

Temperature Dependence of Positron Annihilation Rates in Noble Gases

C. Kurz, R. G. Greaves, and C. M. Surko

Physics Department, University of California, San Diego, California 92093-0319

(Received 10 May 1996)

The dependence of positron annihilation rates on temperature has been measured for the noble gases. These measurements were carried out using a new technique which, for the first time, makes the positron energy range between $k_B T = 0.025$ and 1 eV accessible to experiments. Positrons in a Penning trap are heated to about 1 eV, and then cool to room temperature by collisions while maintaining a Maxwellian velocity distribution. The positron annihilation rate is measured during the cooling. Experimental data are in good agreement with theoretical calculations. [S0031-9007(96)01323-3]

PACS numbers: 36.10.Dr, 25.30.Hm, 71.60.+z, 78.70.Bj

The interaction of positrons with ordinary matter is of fundamental importance and has been the subject of numerous experimental and theoretical studies [1]. Furthermore, an understanding of such interactions is important for diverse technological applications such as the characterization of surfaces and solids using positron beams [2,3] and the development of models of the slowing down and annihilation of galactic positrons, which is the most intense γ -ray line of astronomical origin [4].

Traditionally positron annihilation cross sections have been measured using high-density gases or liquids, into which positrons are injected directly from a radioactive source [5]. Positron annihilation events are detected in delayed coincidence between a prompt γ ray (emitted by the radioactive nucleus at the same time as the positron) and the 511 keV γ ray produced when the positron annihilates. Positron annihilation rates can be measured at different positron temperatures by varying the test gas temperature and measuring the positron decay rate after they have thermally equilibrated with the test gas. In practice this limits the experimentally accessible temperature range to between 77 and 500 K ($k_B T = 0.007$ – 0.043 eV). At early times in the lifetime spectrum variations in the annihilation rate are observed and interpreted as being due to the dependence of the annihilation cross section on positron energy. While such data are in qualitative agreement with theoretical predictions of annihilation rates as a function of energy [5], they do not permit a quantitative analysis since the positron energy cannot be measured during the slowing down phase. Hence the interpretation of the experimental data is difficult and has to rely on theoretical models.

Positron beams have been used extensively to measure positron scattering cross sections [5]. Their application in annihilation rate measurements has not been successfully demonstrated, however, since annihilation cross sections are many orders of magnitude smaller than scattering cross sections. This fact, in conjunction with the short interaction time of beam positrons with the target, has precluded the use of beams from annihilation rate measurements.

For these reasons energies above 0.04 eV have not been explored experimentally until now, although theoretical

calculations of positron annihilation rates have been available for many years. They generally predict a drop in the annihilation rate with increasing temperature. For example, the polarized orbital model of McEachran *et al.* predicts that the annihilation rate for Xe drops by a factor of 5 if the positron temperature T_{e^+} increases from 0.025 to 0.5 eV. In contrast, the annihilation rate for He is predicted to change by only $\approx 10\%$ over the same temperature range.

The development of techniques for accumulating, storing, and manipulating positrons in Penning traps has permitted a new approach to positron-matter interactions. We have already applied this technique to the investigation of a number of topics relating to the interaction of positrons with large molecules [6] and the physics of electron-positron plasmas [7]. In this paper we describe a new technique using trapped positrons to study the dependence of positron annihilation rates on positron temperature. We present and discuss data obtained for the noble gases in the temperature range of 0.025–0.6 eV.

Positron annihilation rates, Γ , are conventionally expressed by the dimensionless parameter Z_{eff} relative to the Dirac rate for annihilation of positrons on a gas of uncorrelated electrons as $\Gamma = \pi r_0^2 c n Z_{\text{eff}}$, where r_0 is the classical electron radius, c is the speed of light, and n is the number density of the sample gas. For simple molecules, Z_{eff} is of the same order as the number of electrons in the molecule, Z , and is crudely interpreted as being the number of electrons in the molecule participating in the interaction. For more complex molecules, e.g., hydrocarbons, Z_{eff} exceeds Z by many orders of magnitude [6]. The simple interpretation of Z_{eff} no longer applies in this case, but the notation is still retained.

