resonance radiation should, therefore, be very useful for studying the magnetic and chemical states of very thin surface layers.

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Free-Positron Annihilation Mean Life in Diatomic and Rare Gases in Liquid and Solid States

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Measurements have been made on the free-positron annihilation mean life, τ_1, in H_2, D_2, He, N_2, O_2, Ne, Ar, and Xe in liquid and/or solid states. To a fair approximation, the observed mean lives may be represented by the equation: $\tau_1(\text{nsec})=0.31+0.15\times10^{-4}\ E_i^2/\rho_e$, where E_i is the first electron ionization potential in eV, and ρ_e is the number of outermost electrons per unit "atomic volmue" a_0^3. In the H_2 and D_2 isotopes where E_i is constant, τ_1 is linearly dependent on ρ_e^{-1}. It was also found that τ_1 is independent of the ortho-para ratio of the liquid H_2 at 20.4°K.

INTRODUCTION

In a previous report[1] it was shown that the observed free-position annihilation mean life, τ_1, has a linear dependence on the ratio of the first electron ionization potential to the outermost electron density, E_i/ρ_e, for liquid H_2, N_2, and O_2. It was noted, however, that the observed τ_1 in liquid He is 30% larger than the value expected from this relationship. In an effort to look for a consistent relationship among the values of τ_1 in rare and diatomic gases, the measurements were extended to other rare gases. This paper presents the results of a further investigation concerning the dependence of τ_1 in condensed elementary gases on E_i and ρ_e.

When positrons are used to study the electronic structure of solids, it must be remembered that the presence of positrons in the solid affects the electronic configuration. On the other hand, the positrons would cause the same perturbation in each member of a pair of isotopes. The use of H_2 and D_2 made possible an examination of the dependence of τ_1 on ρ_e at constant E_i. Measurements were also made on the τ_1 in liquid H_2 of different ortho-para ratios at constant temperature.

APPARATUS

"Research Grade" gaseous D_2, O_2, Ar, and Ne purchased from the Matheson Company were liquefied and/or solidified in a $1\frac{1}{8}$-in. diam by 5-in. evacuated stainless-steel chamber submerged in a cryogenic liquid bath of appropriate temperature. Gaseous Xe, also of "Research Grade," was solidified in a $\frac{1}{2}$-in. diam by 2-in. chamber submerged in a liquid-nitrogen bath. A small quantity of isotopically pure Na^{22} was sandwiched between 0.0001-in.-thick rubber-hydrochloride films, supported by an aluminum frame, and radially centered in each chamber.

The delayed-coincidence apparatus used in I was further improved by the addition of auxiliary electronics. The dewar system necessitated a 2-in. separation between the positron source and the front surface of each detector. A slow signal for energy discrimination was taken from the tenth dynode of each 56 AVP photomultiplier tube and routed to an amplifier-discriminator system. One slow discriminator was set to accept the upper 40% of the nuclear gamma-ray Compton spectrum, while another discriminator was set with a window to accept the upper 30% of the positron annihilation gamma-ray Compton spectrum. The fast anode pulse from each photomultiplier tube was limited by a tunnel diode (1N2939) and used to trigger a tunnel diode univibrator. The univibrator pulse was used to turn off an npn transistor (2N797) that was normally in saturation. The fast square positive pulses thus produced were sent to a time-to-pulse height converter (hereafter referred to as TPHC) of Simms' design.[2] The TPHC output was sent to a 400-channel pulse-height analyzer for gating and storage. The gating system used was similar in principle to that described by Schwarzchild,[3] in that it

[1] D. C. Liu and W. K. Roberts, Phys. Rev. **130**, 2322 (1963); hereinafter this is referred to as I.

[2] P. C. Simms, Rev. Sci. Instr. **32**, 894 (1961).

[3] A. Schwarzchild, Nucl. Instr. Methods **21**, 1 (1963).

provided the necessary energy discrimination and reduced the effect of pulse pileup. In addition, it reduced the chance-coincidence rate by a factor of two.

The time calibration of the TPHC was performed with various lengths of RG71/U cable. The integral linearity of the converter was good over a range of approximately 100 nsec. Its differential linearity was $\pm 10\%$ in the worst region. The differential linearity was examined with a pulse generator in conjunction with a sampling scope to generate a prompt and time-swept signal, respectively, for the trigger circuits. To assure that the poor differential linearity was not caused by the sampling scope circuitry, various lengths of RC71/U cable were inserted between the triggers and the converter. No change in the differential linearity was observed. It is felt that, for subnanosecond mean-life measurements, not only the integral linearity but the differential linearity of the TPHC must be known before one can be confident of the results. It was possible to choose a portion of the converter that has a $\pm 2\%$ differential linearity.

