Positron annihilation in low-temperature rare gases. II. Argon and neon

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Lifetime measurements of slow-positron and ortho-positronium (o-Ps) annihilation have been made in argon and neon gases at room temperature and below. The argon experiments cover the temperature range 115–300 K and the density range 0.0356–0.0726 g/cm^3 ($\simeq$20–40 amagat). The slow-positron spectra in argon exhibit a departure from free-positron annihilation below 200 K. The departure becomes more marked as the temperature is lowered. No deviation from free o-Ps pickoff annihilation is observed in argon at low temperatures. The neon measurements cover the temperature range 30–300 K and the density range 0.032–0.89 g/cm^3 ($\simeq$35–980 amagat). No effect of temperature on the slow-positron spectra throughout the temperature and density ranges investigated in neon is observed. The spectra are very exponential with a corresponding decay rate which is temperature as well as time independent and is directly proportional to density over the ranges investigated. The o-Ps data are more eventful in that the o-Ps lifetime at near-liquid densities is approximately 20 nsec, a factor of nearly 4 greater than the value obtained using the pickoff-annihilation coefficient obtained at lower densities. This is evidence for positronium-induced cavities in low-temperature neon. A brief discussion of the argon and neon results is given in the context of the explanations offered for the low-temperature effects observed in helium gas.

I. INTRODUCTION

The preceding paper dealt with large deviations from free-positron and ortho-positronium (o-Ps) annihilation for positrons in gaseous helium at low temperatures. The experiments described in this article were carried out to determine if similar effects would also occur for other rare gases, in this case argon and neon. In addition to investigating these gases at low temperatures, special care was taken to obtain measurements at room temperature with the lowest levels of impurities possible. This is of particular importance for the neon experiments since there have been no published lifetime spectra, except the low-purity data obtained by Osmon,[1] for positrons in neon. On the other hand, there have been several published lifetime experiments in argon gas where the mutual agreement between various investigators' data is very good, and hence the problem of gas purity is probably of less concern for this gas.[2–5]

All of the lifetime experiments to date in argon have dealt with the problem in interpreting the slow-positron component in terms of the annihilation and scattering cross sections for positron collisions with single argon atoms. Section III of this paper reviews the features of lifetime spectra which are accountable in terms of density-independent cross sections for positron–single-atom collision. This provides a point of reference from which the presentation and discussion of the data in Sec. IV and V can relate to the question of whether or not the low-temperature measurements indicate a deviation from the free-particle picture of slow-positron as well as o-Ps annihilation in the gases. The effects observed and conclusions drawn are summarized in Sec. VI.

II. EXPERIMENTAL PROCEDURE

The apparatus employed in the argon and neon experiments was the same as used for the helium measurements described in the previous paper. However, the methods of cooling the gas samples and precautions taken to ensure their purity levels differed from the helium experiments and warrant discussion here.

All of the argon measurements were carried out using the variable-temperature cryostat described in the helium paper. However, instead of cooling the sample chamber with a flow of cold helium vapor, it was found that the sample could easily be cooled to as low as 100 K by filling the cryostat with liquid nitrogen in the inner cryogen well and allowing a very slow rate of cold nitrogen vapor to flow past the sample. With the vapor flow completely turned off, the equilibrium temperature of the cryostat tail section was approximately 130 K. Thus, the additional amount of cooling supplied by the small flow of liquid nitrogen to the vaporizer block below the sample chamber served mainly to lower the temperature of the tail section surrounding the sample chamber rather than to cool the sample directly, as is more the case when liquid helium is the cryogen. Above 130 K, the cryostat was used in the "exchange gas mode"; i.e., there was no flow of cryogenic vapor past the sample chamber. This was much more convenient than trying to adjust the flow of nitrogen to the tail section in a way that would cool the tail section without allowing liquid nitrogen to

accumulate and surround the sample chamber. Regardless of the method of cooling, the balancing power supplied by the control unit to the heating coils on the sample chamber was less than 100 mW with a corresponding upper limit on the temperature differential between the top and the bottom of the sample chamber of 0.1 K.

The temperature range, 30–50 K, investigated in the neon experiments was covered using the same variable-temperature cryostat with liquid helium as the cryogen. Above 35 K, data could be taken with or without a flow of helium vapor past the sample. No change was observed in the data when changing from one method to the other. Most of the data was taken in the exchange-gas mode as a matter of expediency. Both the platinum and germanium resistance thermometers used in the helium experiments were tested by comparing the published vapor-pressure curves of liquid neon with measured vapor pressures as a function of temperature.[6] In this way we were able to verify the manufacturers' calibrations of the resistors to within an error of 0.25% over the temperature range 35–45 K. This accuracy was also established for the platinum thermometer over the range 95–130 K by measuring the vapor pressure of liquid argon in the sample chamber.[7]

