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Positron Annihilation in Nitric Oxide and Nitrogen Dioxide*

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Positron-annihilation lifetimes in nitrogen dioxide and nitric oxide gases are carefully determined. Nitrogen is used as a dilutant. The free-positron-annihilation rate in units of the effective number of electrons per molecule $Z_{\rm eff}$ for nitrogen dioxide as a mixture of dimers and monomers is determined to be 720 to 1090 and the free-positron-annihilation rate for nitric oxide is $Z_{\rm eff}=34$. The positronium quenching rate in nitrogen dioxide gas is very high and the conversion reaction by NO_2 is found to be mainly responsible for the quenching. The quenching rate is determined to be $460D$ nsec^{-1} amagat^{-1}, where D is the partial density. The positronium quenching rate in nitric oxide is determined to be only about $0.15D$ nsec^{-1} amagat^{-1}, and it changes with the concentration of nitrogen.

I. INTRODUCTION

When energetic positrons are stopped in a gaseous medium, their energy is rapidly moderated via collisions to energies of about tens of electron volts.[1] Below the ionization potential of the medium, a positron may capture an electron during the collision and form an atom, positronium (Ps). The kinetic energies possessed by positrons which are able to form stable Ps atoms range from the ionization potential of the medium I_A to I_A-I_P, where I_P is the ionization potential of Ps, 6.8 eV. Since for most of the gases $I_A > I_P$, this range is, in general, above the thermal energy. Positrons falling below this range are no longer able to form Ps.

Certainly, positrons may annihilate with one of the electrons in the medium atom or molecule during the collisions. This type of annihilation is called either "free" or "direct" annihilation. If the electrons in the medium molecules are assumed to be free electrons, the free-positron-annihilation rate λ_{e^+} should follow the rate predicted by the Dirac theory.[2] This rate is $NZ\pi r_0^2 c$, where N is the number of molecules in a unit volume, Z is the number of electrons per molecules, r_0 is the classical radius of the electron, and c is the speed of light. In many gases the free-positron-annihilation rates have been found to be higher than the rate predicted by the Dirac theory.[3] In addition, the free-positron-annihilation rates have been found to be energy dependent.[3,4] Usually, the free-positron-annihilation rate is expressed in a formula

$$\lambda_{e^+}=NZ_{\rm eff}(E)\pi r_0^2 c\ ,$$

where $Z_{\rm eff}(E)$ is a convenient parameter. Obviously, the values of λ_{e^+} are density dependent.

There are two ground states of the Ps atom. Para-positronium (p-Ps) has total spin $S=0$, is a singlet, annihilates with a mean life of 0.125 nsec in free space, and produces two colinear γ rays. This mean life is too short to be directly resolved in the positron-lifetime spectrum. Ortho-positronium (o-Ps) has total spin $S=1$, is a triplet, annihilates with a mean life of 140 nsec in free space, and produces three coplanar γ rays. The normal ratio of o-Ps to p-Ps produced is 3.

In the presence of matter, the mean life of triplet or o-Ps is drastically shortened from its vacuum value of 140 nsec by quenching processes. Before self-annihilation occurs, the o-Ps atom makes many collisions with surrounding molecules. In these collisions, the positron bound in o-Ps may annihilate with a target electron of opposite spin. This process, called pickoff quenching, is present in all substances. In paramagnetic gases, such as O_2, NO, and NO_2, another quenching process, conversion quenching,[5] may also take place. Collisions with a molecule containing an unpaired electron can convert the o-Ps to p-Ps with subsequent rapid annihilation. In addition, chemical reaction or oxidation may occur, and the mean life of o-Ps will also be quenched by this process.[6]

Abid and Paul[7] investigated the positron-annihilation rate in nitric oxide (NO) at pressures of a few atomospheres and found a quadratic dependence of λ_{e^+} on pressure and a $Z_{\rm eff}$ that ranged from 2 to 4 times Z (30). Several other investigators have studied the quenching of o-Ps by NO.[8–11] However, no accurate values have been given. The quenching rate for small amounts of this gas was found to be so large that it was often used to suppress the long-lifetime component owing to o-Ps annihilation in these gases. For nitrogen dioxide (NO_2), the only published data for the free-annihilation rate are that of Osmon[8] who estimated that λ_{e^+} is greater than 0.3 nsec^{-1} atm^{-1} for pressures of a few atmospheres. This rate corresponds to $Z_{\rm eff} \simeq 1500$. No accurate value for the quenching rate of NO_2 has been reported.