The experiment was performed in the multistage positron trap shown in Fig. 1. Operation of the trap, described in detail elsewhere [7], can be summarized briefly as follows. Positrons from a 50 mCi ^{22}Na source are moderated to a few eV by a solid neon moderator [8]. They are then magnetically guided into the four-stage trap, where they lose energy by inelastic collisions with a nitrogen buffer gas. Positrons accumulate in the fourth stage of the trap and cool to room temperature there

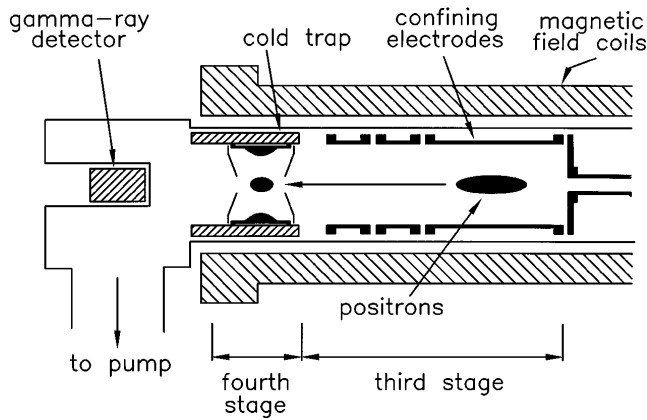


FIG. 1. Schematic diagram of the positron trap.

within ≈ 3 s. We can collect up to 10^8 positrons and store them for more than 1 h at a vacuum base pressure of $\sim 4 \times 10^{-10}$ torr, after the buffer gas is switched off. Typical test gas pressures vary between 10^{-6} torr for Xe to about 5×10^{-5} torr for He.

The experiment consists of repeated cycles of positron filling, heating, and annihilation. The fill cycle lasts for 25 s. The positron beam is then switched off, and the trapped positrons cool down to room temperature. Following this, the buffer gas is switched off and after a pump-out delay of 10 s the pressure has dropped from around 10^{-7} to about 10^{-9} torr. Subsequently the test gas is admitted into the trap region, allowing 8 s for the test gas pressure to stabilize. The positrons are then heated by applying a pulse of broadband RF noise to one of the confining electrodes ($\Delta f \approx 20$ MHz), which heats the positrons to 0.6–0.8 eV. We typically use RF pulses of approximately 10 ms duration with a peak-to-peak amplitude of about 50 mV. Attempts to increase the RF amplitude or duration above those levels do not lead to a corresponding increase in positron energies but rather begins to expel positrons from the trap. The expulsion mechanism is not understood at present, but it is the main factor that limits the maximum achievable temperature. In the case of the noble gases, the positrons cool by elastic collisions with the test gas and reach room temperature after typically 5–10 s. Concurrent with the cooling, the positrons annihilate on the test gas. Their annihilation rate is measured by a NaI(Tl) detector counting the annihilation γ rays (cf. Fig. 1). To correct for drifts in the positron filling rate, we measure the number of positrons remaining after the measurement cycle by dumping them onto an annihilation plate and adjusting the fill time accordingly for the next cycle. We acquire data in this manner for about 2–3 h, accumulating a total of approximately 5×10^5 counts. The background count rate is measured separately and is less than 10% of the signal level for the data presented here.

Before and after each run we measure the positron temperature, $T_{e^+}(t)$, as a function of elapsed time since the end of the heating pulse. This is accomplished by

reducing the depth of the confining well to zero and analyzing the number of positrons escaping the trap as a function of well depth. The effect of the changing space charge during the positron dump on the velocity distribution of positrons escaping the well has been discussed in detail elsewhere [9]. This effect can be neglected if only the first few percent of the dumped positrons are used to calculate the temperature [cf. inset to Fig. 2(a)].

The possibility exists that a strong temperature dependence of the positron annihilation rate could potentially lead to a non-Maxwellian positron energy distribution through preferential depletion of positrons with energies for which Z_{eff} of the test gas happens to be high. To estimate the impact of this effect on the positron velocity distribution, one should compare the thermal equilibration time within the positron cloud to the variation in the positron annihilation time over the investigated