Table I shows the impurity levels present in the argon and neon gas samples, as quoted by the supplier.[8] Before introducing a gas from the supply cylinder into the system, the system was evacuated for a period of several days with periodic flushing with high-purity helium gas. The method guaranteed that the only possible contamination that could have been introduced into the sample gas would have to be a result of outgasing of the walls of the copper sample chamber and stainless-steel-tubing wall surfaces. However, checks were made to determine that this source of contamination had no appreciable effect on the data. For example, the lifetime spectra taken in argon gas at room temperature showed no observable changes when obtained at the beginning and the end of a month period with the same initial charge of gas. In an attempt to obtain a more definite measure of the system's cleanliness, the neon-gas sample used in the experiments was recycled into the supply cylinder for the purpose of reanalysis. This was done by reducing the sample chamber temperature to 30 K and allowing the contents of the supply cylinder to condense into the sample chamber. The neon was then allowed to warm up to room temperature where it was held in the system for three days, after which time it was vented back into the supply cylinder. The amount of gas recycled this way represented about twice the amount of gas normally in the system. The supply cylinder was then disconnected from the system and sent to the supplier for mass-spectrographic analysis. The amount of impurities was found to be below the upper limits guaranteed by the supplier.

III. FREE-POSITRON DIFFUSION PICTURE

The usual picture of a free positron's behavior in the gas can be accounted for entirely in terms of the scattering and annihilation probabilities associated with the collision of a plane-wave positron with a *single* gas atom. Equivalently, the free-positron picture assumes that the positron spends more of its time in transit between collisions than in the vicinity of any one atom, and that the positron annihilation and scattering probability in a gas is proportional to the gas density. In order to apply this picture to the task of interpreting the lifetime spectra of slow positrons, we can express the annihilation activity of the hypothetical slow positron ensemble as follows:

$$A(t') = \pi r_0^2 c n_0 \int_0^{E_0} Z(E) G(E, t') dE \,. \tag{1}$$

$Z(E)$ is the overlap of the scattered-positron wave function of energy E with the electronic wave function of the target atom, and the constant term $\pi r_0^2 c$ is defined in the preceding paper.[9] $G(E, t')$ is the number of positrons, per unit energy, with energy E at time t'. What is particularly relevant to the free-positron picture, when using the above equation, is the assumption that n_0 is the average bulk-gas number density. We have also incorporated the approximation that the slow-positron ensemble is formed at $t' = 0$ with energies ranging from zero up to the lower limit E_0 for the positronium formation. The values of this threshold are 8.9 and 14.6 eV for argon and neon, respectively. The full diffusion equation for $G(E, t')$ is derived from the same formalism used in the classical Boltzmann transport equations, except with the extra inclusion of an annihilation term in the case of positrons.[10] Letting t' now be equal to $n_0 t$, where t is the real time, the diffusion equation can be written as

TABLE I. Upper limits on impurities (in parts per million) as quoted by supplier.[a]

Sample	N_2	O_2	CO_2 & CO	He	Ne	H_2O	THC[b]
Argon	1	0.5	1	•••	•••	1	0.5
Neon	4	1	1	10	•••	•••	0.5

[a] Impurities not listed explicitly are below the average detectability limits of 0.5 ppm.

[b] THC means total hydrocarbons (methane series).

$$\frac{\partial G}{\partial t'}(E, T') = \frac{(8m)^{1/2}}{M}\frac{\partial}{\partial E}\left[\left(E^{3/2} - \frac{kTE^{1/2}}{2}\right)\sigma(E)G(E, t') + kTE^{3/2}\sigma(E)\frac{\partial}{\partial E}G(E, t')\right] - Z(E)G(E, t'), \quad (2)$$

where $\sigma(E)$ is the momentum-transfer cross section for positron-atom elastic collisions and $\pi r_0^2 c$ has been set equal to unity.

The equilibrium, i.e., time-independent, decay rate of the positrons when they ultimately reach thermal equilibrium with the gas atoms is of particular importance. As thermal equilibrium is approached (2) becomes separable with $G(E, t') = g(E)\exp(-Z_e t')$, where $\lambda_e = Z_e n_0$ is the observed equilibrium decay rate when the spectra are plotted against real time. The equilibrium energy distribution $g(E)$ and decay constant Z_e are obtained by solving the following set of equations:

$$Z_e = \int_0^{E_0} g(E)Z(E)\,dE, \quad (3)$$

$$-\int_E^{\infty}[Z(E') - Z_e]g(E')\,dE' = \frac{(8mE)^{1/2}}{M}\sigma(E)\left[\left(E - \frac{kT}{2}\right)g(E) + kTE\frac{\partial g}{\partial E}(E)\right], \quad (4)$$

where it is understood that $g(E)$ vanishes before $E = E_0$. Therefore if a lifetime spectrum of slow positrons is to be interpreted in terms of the free-positron picture, the density independence of Z_e, or equivalently the direct proportionality of λ_e to the density, is a necessary condition to be met.

