps}=7.06+0.203D$ μsec^{-1} [14], was used. At low temperatures, λ_f increased rapidly with the increase of density, and the positron component approached to the prompt peak too closely to separate them each other. For such case, we used another system (the high time resolution system) for the measurement of λ_f. This system was a fast-slow coincidence spectrometer, and consisted of fast plastic scintillators (Pilot-U; 2" diameter × 2") and 2" photomultipliers (Hamamatsu Photonics R1246A and RCA 8575 for start and stop detectors, respectively). The time resolution was 0.55 nsec FWHM for ^{60}Co. The spectrum measured by this system was directly analyzed by the simple exponential fit program. It was fitted into two exponential components and a constant of background. Due to its low intensity, λ_{o-Ps} was fixed to the value which was obtained from the spectrum measured by the high counting rate system at the same period. The fit region was chosen by examining the resultant λ_f and the variance of fit in order to assure to exclude contribution of the shoulder region, if any. The present results of λ_f larger than $\sim$400 μsec^{-1} were measured by this system.

III RESULTS AND DISCUSSION

A. Room temperature

In order to check the reliability of the measuring systems, measurements have been carried out at 296 ± 2 K at several density points, and λ_f has been compared with the existing values. λ_f against density is shown in Fig. 1-a. Griffith and Heyland[8] performed precise measurement at room temperature, and obtained the empirical relation of $Z_{eff}=20e^{-0.0046D}+10.6$ where $Z_{eff}=\lambda_f/\pi r_0^2 c n_0 D$, r_0 is the classical electron radius, c the velocity of light and n_0 the standard number density. Z_{eff} against density is also shown in Fig. 1-b. The present result is in good agreement with that by Griffith and Heyland. In the case of free positron annihilation in gases without collective effects, Z_{eff} is expected to be constant, and can be straightly compared with theoretical values obtained from scattering theories. However, Z_{eff} decreases gradually with the increase of density in both results by Griffith and Heyland, and us. This behavior of Z_{eff} has been attributed to the energy dependence of Z_{eff} by Rytsölä et al[3].