It was the aim of this investigation to systematically study the free-annihilation and o-Ps quenching rates in NO and NO_2 as a function of density accurately, by measuring variations in the positron-lifetime spectra of these gases with a delayed-coincidence system of good time resolution. Since the positron-annihilation rates (particularly the *o*-Ps-annihilation rate) in both NO and NO_2 are as great as shown in this report, the accurate determination of the values of these rates requires the densities of the gases to be reduced to appropriate pressures less than the atmospheric pressure. However, at such low densities the stopping power of the gas is so weak that most of the positrons emitted by the Na^{22} source will hit the wall of the vessel, annihilating there instead of in the gas. For example, it was found by us that only about 20% of positrons emitted by Na^{22} are stopped in NO at 3 atm and 5% are stopped at 1 atm in the gas vessel used. At lower pressures the fraction of positrons stopped in the gas will be less. Therefore, a carrier gas must be introduced in order to provide the necessary stopping power to stop the positrons in the gases. Nitrogen gas was used because the positron-annihilation rates in nitrogen have been determined accurately[4]; it is relatively inert towards positron annihilation, and the slowing down of positrons in it is relatively fast.

For a detailed description of positron annihilation, the reader is referred to several recent review articles.[3,12–14]

II. EXPERIMENTAL

To accurately measure the range of positron lifetimes observed in the oxides of nitrogen requires an exceptionally flexible delayed-coincidence system. Because of the strong quenching effect of these gases, the positron lifetimes are very short for partial pressures $\simeq 1$ atm. On the other hand, the lifetime of *o*-Ps for low concentrations of the quenching gases mixed with N_2 is of the order of 100 nsec. In order to determine the entire range of lifetimes with the same accuracy, a delayed-coincidence apparatus used for previous works[4,15] was again used here.

The gas-handling system, constructed of stainless steel, is grease free, with all joints either welded or sealed with metal packing. The sample chamber itself is a spherical stainless-steel sphere 5 in. in diameter with its inside surface polished.[4] Gas pressure in the sample chamber was measured with a gauge calibrated to an accuracy of better than 1%.

The gas-handling system and sample chamber were filled with J. T. Baker or Matheson prepurified N_2 (minimum stated purity 99.997% and an O_2 content of less than 2 ppm) to a pressure of about 400 lb/in.^{2}. It was then evacuated to a pressure of a few microns. The cycle of flushing and evacuation was repeated several times. After the final evacuation, the system and chamber were filled with one of the oxides of nitrogen. Both NO (minimum stated purity 99%) and NO_2 (minimum stated purity 99.5%) were obtained from the J. T. Baker Chemical Co. No further purification of NO_2 gas was carried out. The purification of NO gas will be described in Sec. III.

The range of density of pure NO studied was about 2–7 amagats. Only one lifetime spectrum was recorded for pure nitrogen dioxide corresponding to a density of 3.4 g/liter. Higher pressures and densities could not be reached at room temperature, as the boiling point of nitrogen dioxide is 21.2 °C.

At the lowest density of each pure gas studied, N_2 was introduced into the chamber to raise the total pressure to about 400 lb/in.^{2} A series of measurements was then taken at decreasing pressures until the total system pressure was about 100 lb/in.^{2} The dilution process was repeated until the *o*-Ps lifetime approached that for pure N_2 over the given range of total pressures.