The above corollary is true, provided $\sigma(E)$ and $Z(E)$ are density independent. At densities where the interatomic distances are less than the range of the positron-atom interaction, it is not unreasonable to expect some density dependence for these quantities. At such densities, the presence of the neighboring atoms' repulsive cores would tend to shield or cut off the long-range polarization between the positron and each individual gas atom. This would result in a reduction of $Z(E)$ and $\sigma(E)$ at low (near thermal) values of E. The reduction of $Z(E)$ may be slight, but $\sigma(E)$ could exhibit a large decrease corresponding to an increase in n_0.[11] Even if we were to neglect the decrease in $Z(E)$, a decrease in Z_e would still be possible as a result of the reduction in $\sigma(E)$. This can be seen by taking into account the fact that $Z(E)$ increases with decreasing E at low energies and, hence, the left-hand side of (4) is greater than zero. In which case as $\sigma(E)/Z(E)$ decreases with increasing n, the most probable value of E [where $\partial g(E)/\partial E = 0$] increases slightly. This shift of the equilibrium distribution towards higher energies thus results in a decrease in Z_e. In summary: as the gas density is increased at a given temperature Z_e, *if anything*, would decrease for free positrons in a uniformly distributed gas. If the gas is not uniformly distributed, e.g. some of the atoms exist in clusters, this may not be true.

In the case of o-Ps, the annihilation rate is the direct sum of the self-annihilation rate, λ_{vac}, plus the "pick-off" annihilation rate λ_{po} resulting from annihilations between the positron of the o-Ps atom with an electron of the surrounding gas atoms. In principle, λ_{po} depends on the o-Ps kinetic energy and hence exhibits a time dependence which is subject to the same type of diffusion analysis outlined above for free-positron annihilation. At present, there has yet to be reported any appreciable deviation from exponential decay (i.e., a time-independent annihilation rate) of o-Ps for times greater than the time t_e at which the slow-positron component begins to vanish exponentially as well. For $t < t_e$ the o-Ps cannot be separated from the nonexponential shoulder region of the slow-positron component without invoking the assumption that the o-Ps component remains an exponential in this region. Thanks to a recent experiment by Bird and Jones,[12] we can now invoke this assumption with confidence when subtracting the o-Ps component from the total lifetime spectra for positrons in argon gas. Their experiment was able to demonstrate unambiguously that the o-Ps component is a single exponential, within normal statistical errors, throughout the spectra down to values of $n_0 t \simeq 10$ nsec amagat, nearly 40 times less that $n_0 t$ for slow positrons. Here the amagat density unit is understood to be equal to 2.69×10^{19} atoms per cm^3, the ideal-gas number density at 273.2 K and 1 atm pressure. Equivalently, the density can be expressed in terms of the mass density $\rho = Mn_0$, where M is the atomic mass of the gas. For argon, 1 amagat $= 1.78\times10^{-3}$ g/cm^3 and for neon, 1 amagat $= 9.01 \times 10^{-4}$ g/cm^3.

Because of the time independence of the o-Ps annihilation rate, it can be assumed that λ_{po} does not depend appreciably on the o-Ps kinetic energy at thermal energies and hence not on gas temperature as well, when the o-Ps is thermalized in the gas. With regard to the density dependence of free o-Ps annihilation rates, it should be kept in mind that the range of the interaction between the neutral o-Ps and gas atoms is shorter than the range of the positron–gas-atom interaction; and hence direct proportionality of λ_{po} to n_0 should be expected for free o-Ps.

IV. EXPERIMENTAL RESULTS

A. Argon

A series of lifetime spectra for slow positrons annihilating in argon gas at different temperatures, but at a fixed gas density, is shown in Fig. 1. By varying the range of allowable photomultiplier pulses corresponding to the detection of positron emission and annihilation, the full width at half-maximum (FWHM) time resolution of the timing equipment could be varied from 0.7 to 1.2 nsec. The argon spectra shown in the figure were obtained with a FWHM of 1.0 nsec. Spectra obtained at other densities are qualitatively similar in appearance, with the location of the peak in each spectrum being inversely proportional to density and exhibiting no appreciable temperature dependence. Figure 2 shows the values of λ_e and the corresponding Z_e plotted against temperature. An increase in temperature from 200 K to room temperature was found not to have a noticeable effect on the spectra, aside from the expected relatively small decrease in λ_e associated with free positrons thermalizing at higher energies as the temperature is increased.[3] At room temperature, the value of λ_e/ρ was found to be 5.3 ± 0.2 $\mu\text{sec}^{-1}\text{amagat}^{-1}$ as compared to the value 6.4 ± 0.2 $\mu\text{sec}^{-1}\text{amagat}^{-1}$ at 200 K. The spectra are also characterized by a shoulder width t_s. Using the convention of Paul and Leung,[13] that t_s is to be taken as the time required for the positrons to attain a decay rate equal to 90% of the equilibrium value λ_e, it is found that $\rho t_s = 350 \pm 30$ nsec amagat for each of the spectra obtained. This is in good agreement with others,[2-5] and further attests to the gas purity.

The o-Ps decay rates obtained from the argon lifetime spectra are shown in Fig. 3 along with the range of values obtained by other investigators. The low-temperature value shown by the open circles at each given density is the weighted average of all of the individual values obtained at the indicated density. This averaging was done since no systematic dependence of the decay rate in temperature was observed, nor did any of the measured values deviate from the plotted average by more than three times the error limits shown by the size of the plotted points.