B. Low temperatures

The measurements have been performed in the density ranges of 4.2–25.5, 3.5–

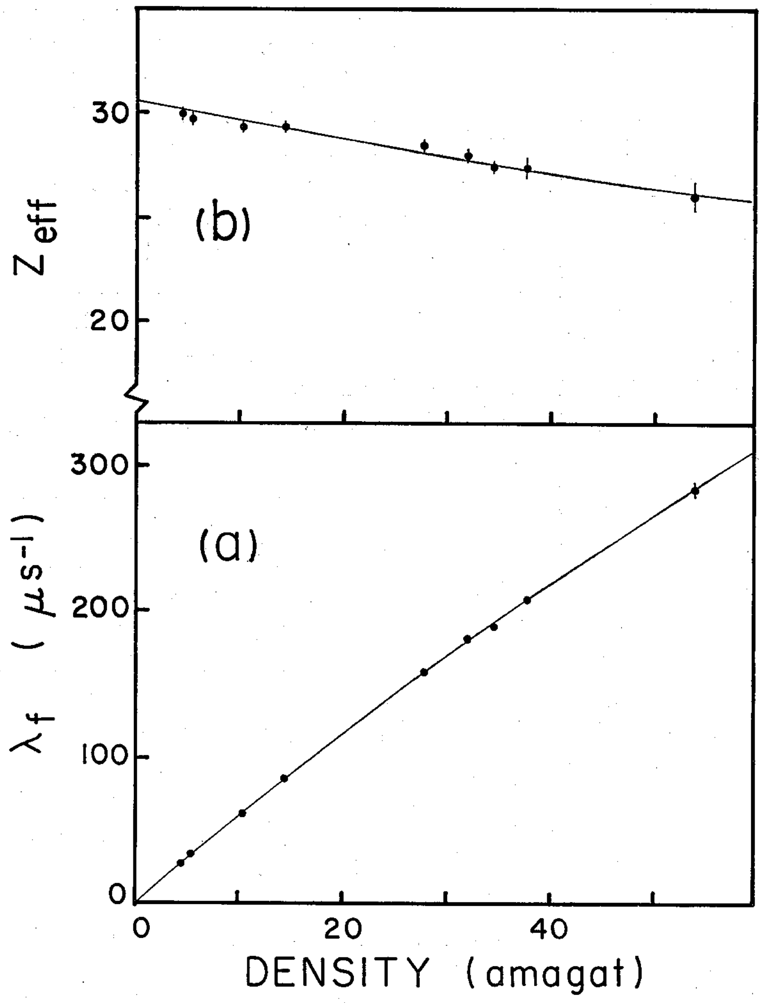

Fig. 1-a The positron annihilation rate λ_f against density at room temperature. A solid line represents the relation obtained by Griffith and Heyland.[8]

Fig. 1-b Z_{eff} against density at room temperature. A solid line represents the relation by Griffith and Heyland.

76.4, 3.4–428, 3.4–9.5 and 3.3–170 amagat at 100, 120, 130, 150 and 170 K, respectively. The results at densities lower than 14 amagat are shown in Fig. 2. The data even at low densities are apparently larger than the result at room temperature. At 100 K, λ_f increases rapidly with density from the lowest density point. At the other temperatures, at sufficiently low densities λ_f appears to increase approximately linearly with density. Z_{eff} against density also is shown in Fig. 3. At 100 K, Z_{eff} increases steeply with density, reflecting the rapid increase of λ_f. Even at 120–170 K, Z_{eff} gradually increases with density, and this means that λ_f increases faster than the linear relationship. Thus the constant value of Z_{eff} for each temperature has not been obtained from the present data. Rough linear-extrapolations of Z_{eff} plots to zero density seem to give values of $\sim$31 at 150 and 170 K. However, such estimation of Z_{eff} at very low densities seems

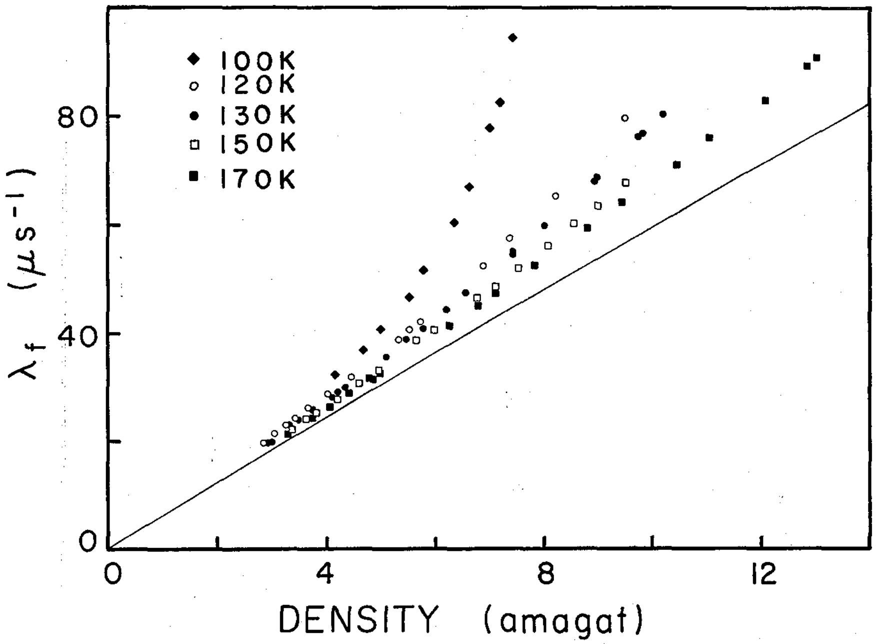

Fig. 2 The positron annihilation rate λ_f against density, in the region less than 14 amagat at low temperatures. A solid line represents the same relation as that in Fig. 1-b.

to be unproper, at least for 100, 120 and 130 K, because of somewhat concave shapes of Z_{eff} behaviors. Sharma and McNutt[9] reported that in gaseous N_2 at 77 K, λ_f increases linearly with density up to 2. 3 amagat and arises very rapidly from the linear relationship above this density. The constant value of Z_{eff} at 77 K was calculated to be 40. 0±0. 9 from their linear region. The constant Z_{eff}, namely the linear dependence of λ_f on density, might be found also at 100–170 K at lower regions than the present lowest density points.