For positrons annihilating in aluminum, the delayed-coincidence system showed a full width at half-maximum of 0.9 nsec and a mean life of 0.26 nsec. By the generation of time spectra of various mean-life values with a measured prompt time spectrum of Co^{60}, it was found that any mean life that is 0.35 nsec or longer can be deduced accurately from the slope of the logarithmic count rate versus time delay plot.

RESULTS AND DISCUSSION

In Table I are summarized the results of all the τ_1 measurements made on four diatomic and four rare gases in liquid and/or solid states. The values of τ_1 in H_2, N_2, and O_2, which have been reported in I, were reproduced with the present instrumentation to within the indicated experimental errors. They are repeated here for completeness, since it is intended that the consistency among the values of τ_1 in all these condensed gases be discussed.

TABLE I. Free-positron annihilation mean life in condensed diatomic and rare gases.

Substance	T (°K)	τ_1 (nsec)	E_i[b] (eV)	ρ_e[c] ($10^{-3}a_0^{-3}$)	E_i^2/ρ_e [10^4 (eV)^2 a_0^3]
Liquid H_2	20.4	0.92 ±0.04[a]	15.6	6.29	3.87
Solid H_2	13	0.80 ±0.03[a]	15.6	7.53	3.23
Liquid D_2	20.4	0.83 ±0.03	15.6	7.49	3.24
Solid D_2	13	0.74 ±0.03	15.6	8.86	2.75
Liquid He	4.2	1.90 ±0.06[a]	24.5	5.57	10.75
Liquid N_2	77.3	0.56 ±0.02[a]	15.5	15.5	1.55
Solid N_2	62	0.48 ±0.02[a]	15.5	18.7	1.28
Liquid O_2	90.1	0.45 ±0.02[a]	12.5	25.5	0.61
Solid O_2	4.2	0.38 ±0.02	12.5	34.3	0.46
Solid Ne	20.4	0.43 ±0.03	21.5	38.9	1.19
Liquid Ar	86	0.50 ±0.02	15.7	19.0	1.30
Solid Ar	77.3	0.43 ±0.03	15.7	22.1	1.11
Solid Xe	77.3	0.40 ±0.03	12.1	14.9	0.98

[a] Values had been reported in I and were presently reproduced to within the indicated experimental errors.
[b] Values are taken from the *Handbook of Chemistry and Physics*, edited by Charles D. Hodgman (The Chemical Rubber Publishing Company, Cleveland, Ohio, 1962), 44th ed.
[c] See text for explanation.

The values of ρ_e, the outermost electron density, in these substances are given as the number of electrons per unit "atomic volume," which is a_0^3, where a_0 is the Bohr radius. A word should be added about the outermost electrons per atom as interpreted for the present purpose. The s state electrons are more strongly bound to the atom because they penetrate more closely to the nucleus than do the corresponding p state electrons of the same principal quantum number. For example, x-ray spectra of Ar show an energy difference of 24 eV between M_I and the (M_{II},M_{III}) doublets.[4] It is reasonable to assume that the s state electrons are part of the core electrons and the annihilation process involves principally the outermost p state electrons. In the calculation of ρ_e, therefore, the number of electrons per atom were taken as follows: 1 for H_2 and D_2, 2 for He, 3 for N_2, 4 for O_2, and 6 for Ar, Ne, and Xe.

The plot of τ_1 versus ρ_e^{-1} in Fig. 1 is of interest, although a quantitative correlation is not expected since the values of ρ_e, estimated by smearing the outermost electrons over the molar volume, do not describe adequately the actual electron density and the potential experienced by the positron. This is evident by the observed τ_1 in liquid He, which is twice that expected if a consistency exists among the values of mean life in all condensed gases. Moreover, the values of τ_1 in liquid O_2 and solid Ne, Ar, and Xe are approximately constant, although the values of ρ_e for these substances differ quite considerably. On the other hand, the values for τ_1 in liquid and solid H_2 and D_2 fall on a straight line. This indicates that for an isotopic pair, where the interaction between the positron and each member of the pair is essentially the same, τ_1 depends linearly on ρ_e^{-1}. Furthermore, it is seen that the values of τ_1 in the ele-

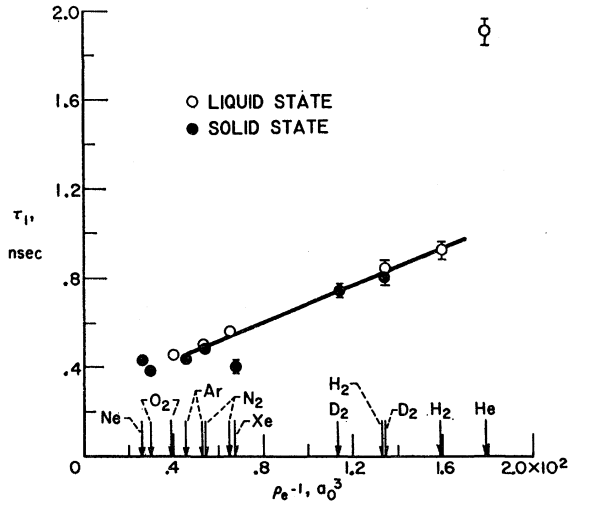

FIG. 1. A test of the τ_1 in condensed elementary gases versus ρ_e^{-1}. The line is drawn to show the agreement among materials that have essentially the same E_i.