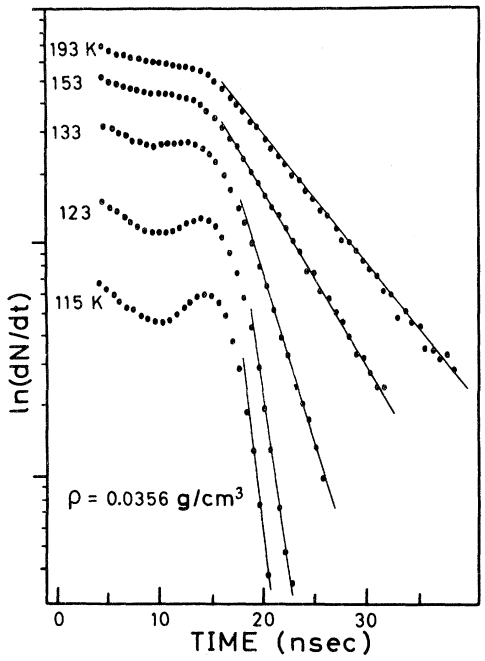

FIG. 1. Lifetime spectra of slow positrons in argon gas. For convenience, the spectra have been plotted with a common decade scale shown on the left. The actual counting statistics for each spectrum is roughly indicated by assigning 10^4 counts to the first plotted point in each spectrum.

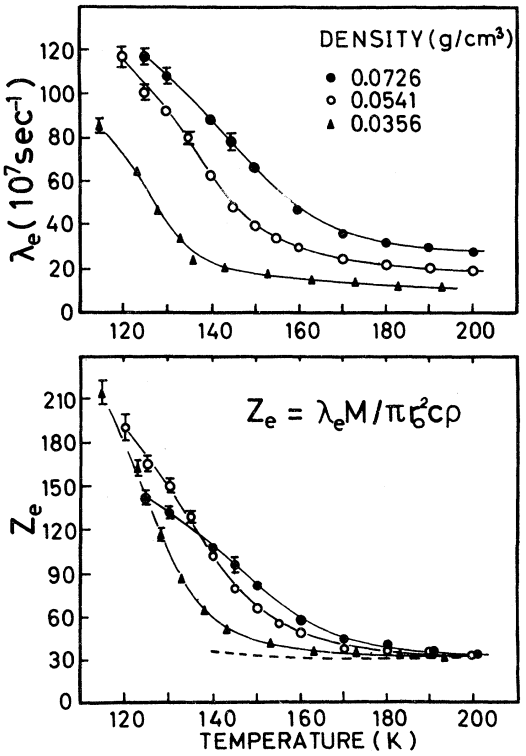

FIG. 2. Equilibrium decay rates and Z_e for slow positrons in argon gas. The mass density and atomic mass of the argon are denoted by ρ and M, respectively. The dashed line is a fit to the values obtained by Miller *et al.* over the low-density range 0.014–0.018 g/cm^3 where the decay rate is proportional to density (see Ref. 3). Error bars are omitted where they fall within the size of the plotted points.

B. Neon

Figure 4 is a lifetime spectrum for slow positrons annihilating in neon gas at room temperature. Aside from the presence of the prompt component, it is very difficult to discern any nonexponential region of the spectrum. Goldanskii claims to see a shoulder in his room-temperature spectra, ($\rho t_s = 500$–900 nsec amagat), but he states that it is considerably weaker than that which occurs in helium and is difficult to locate.[14] Since the helium and argon data exhibited unexpected properties at low temperatures and high densities, it was felt that the same thing should happen in neon gas. In an effort to maximize the densities while minimizing the temperature, spectra were also taken at temperatures above the critical point and, in particular, at 77.3 K. In contrast to the helium and argon results, no effect of temperature on the spectra could be found; i.e., the spectra were identical to the one shown in Fig. 4. The equilibrium decay rates are shown in Fig. 5. It is seen that λ_e is directly proportional to ρ and that λ_e/ρ, or equivalently Z_e, does not vary noticeably with temperature. A least-squares fit to the decay rates in the low-statistical-error region, 0.033–0.22 g/cm^3, yields $\lambda_e/\rho = 1.20 \pm 0.03$ μsec^{-1}amagat^{-1} which is in agreement with Goldanskii's room temperature value of 1.1 ± 0.1 μsec^{-1}amagat^{-1}. The insert in Fig. 5 shows that the values of λ_e, even at near-liquid densities, are in good agreement with the linear extrapolation of the fit to the lower-density points. On the assumption that the absence of low-temperature effects was perhaps due to it taking too long for positrons to reach low energies in neon, a few spectra were also taken with various amounts of helium gas added to the neon. Thus if the time required for positrons to reach low energies in neon was greater than the time range of our apparatus, the addition of helium should reduce it to within its range. With fractions of helium ranging from 5 to 80%, no peak or other marked features were observed in the spectra for temperatures as low as 28 K.

Figure 6 shows that values of o-Ps decay rate at various densities and temperatures. The pair of lines indicate the range of values obtained at room temperature by Goldanskii.