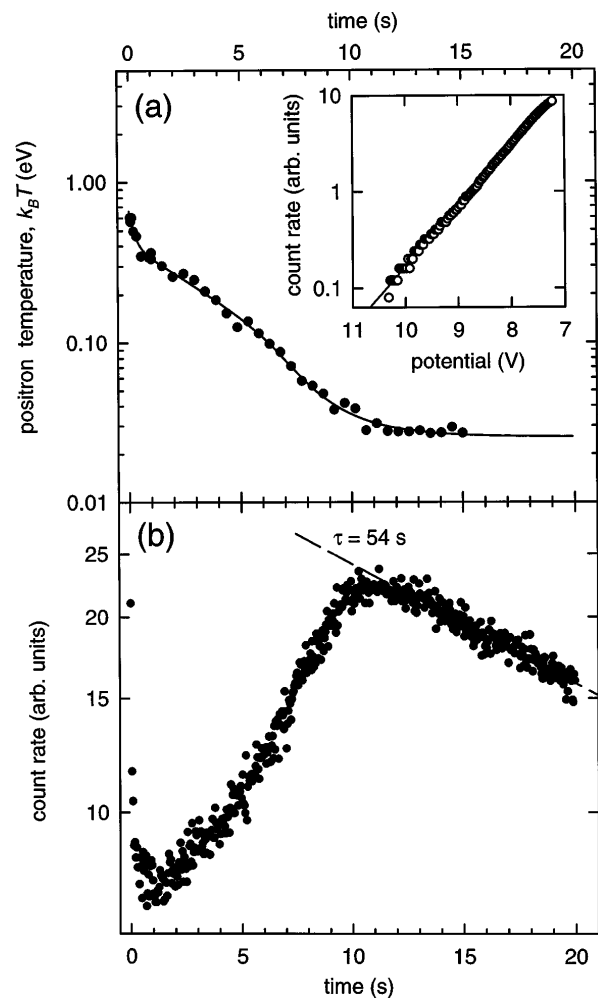


FIG. 2. (a) Positron cooling on xenon following a 5 ms pulse of RF noise. Inset: Energy distribution of positrons at $t = 1$ s. The data shown here represent only the first escaping positrons for which space charge effects are negligible. (b) Annihilation rate during the same time interval. Dashed line: exponential decay with $\tau = 54$ s.

temperature range. The collisional positron equilibration time is given by $\tau_{\text{eq}} = 1.5 \times 10^{10} T_{e^+}^{3/2} / n_{e^+}$ [10], where the positron temperature, T_{e^+} is specified in eV and the positron density, n_{e^+} , in m^{-3} . At $T_{e^+} = 0.5$ eV and a typical positron density in the trap of $n_{e^+} = 5 \times 10^{11} \text{ m}^{-3}$, we have $\tau_{\text{eq}} = 11$ ms. This should be compared to the largest variation in positron annihilation time, which occurs for Xe and is 250 s at $T_{e^+} = 0.3$ eV versus 50 s at $T_{e^+} = 0.025$ eV. Thus the equilibration time is more than 3 orders of magnitude shorter than both the annihilation time and the cooling time. Therefore the positron velocity distribution is not perceptibly influenced by the energy dependence of the annihilation cross section and remains Maxwellian throughout the entire cooling phase.

A full measurement of $T_{e^+}(t)$ comprises a number of individual dumps at different times during the cooling. The solid line in Fig. 2(a) is a fit to the experimental data, which is used to find the positron temperature at any arbitrary time after the heating pulse. Note that the shape of the full cooling curve is not a simple exponential decay, which presumably reflects variations in the elastic scattering cross section with positron energy.

Figure 2(b) shows the γ -ray count rate obtained during the cooling phase in Xe. This rate is proportional to the annihilation rate, $\dot{N}(t)$, where $N(t)$ is the number of positrons in the trap at time t . At later times, after the positrons have already cooled down to room temperature, the count rate shows an exponential decay, consistent with a constant value for Z_{eff} and a positron lifetime of 54 s. Earlier, however, when the positron temperature is still high, the annihilation rate is markedly lower. We interpret this as an effect of the temperature dependence of Z_{eff} . A comparison of Figs. 2(a) and 2(b) shows that the time when the positron temperature has reached room temperature coincides with the time when the γ -ray signal starts to follow a purely exponential decay. The fact that these two features coincide is a strong indication that the observed variations in the positron annihilation rate are indeed caused by changes in the positron temperature. In addition, good alignment confirms that the experimental conditions have remained stable during data acquisition.

As noted above, we have observed that when a certain threshold temperature is reached during RF heating, positrons are expelled from the confining well and annihilate on the walls. This continues even for a short time after the heating pulse has been switched off. The high annihilation rates observed for the first few data points in Fig. 2(b) are due to this effect, and all results presented in this paper have been truncated at this threshold temperature.

We obtain relative values of $Z_{\text{eff}}(t)$ from the raw data by normalizing the count rate at each time to the number of positrons present in the trap at that time,

$$Z_{\text{eff}}(t) \propto \frac{\dot{N}(t)}{N_0 - \int \dot{N}(t) dt}, \quad (1)$$

where the constant of integration, N_0 , represents the number of positrons in the trap at time $t = 0$. It is chosen so as to yield a constant value for $Z_{\text{eff}}(t)$ after the temperature has leveled off at room temperature. We obtain the temperature dependence of Z_{eff} by plotting $Z_{\text{eff}}(t)$ versus $T_{e^+}(t)$. To facilitate interpretation, the data shown in Fig. 3 have been accumulated into conveniently spaced temperature bins.