The behaviors of λ_f over the whole density ranges are shown in Fig. 4, partly omitted in the lower-density regions. At all temperatures except 150 K, λ_f increases rapidly with density, differing from the behavior at room temperature. The rate of increase of λ_f against density becomes smaller at higher density portion after a rapid rise. Particularly at 130 K, λ_f shows a plateau above about 150 amagat, and stays approximately constant up to the maximum density points. At 170 K, λ_f also shows a convex shape at higher density portion, although it does not show a plateau in the present range.

Recently, Rytsölä et al[3] measured λ_f in the wide range of temperature of 60–400 K in solid, liquid and gaseous N_2. They analyzed the lifetime spectrum, fitting it into two exponential components using a fixed value of λ_{o-Ps} calculated from the relationship of $\lambda_{o-Ps}=(7.24+0.207D)$ μsec^{-1} by Coleman et al.[15] The fitting procedure by Rytsölä et al differs from that by us in the value of the fixed λ_{o-Ps}. For exact analysis, we have used our experimental value measured by the high counting rate system at the same period. Certainly, the experimental value of λ_{o-Ps} does not so much differ from the above calculation, as verified by our previous results[16]. However, at sufficiently high

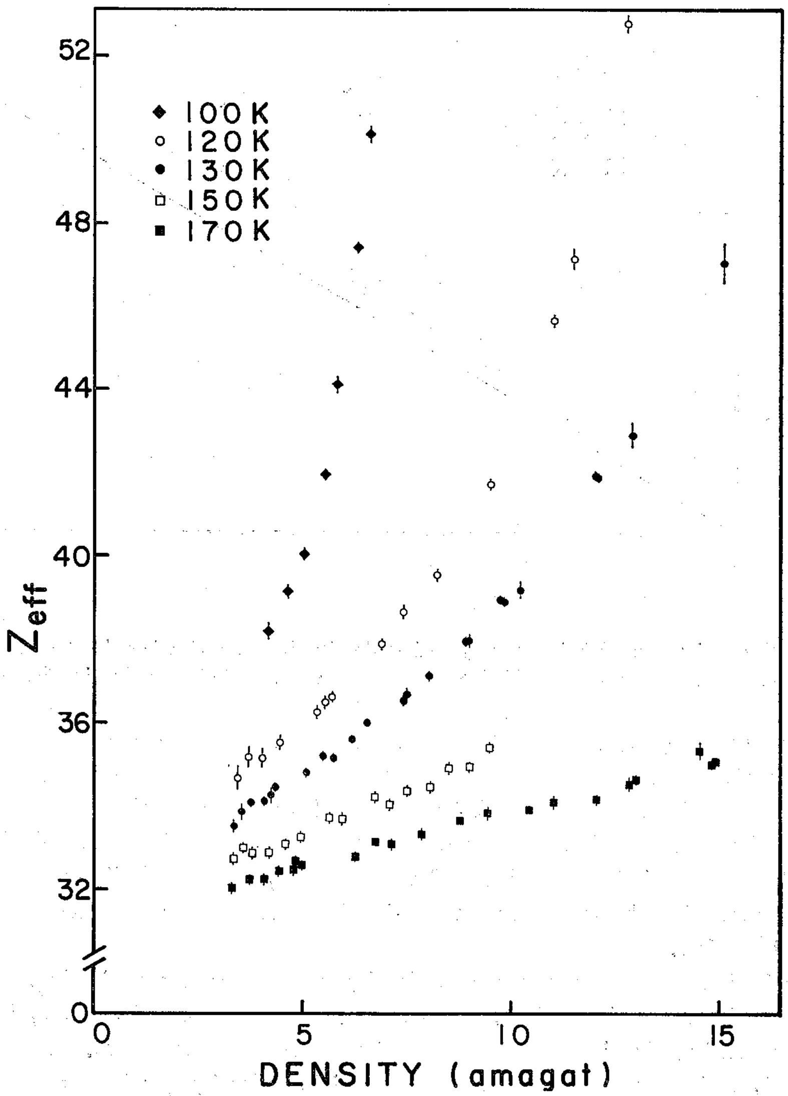

Fig. 3 Z_{eff} against density, in the region less than 16 amagat.