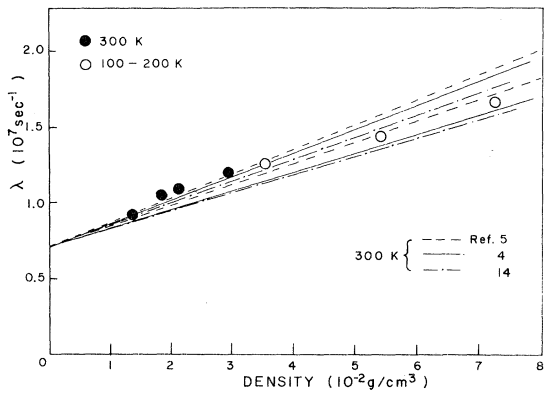

FIG. 3. Decay rates of o-Ps in argon gas. The error bars fall within the size of the plotted points. The straight lines are linear fits to other investigators' results with the constraint $\lambda = 0.71 \times 10^7$ sec^{-1} at $\rho = 0$.

V. DISCUSSION

A. Argon

The lifetime spectra of slow positrons annihilating in argon gas at low temperature are distinguished by the same type of large peak that is present in the low-temperature helium spectra. However, for argon there is no noticeable temperature dependence for the location of the peak. Thus if we identify the location of the peak as the time required for the bulk of the positrons to slow down to some energy E_R where $Z(E)$ under-

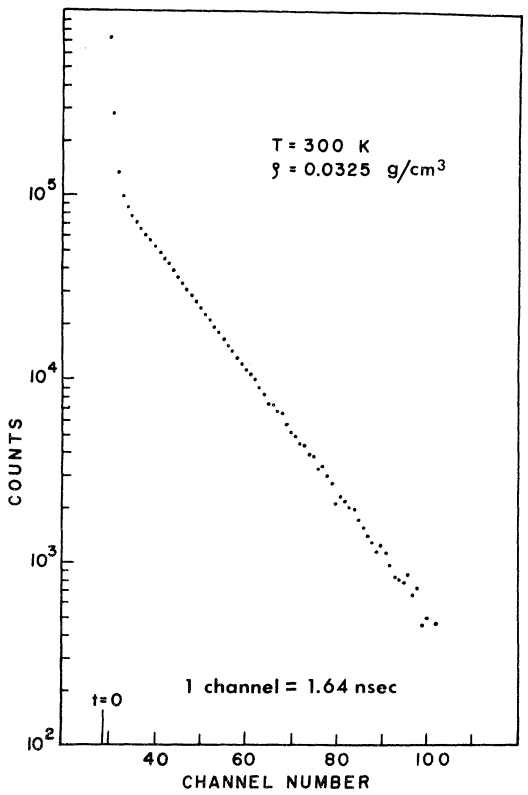

FIG. 4. Slow positron plus prompt-annihilation component of positron-lifetime spectrum in room-temperature neon gas. The FWHM resolution is 0.9 nsec.

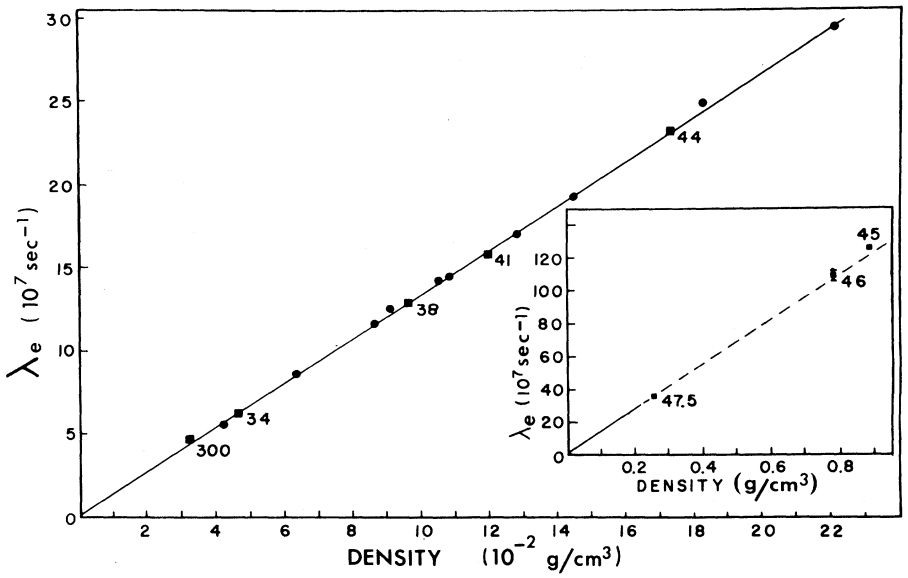

FIG. 5. Equilibrium decay rates of slow positrons in neon gas. The unlabeled points were obtained at 77.3 K. The square points were obtained at the temperatures indicated in units of degree Kelvin. Error bars, where omitted, fall within the size of the plotted points. The straight line is a least-squares fit to the decay rates over the density range $0-22\times10^{-2}$ g/cm^3. The dashed line in the high-density insert is a linear extrapolation of the fit to the lower-density points. The density of liquid neon at atmospheric pressure is 1.21 g/cm^3.

goes an abrupt enough increase to produce the peak, then E_R exhibits no apparent temperature dependence. Further we can also conclude that E_R is independent of density as well, because of the location of the peak is inversely proportional to gas density.