In Fig. 3 we present our results for the noble gases together with the theoretical predictions of McEachran *et al.* [11]. To facilitate comparison, the theory curves have been convolved with a Maxwellian distribution function and normalized to their values at room temperature. We observe a clear trend in the relative variation of Z_{eff} : whereas Z_{eff} for Xe drops by a factor of 5 over the investigated range of temperatures, the corresponding variations for the other noble gases continue to decrease with their atomic number; Z_{eff} for He and Ne varies only by about 10% over the same range. Within the scatter of our data, He and Ne exhibit the same temperature dependence of Z_{eff} . The agreement between theory and experiment is remarkably good over the entire temperature range. Small deviations are noticeable for Kr and Ar above about 0.15 eV, where the experimental data are consistently higher than the theoretical predictions. Recently Van Reeth *et al.* [12] have published new calculations for He, which, however, cannot be distinguished from the theory of McEachran at the present level of accuracy of the experiment.

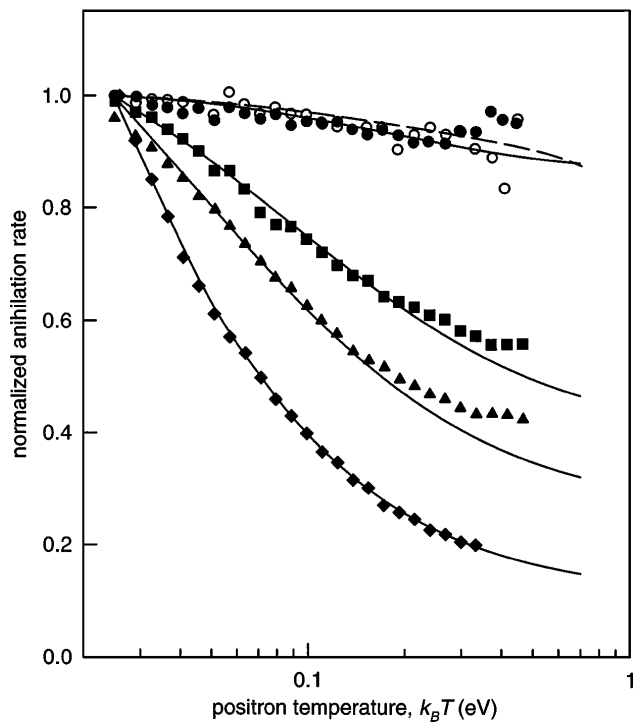


FIG. 3. Positron annihilation rates on the noble gases, normalized to unity at room temperature: \circ He, \bullet Ne, \blacksquare Ar, \blacktriangle Kr, \diamond Xe; theory calculations by McEachran *et al.* are denoted by solid lines for Ne, Ar, Kr, and Xe, and by the dashed line for He.

Lee *et al.* [13] have measured Z_{eff} for Ar by varying the temperature of the high-density test gas. Their results, again normalized to unity at room temperature, are in excellent agreement with our measurements in the temperature range over which the two experiments overlap (cf. Fig. 4). Wright *et al.* [14] have found strong evidence of clustering effects in Xe by studying the temperature dependence of the annihilation rate. For this reason, their measured temperature dependence cannot easily be compared with our results. Test gas densities for the measurements reported here are of the order of 10^{17} m^{-3} , thus making clustering effects negligible.

It is interesting to note that, although the theory of McEachran *et al.* compares well with the relative temperature dependence of Z_{eff} , there exists a discrepancy between the predicted and measured values of Z_{eff} at room temperature. The temperature averaged values calculated by McEachran *et al.* are 3.85, 7.0, 28, 58, and 217 for He, Ne, Ar, Kr, and Xe, respectively. Most noticeably for the heavier noble gases, the experimental values are significantly higher than these theoretical ones. For Xe the experimental data for Z_{eff} fall in the range between 300 and 400 [14–16], and for Kr they lie between 65 and 90 [6,14,17].

We see two avenues to extend the scope of these experiments. The cooling curves for the noble gases are not exponential, indicating that the cooling rate is energy dependent [cf. inset to Fig. 2(a)]. We expect to be able to use these time dependent measurements of the positron velocity distribution in the trap to extract information