densities, particularly at 130 K, the experimental value of λ_{o-Ps} is considerably smaller than the calculation. Use of the calculated value for λ_{o-Ps} might lead to errorneous results of λ_f. For comparison, we have also tried the similar fit to the same spectrum with a fixed value of λ_{o-Ps} calculated by the same relation[15]. The value of λ_f obtained by this procedure was larger than the corresponding value in Fig. 4, and particularly in the plateau region at 130 K the difference exceeded twice of the standard deviation and the variance of fit became worse. In the case that the fitting region of the spectrum was limited into the region where the positron component appeared to decay, the results obtained by the two kinds of procedures agreed within the standard de viation with each other and they also agreed well with those in Fig. 4. In spite the

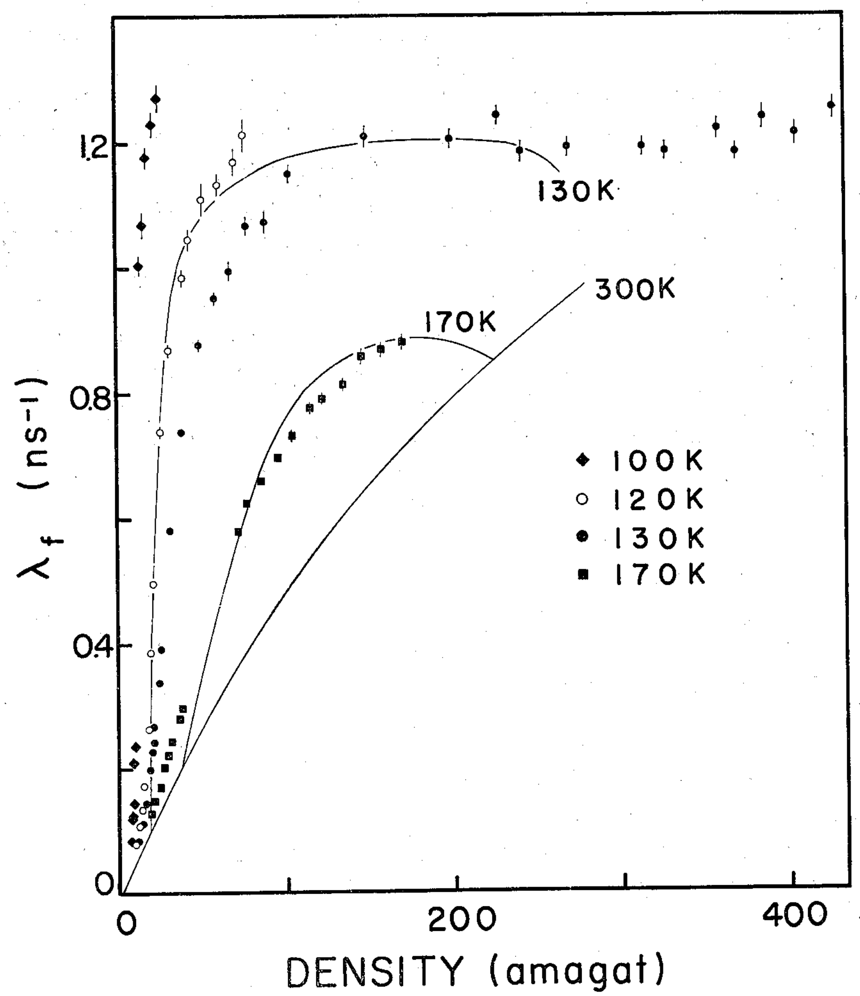

Fig. 4 The positron annihilation rate λ_f against density. The data points at densities less than 20 amagat are partly omitted for clarity. Solid lines represent the calculation by Rytsölä et al[3] for 130 and 170K, and the same relation as that in Fig. 1-a. for 300 K.