The density dependence of λ_e expected on the basis of the free positron picture differs very much from the experimental data. Although the sensitivity of the argon decay rates to changes in temperature and density is not as extreme as it is in the case of helium, we are nevertheless drawn to the conclusion that the argon decay rates at low temperatures and high densities are indicative of the positron becoming "trapped" into an environment of a higher-than-ambient gas density. An alternative to such an interpretation which would be in keeping with the free-positron picture would require the improbable situation that as E decreases, $Z(E)$ for positron–single-atom collisions exhibits a large increase with respect to decreasing interatomic spacing.

In view of the difficulty encountered in the preceding paper when trying to use the cluster model

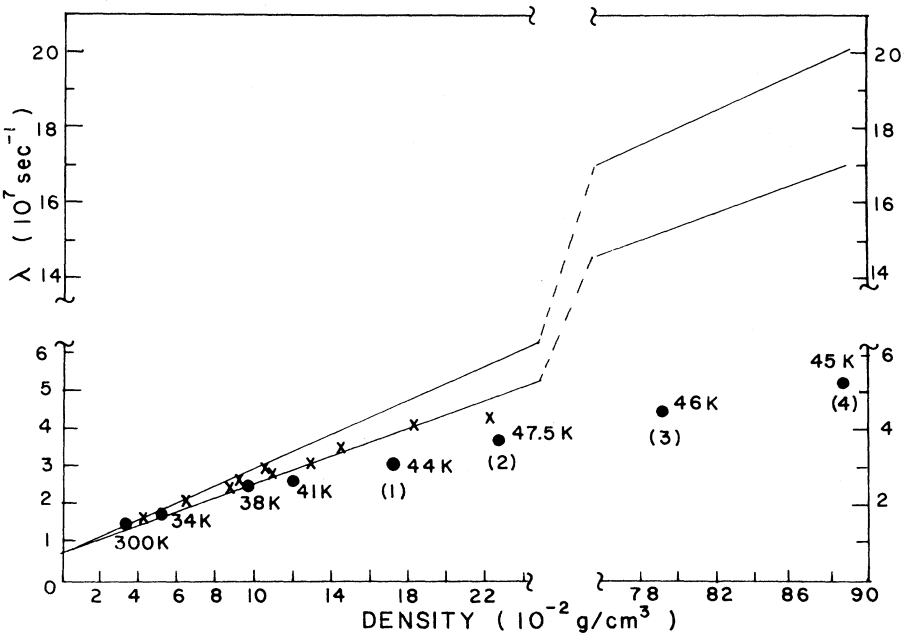

FIG. 6. Decay rates of o-Ps in neon gas. The crossed points were obtained at 77.3 K. The pair of lines bracket the upper and lower limits of the values obtained by using Goldanskii's pickoff annihilation coefficient measured at room temperature over the density range 0–0.26 g/cm^3 (Ref. 14).

to quantitatively account for the decay rates in helium, it would be of little advantage here to pursue the model to its limits in order to account for the argon data. It suffices to say that if we propose a similar electrostriction or condensation of argon atoms about a "localized" positron at low temperatures and high densities, then it is found that the reduced sensitivity of λ_e to changes in temperature, as compared with helium, is consistent with a model. The relevant quantity in the cluster model is α/T, the atomic polarizability divided by temperature. In the ideal-gas approximation, the gas pressure at a given distance from the center of the positron cluster varies exponentially with α/T.[15] Although the atomic polarizability of argon is about eight times that of helium, α/T is only one-third that of helium when T is taken to be 5–7 K for helium and 120–150 K for argon. Thus, the degree of electrostriction about a localized positron in argon should be less than for helium at the appropriate temperatures.

If we were to label the argon spectra "high-temperature" and "low-temperature spectra" when they were accountable in terms of the free-positron picture and when the free-positron picture was not adequate, respectively, we could not rely on the appearance of the spectra alone to make such a distinction. As seen in Fig. 1, the peak in the spectrum becomes weaker as the temperature is increased, because of the corresponding decrease in λ_e (or Z_e). At the same time, $(\partial Z_e/\partial\rho)_T$ decreases from a positive value towards zero, the limit required for the free-positron picture to hold.[16] Comparing Figs. 1 and 2, it is seen that at temperatures greater than $\simeq 170$ K the only way a spectrum could be distinguished as still being a low-temperature spectrum is by examining the vanishing density dependence of Z_e. In other words, the mechanism responsible for the peak in the low-temperature spectrum does not seem to vanish as the temperature is increased, but rather just results in a weaker bound state for the positron which gradually gives way to the positron being free. This behavior leads us to conjecture that the mechanism responsible for the shoulder terminus in the free-positron spectra could be the same mechanism which aids in the trapping of the positron into a high-density complex at low temperatures. An example of such a mechanism would be a virtual or a highly unstable positron–single-argon bound state[17] which is stabilized into a positron-multiatom complex at low temperatures and high densities.