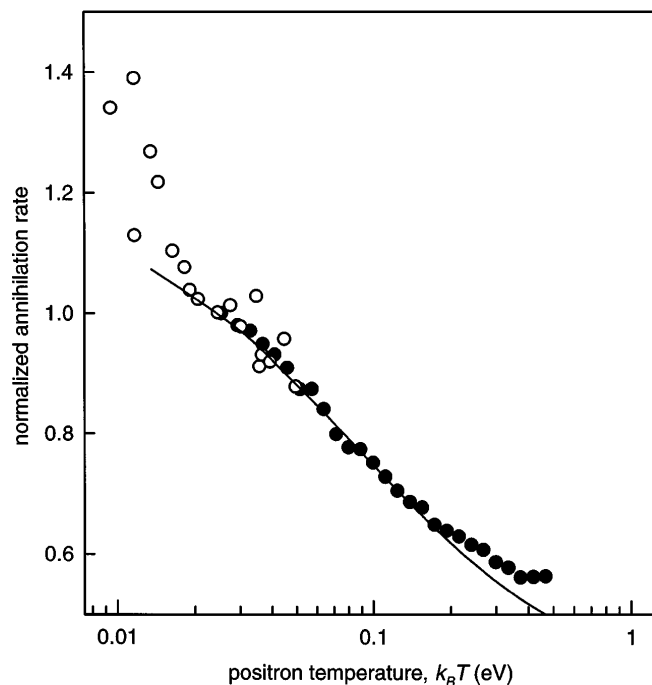


FIG. 4. Annihilation data for Ar, normalized to unity at room temperature. ● this experiment, ○ Lee *et al.* [13], theory denoted by solid line.

about the corresponding scattering cross sections as a function of energy. A second, natural extension of the experiment is to perform $Z_{\text{eff}}(T_{e^+})$ measurements on large organic molecules which have been shown to exhibit anomalously high values of Z_{eff} [6].

In summary, we have developed a new technique for measuring the energy dependence of positron annihilation rates in an important range of energies that was not previously accessible to experiment. Applying this technique to the noble gases, we find excellent agreement with the predictions of McEachran *et al.* and Van Reeth *et al.* regarding the relative variation of Z_{eff} with positron temperature. We expect to be able to extend this technique to a variety of other substances including hydrocarbon molecules. These experiments are likely to provide an important benchmark for theoretical calculations venturing beyond the noble gases and simple diatomic molecules calculated previously.

We thank K. Iwata, G. Gribakin, and M. Charlton for helpful comments, and E.A. Jerzewski for expert technical assistance. This research was supported by NSF Grant No. 9221283.

- [1] Proceedings of the XIX International Conference on the Physics of Electronic and Atom Collisions, Workshop on Positron Collisions, Can. J. Phys. (to be published).
- [2] P.J. Schultz and K.G. Lynn, Rev. Mod. Phys. **60**, 701 (1988).
- [3] A.P. Mills, Jr., Science **218**, 335 (1982).
- [4] R. Ramaty, M. Leventhal, K.W. Chan, and R.E. Lingens, Astrophys. J. Lett. **392**, L63 (1992).
- [5] T.C. Griffith and G.R. Heyland, Phys. Rep. **39**, 169 (1978).
- [6] K. Iwata *et al.*, Phys. Rev. A **51**, 473 (1995).
- [7] R.G. Greaves, M.D. Tinkle, and C.M. Surko, Phys. Plasmas **1**, 1439 (1994).
- [8] A.P. Mills, Jr. and E.M. Gullikson, Appl. Phys. Lett. **49**, 1121 (1986).
- [9] D.L. Eggleston *et al.*, Phys. Fluids B **4**, 3432 (1992).
- [10] K. Miyamoto, *Plasma Physics for Nuclear Fusion, Revised Edition* (The MIT Press, Cambridge, MA, and London, England, 1989), p. 96.
- [11] R.P. McEachran, D.L. Morgan, A.G. Ryman, and A.D. Stauffer, J. Phys. B **10**, 663 (1977); R.P. McEachran, A.G. Ryman, and A.D. Stauffer, *ibid.* **11**, 551 (1978); **12**, 1031 (1979); R.P. McEachran, A.D. Stauffer, and L.E.M. Campbell, *ibid.* **13**, 1281 (1979).
- [12] P.V. Reeth *et al.*, "Annihilation in Low Energy Positron-Helium Scattering," J. Phys. B (to be published).
- [13] G.F. Lee and G. Jones, Can. J. Phys. **52**, 17 (1974).
- [14] G.L. Wright, M. Charlton, T.C. Griffith, and G.R. Heyland, J. Phys. B **18**, 4327 (1985).
- [15] P.G. Coleman, T.C. Griffith, and G.R. Heyland, J. Phys. B **14**, 2509 (1981).
- [16] T.J. Murphy and C.M. Surko, J. Phys. B **23**, 727 (1990).
- [17] P.G. Coleman, T.C. Griffith, G.R. Heyland, and T.L. Killeen, J. Phys. B **8**, 1734 (1975).