[4] R. D. Hill, E. L. Church, and J. W. Mihelich, Rev. Sci. Instr. 23, 523 (1952). (See table of critical x-ray absorption energies.)

ments Ar and N_2, which have essentially the same first electron ionization potential E_i, as the molecular H_2 and D_2 (Table I), also fall on the line extrapolated from the hydrogen isotopes data. These experimental facts imply that τ_1, in addition to being a function of ρ_e, is dependent on E_i.

For obvious physical reasons, the positron-electron interaction, whether it causes electronic polarization or results in electron capture, should depend on how tightly the electrons are bound to the atom or molecule. Figure 2 shows a test of the τ_1 dependence on E_i/ρ_e. A good fit exists only among the values of τ_1 in the diatomic gases and Ar. This correlation had been reported earlier in I when we had only the results of liquid H_2, N_2, and O_2. The fit is probably fortuitous because, with the exception of O_2, all have nearly the same E_i and thus should fall on a straight line as in Fig. 1, where τ_1 is plotted against ρ_e^{-1}.

Figure 3 shows the test of τ_1 versus E_i^2/ρ_e. The line drawn is the result of a least-square fit of all the data points. It is seen that to a fair approximation the mean life may be expressed by the equation of this line

$$\tau_1(\text{nsec})=0.31+0.15\times10^{-4}E_i^2/\rho_e.$$

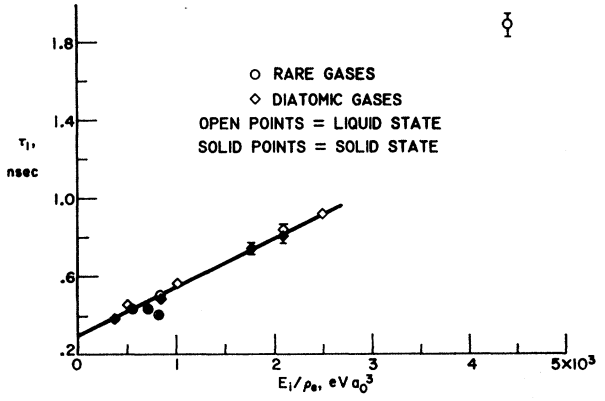

FIG. 2. A test of the τ_1 dependence on E_i/ρ_e. The line drawn is a best fit for the diamond points only.

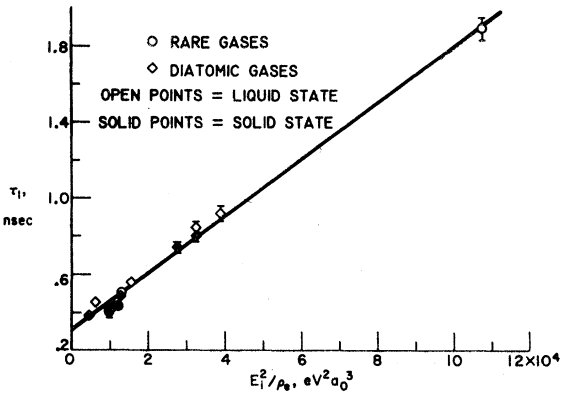

FIG. 3. A test of the τ_1 dependence on E_i^2/ρ_e. The line drawn is a least-square fit through all data points.

The intercept at the τ_1 axis, where E_i is zero, has a value of 0.31 nsec. This value may be thought of as the positron mean life in a plasma in which the electrons are not bound, so that no work is required of the positron to capture an electron. It agrees well with the average of mean-life values observed in alkali metals[5] where the conduction electrons may be approximated as a free-electron gas.

The values of τ_1 in liquid H_2 of various ortho-para ratios were also measured. Starting with the 75/25 ortho-para percent-ratio liquid at 20.4°K, τ_1 was measured as a function of time for 135 h at constant temperature. It was found that, within the experimental error limits of $\pm5\%$, the values of mean life in liquid H_2 of various ortho-para percent ratios, in the range from 75/25 to 35/65, are constant and the same as that observed for the 99% para-H_2 liquid, which was catalytically liquefied and aged at 20.4°K.

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[5] R. E. Bell and M. H. Jørgensen, Can. J. Phys. 38, 652 (1960).