Recent observations of diatomic concentrations in argon gas[18] raise the question of the temperature and density ranges that the argon gas itself is homogeneous enough for the free-positron picture to apply, quite apart from the additional problem of positron-induced complexes. This subject is worth future investigation to see if the low-temperature spectra are reflecting anomalies in the intrinsic gas structure.

Finally, it is important to note that the shoulder preceding the peak in the argon spectra becomes noticeably steeper as the temperature is decreased. This is interpreted as being due to the temperature dependence of λ_e being reflected into the region near $t=0$. The positrons which are initially close to (above and/or below) E_R can contribute steep components to earlier portions of the shoulder preceding the peak which is produced by the higher-energy positrons that take longer (~ 300 nsec amagat) to reach E_R. An attempt to confirm this explanation was made by subtracting a high-temperature spectrum from a low-temperature spectrum, both of which were normalized to equal areas and taken at the same density. The reasoning behind this was that the positrons which annihilate before reaching E_R or below should produce identical spectra at high and low temperatures. Therefore, when the total spectra are subtracted, the effects due to the lower-energy positrons being trapped at earlier times than the higher-energy positrons should become enhanced. This was indeed found to be the case. Unfortunately, the statistical error in the difference of counts per channel was very large, and the prompt components prevented any meaningful subtraction near $t=0$. What could be established, however, was that the subtraction produced a component in the shoulder region which was similar in appearance to the difference of the spectra in the region following the peak. Thus, there is a strong possibility that, under appropriately ideal experimental conditions, the subtraction of spectra could reveal the manner in which the initial distribution is manifested in the disparity in slowing-down times required for different energy groups of positrons to reach the bound state at low temperatures. Ultimately this could provide a good estimate of not only E_R, but $\sigma(E)$ as well.

The o-Ps decay rates exhibit much less unusual features and are probably representive of free o-Ps annihilation in the gas. Although the results are not good enough to convincingly establish the linearity of the decay rate versus density, they are in agreement with many investigators' ranges of room temperature values. Further, the fact that there is no temperature or hence pressure dependence at any one density, indicates the lack of an o-Ps induced cavity in the gas. If the Van der Waals attraction between o-Ps and argon

dominated over the repulsive spin-exchange, then we would expect that the o-Ps could not form a cavity in the gas. However, angular correlation measurements in *liquid* argon show that positronium-induced cavities are present at such high densities.[19] Thus, it is more probable that the absence of cavity formation here is due to the barrier height against adiabatic penetration of o-Ps into argon still being positive but not large enough at the gas densities investigated. For example at our highest density of 0.0726 g/cm^3, it can be shown that a barrier height slightly less than 1 eV would be insufficient to allow the zero-point pressure of the o-Ps inside a cavity to equal the ambient gas pressure of 14.9 atm, the lowest pressure possible to attain this density. But below condensation temperatures where the gas condenses to much higher densities at atmospheric pressure or less, a stable cavity would be possible. Future lifetime measurements of o-Ps lifetimes in liquid argon could confirm this by producing an anomalously long lifetime as is the case for helium.

B. Neon

There is no indication of the trapping of slow positrons in neon gas throughout the temperature and density ranges investigated. The proportionality of λ_e to ρ is preserved up to gas densities as high as 10^3 amagat and at the corresponding temperature 45 K, where the de Broglie wavelength of thermalized free positrons is more than 50 times the mean interatomic spacing. It is perhaps surprising to observe the annihilation rate conforming to the behavior predicted by the free-positron picture at such extreme temperatures and densities. A possible explanation of this result is that the positron-neon interaction is short range (< 10 Å) as opposed to being dominated by the long-range induced-polarization interaction at thermal energies.

Another "unusual" feature of the neon spectra, when compared with the high-temperature helium and argon spectra, is the lack of any appreciable deviation from exponential decay. Ordinarily, the lack of a shoulder in rare-gas lifetime spectra would imply the presence of polyatomic gas which thermalize the positrons within times close to $t=0$.[13] However the analysis of our gas purity as well as agreement between room-temperature spectra and spectra obtained at 30 K, where polyatomic impurities (except for H_2) would be frozen out, makes it difficult to believe that gas purity presents a problem here. Another implication of an exponential spectrum which is not *necessarily* true in this case is that $Z(E)$ is energy independent at near-thermal energies. It is possible, for example, to visualize $Z(E)$ changing slowly with energy over an energy region where the energy loss rate of the positrons, by coincidence, happens to be very small. This is indeed the case if we accept the annihilation and scattering cross sections calculated by Montgomery and LaBahn, which yield a calculated value of Z_e that is close to our measured value of 5.9 ± 0.1.[20] We have used the Montgomery-LaBahn (ML) cross sections in Paul's slowing-down approximation[2] of the full diffusion equation to produce calculated lifetime spectra, and find that the spectra exhibit only a very small deviation from exponential decay with no well-defined shoulder-vs-exponential region. The slowing-down approximation overestimates the rate of change of the instantaneous annihilation rate and hence, the full diffusion equation would produce even a more diffuse shoulder if the ML cross sections were used. This result emphasizes the role played by $\sigma(E)$ in determining the shape of the lifetime spectra since the $Z(E)$ calculated by ML has nearly the same energy dependence of the $Z(E)$ used by Leung and Paul[21] to produce good fits to the high-temperature helium spectra which have an easily discernable shoulder. However, unlike helium, the energy loss rate $[\sim\sigma(E)E^{3/2}]$ for positron-neon collisions shows no marked up-turn as the positrons slow down through the 2–0 eV energy region where $Z(E)$ undergoes most of its increase. This does not of course imply that the resulting lifetime spectrum is perfectly exponential. Nevertheless, such a behavior of the energy loss rate could account for the very diffuse, if not unidentifiable, shoulder in the neon spectra.