The experimental-lifetime spectra measured typically consisted of three resolvable components plus a background. The shortest-lived component ($\lambda_1 \sim 3.0\ \text{nsec}^{-1}$) contained at least two unresolved components: the annihilation of positrons bound in *p*-Ps and positrons annihilating in the walls of the container. The two longer-lived components are attributed to the annihilation of free positrons and to the quenching of *o*-Ps.

The data were analyzed by using a standard iterative least-squares technique.[16] A typical annihilation-lifetime spectrum resolved into two components and background is shown in Fig. 1. The detailed data-reduction process varies slightly according to the pattern of the annihilation-lifetime spectrum.

III. RESULTS AND DISCUSSION

A. Nitrogen Dioxide

As nitrogen dioxide boils at 21.2 °C, just below the room temperature 22 °C, only one lifetime spectrum was taken in the pure gas at the saturated-vapor pressure at 22 °C. The density of the gas is 3.4 g/liter at this temperature.[17] This density is far greater than the density if we assume all the nitrogen dioxide molecules to be monomers. Actually there are more dimers than monomers under this condition. The dissociation of the dimers to monomers,

$$N_2O_4 \rightleftarrows 2NO_2 , \qquad (1)$$

is pressure dependent. From the density of the gas and assuming both the dimers and monomers to be nearly ideal gases, the equilibrium constant

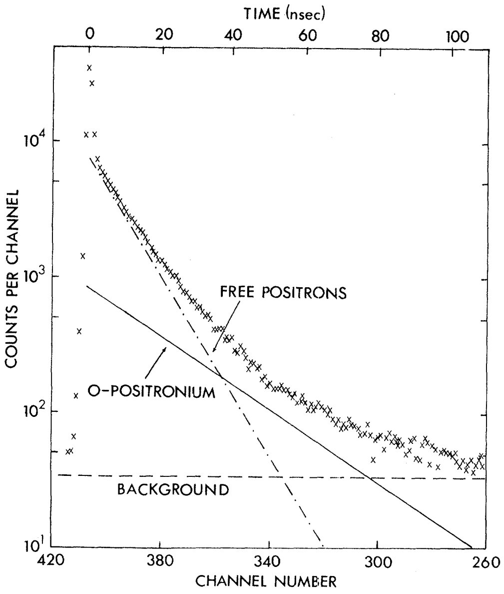

FIG. 1. Positron-annihilation-lifetime spectrum in a nitrogen dioxide and nitrogen mixture; NO_2, 6.4×10^{-4} amagat; N_2, 16.9 amagats; $\tau_2=9.62$ nsec, $\tau_3=24.2$ nsec.

is found to be

$$K_P=4\alpha^2P/(1-\alpha^2)=0.0618\text{ atm}, \tag{2}$$

where P is the total pressure and α the fraction of the dimers dissociated. The densities of the monomers NO_2 and the dimers N_2O_4 at various pressures can be calculated by using the above formula.

At the density of the saturated vapor (3.4 g/liter) there is only one long-lifetime component with a mean life of 5.9 ± 0.2 nsec in the annihilation-lifetime spectrum. This first long-lifetime component remains there as the density of the nitrogen dioxide gas is diluted by an "inert" gas, nitrogen. No separable second long-lifetime component with mean life greater than 0.6 nsec appears until the density of the nitrogen dioxide gas is reduced to less than 0.1 g/liter. At this density (0.1 g/liter) the fraction of N_2O_4 dissociated is about 60%. We shall use "nitrogen dioxide gas" to represent the gas with both monomers and dimers in equilibrium.

At the above density of the nitrogen dioxide gas or less, the mean life of the first long-lifetime component for the gas mixture approaches the mean life of the nearly thermalized free-positron annihilation in pure nitrogen and does not change appreciably with the density of the nitrogen dioxide gas. However, the mean life of the second long-lifetime component changes with the density of the nitrogen dioxide gas. Therefore, the first long-lifetime component is attributed to the free annihilation of positrons in the gas mixtures and the second long-lifetime component to the annihilation of *o*-Ps and *p*-Ps.