difference of the fixed values of λ_{o-Ps}, the agreement between the present resluts and those of Rytsölä et al is good. This might be due to that their results did not so much influenced by the use of the invalid value for λ_{o-Ps} because of a small number of events of o-Ps in their spectrum.

This behavior of λ_f in N_2, namely a rapid increase and a successive plateau, is quite similar to that in He at low temperatures, although overall feature of the former is much smoother. These phenomena are attributed to the formation of a cluster around a positron. The theoretical result of Rytsölä et al[3] by using the density functional method, is also shown in Fig. 4. In this calculation, the spherically averaged positron-N_2 molecule potential was used, which was represented as a sum of a Coulombic repulsive part and a symmetric polarization part with a cutoff radius r_c neglecting the asymmetry of the N_2 molecule. The calculation by using a singe value of r_c as a fitting parameter did not provide the overall agreement with the experiment over the wide range of the temperature, although the choice of $r_c=1.43$Å, which corresponds to the positron–N_2 scattering length of -1.06Å, gives a better fit than other choice. Further theore tical effort is hoped, since this kind of calculation is one of the approaches which may provide a scattering length of a positron-molecule collision.

The behavior of λ_f at sufficiently low densities at low temperatures has been measured in detail for the first time in the present work. Even in these low densities λ_f does not increase linearly with density as seen in Fig. 2, or equivalently Z_{eff} gradually increases with density as seen in Fig. 3, and this means that the behavior of the positron can not be interpreted in terms of the simple picture of the positron scattering with a single N_2 molecule. In many gases very gradual decrease of Z_{eff} with the increase of density has been observed at the temperatures considerably far from the critical temperature T_c, and this behavior has been attributed to the multiple scattering[17–19] or the energy dependence of Z_{eff}.[3] The present results showing gradual increase, however, contrast to the behavior mentioned above, so that such interpretations can not be applied. It is reasonable that the present increase of Z_{eff} is closely related to the annihilation of positrons interacting with the cluster of moleclues. The density functional calculation for N_2 by Rytsölä et al represented the gradual onset of transition to the cluster state. It has been interpreted that this is due to the fact that the change in the free energy of the positron on cluster formation is small compared with thermal energy. Comparison between the density functional calculation and the present result provides further check of this calculation, particularly in the onset region of cluster formation. Contrasing to the picture of the cluster induced by the positron, Albrecht and Jones[20] interpreted their measurement of the behavior of λ_f in low-temperature gaseous Ar in terms of the positron interacting with existing atomic cluster, at least at low densities for which the time scale associated with atomic regroupings is much longer than the positron annihilation time. This mechanism might also be taken into consideration for the present gradual increase of Z_{eff} even at sufficiently low densities.

Although the constant Z_{eff} has not been found in the present density ranges, the lowest density values of Z_{eff} at 100–170K are obviously smaller than the constant value of $Z_{eff}\sim 40$ at 77 K and somewhat larger than $Z_{eff}\sim 30$ at room temperature. This agrees with the experimental result by Rytsölä et al. However, these are considerably larger than the recent theoretical Z_{eff} given in the work by Darewych[6], whose elestic cross section is in agreement with the recent measured values. More advanced theoretical studies are needed to resolve this discrepancy.

In summary, we have shown that the behavior of λ_f in low-temperature gaseous N_2 is not interpreted in terms of the positron interacting with a single N_2 molecule even at sufficiently low densities. Understanding of the behavior of low-energy positrons in N_2 at low densities is not satisfactory. More experiments at further lower densities are hoped. Experiments in low-density N_2 at low temperatures, which aim to the behavior of λ_f and the shoulder region, are now in progress.

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