Although the slow-positron component of the lifetime spectra exhibits no bound-state behavior, the o-Ps decay rates shown in Fig. 6 serve as evidence that the trapping of o-Ps into zero-point cavities is possible in low-temperature neon gas at high densities. By extending the linear fit to Goldanskii's room-temperature o-Ps decay rates out to the highest density point, it is seen that the measured decay rate at this density is approximately four times less than the extrapolated value for free o-Ps in the bulk gas. At lower densities down to 0.16 g/cm^3, the o-Ps decay rates measured at temperatures less than 50 K are still significantly less than the values extrapolated from the room-temperature data. As in the case for the helium data, it should be possible to account for these low-temperature neon values using the simplified cavity model with the potential well depth V_0 of the cavity being the only adjustable parameter. Accordingly, we have applied the cavity model calculation, outlined in the pre-

ceding paper, to the neon o-Ps decay rates [labeled (1)–(4) in Fig. 6] and have obtained the following results: The low-density points at 0.173 and 0.228 g/cm^3 can be accounted for with V_0 equal to 0.119 ± 0.004 and 0.138 ± 0.003 eV, respectively. Calculated decay rates at the higher densities 0.792 and 0.887 g/cm^3 can be brought into agreement with the measured values with V_0 equal to 0.241 ± 0.015 and 0.259 ± 0.016 eV, respectively. The equilibrium cavity radius corresponding to each value of V_0 was calculated to be within the range 9.9–11.1 Å.

Clearly V_0 is not directly proportional to the gas density for the above results. This does not allow us to account for V_0 in terms of a unique value of the scattering length for o-Ps scattering by neon as was done in the case of helium, using the multiple scattering expression for V_0. Therefore, the claim that the neon data indicates cavity formation by o-Ps is not supported quantitatively by the simple cavity model. Aside from the possible shortcomings of the model at the densities and temperatures investigated, our analysis is handicapped by having to rely on the extrapolation of Goldanskii's room-temperature data to densities much greater than his highest-density point at 0.26 g/cm^3. It is thus desirable to obtain a less ambiguous determination of the free o-Ps annihilation rate at high densities in order to proceed beyond the above brief analysis of the data in the future. However such experiments will require a gas-handling system and a quantity of neon that is capable of attaining pressures on the order of a few hundred atmospheres at temperatures in the range 100–300 K.

VI. SUMMARY

As slow positrons in room-temperature argon gas approach thermal equilibrium, they undergo a marked increase in annihilation rate which gives rise to an easily discernible shoulder in the lifetime spectra. This increase can be attributed to the particular energy dependence of the scattering and annihilation cross sections for free-positron collisions with single argon atoms. However at lower temperatures and sufficiently high densities, the spectra exhibit a larger increase in annihilation rate which cannot be accounted for in terms of the free-positron picture. The interpretation given here of this effect is that the positrons become trapped into a complex where the positrons no longer interact freely with the gas atoms. It is further suggested that the initiation of this complex is related to the cause of the increase in annihilation rate in the room-temperature spectra. The o-Ps lifetime measurements do not indicate any significant departure from the density and temperature dependence expected for free o-Ps pick-off annihilation in gases. This does not however rule out the possibility of cavity formation by o-Ps at higher densities and lower temperatures.

The slow-positron lifetime spectra in neon gas are unusually well behaved. In addition to the spectra being very exponential, no departure from free-positron annihilation is observed throughout the temperature and wide density range investigated. This null effect may ultimately prove to be a most valuable check on any future theory offered as an explanation for the low-temperature phenomena in helium and argon gas. The o-Ps lifetimes in neon gas at near-liquid densities are much less than the values obtained by linearly extrapolating the lower-density measurements. Although this result is qualitatively consistent with the cavity model, quantitative verification of positronium-induced cavity formation in neon gas will have to await further experiments and perhaps more sophisticated treatments of the model.

In view of the varied behavior of the gases investigated, it is too early to generalize the low-temperature properties of positron annihilation in rare gases until the effects presented are investigated in more detail, as well as carrying out experiments in the remaining rare gases, krypton and xenon. As of this writing, some low-temperature measurements have already been made in krypton and xenon by T. C. Griffith's experimental positron group, using exchange-gas cooling. It is hoped that the analysis of the results and their publication will help clarify the picture.

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