1. *Free-Annihilation Rate*

The free-annihilation rate of the positrons in nitrogen dioxide gas, (i.e., the annihilation rate of the first long-lifetime component subtracted from the free-annihilation rate of nearly thermolized positrons in pure nitrogen,[4] $5.80D$ μsec^{-1}amagat^{-1}, with the partial density D of nitrogen in the gas mixture) is plotted against the total density of the nitrogen dioxide gas in Fig. 2. No specific relationship between the free-annihilation rate and either the pure monomers or the pure dimers can be found. Therefore, only a combined rate will be given here.

It appears in Fig. 2 that the rate in the gas mixtures with high densities of nitrogen is higher than in those with lower densities of nitrogen. This can be explained by the fact that the positrons are not fully thermalized during their annihilation. The rate for the nearly thermalized positrons (in high densities of nitrogen) is slightly higher than the partially thermalized ones (in lower densities of nitrogen).

The free-annihilation rate of the nearly thermalized positrons is calculated to be

$$\lambda_{e^+}=(0.065\pm0.006)d\text{ nsec}^{-1}\text{g}^{-1}\text{liter}\ , \tag{3}$$

with d being the density in g/liter. The free-annihilation rate of the partially thermalized positrons is calculated to be

$$\lambda_{e^+}=(0.048\pm0.003)d\text{ nsec}^{-1}\text{g}^{-1}\text{liter}\ . \tag{4}$$

This is also very close to the rate in pure nitrogen dioxide vapor at 22 °C without nitrogen as a dilutant.

If we want to express these two values in units of Z_{eff}, the values of the Z_{eff} for the nitrogen diox-

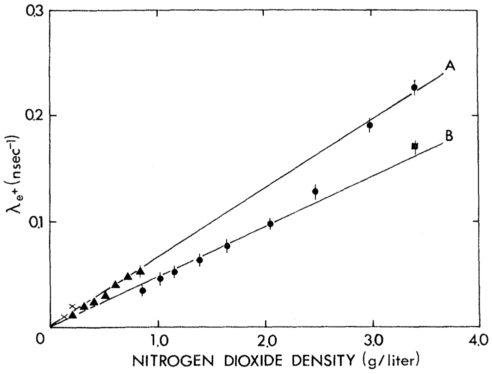

FIG. 2. Free-positron-annihilation rate in nitrogen dioxide (mixtures of monomers and dimers); square, nitrogen dioxide; dots, 3.8% volume nitrogen dioxide in nitrogen; triangles, 1.0% volume nitrogen dioxide in nitrogen; crosses, 0.28% volume nitrogen dioxide in nitrogen.

ide gas with dimers and monomers in equilibrium are 1090 and 720, respectively. These values are slightly lower than the one reported by Osman.[8] Since these values of Z_{eff} are much higher than the numbers of electrons either in NO_2 (23) or in N_2O_4 (46), the attractive interaction between the positron and electrons in either NO_2 or N_2O_4, or both, must be very strong during the collisions. A metastable collision complex of e^+NO_2 may exist during the collisions from either the reaction

$$e^+ + NO_2 \rightarrow e^+NO_2 \tag{5}$$

or

$$e^+ + N_2O_4 \rightarrow e^+NO_2 + NO_2 \tag{6}$$

or both.

2. *Positronium Annihilation*

The quenching rate of *o*-Ps in the nitrogen dioxide gas is very high. Therefore, the quenching rate can only be determined with the presence of a large amount of an inert gas (nitrogen). The second long-lifetime component which is attributed to Ps annihilation does not appear until the density of the nitrogen dioxide gas is less than 0.1 g/liter. We can either plot the quenching rate against the total density of the nitrogen dioxide gas, the density of the dimers only, or the density of the monomers only. It is found that the plot against the density of the monomers only fits best to the theoretical curve. This plot is shown in Fig. 3. For the sake of clarity a log-log scale is used.

The solid curve in Fig. 3 represents the theoretical quenching rate if the quenching is due to the conversion reaction[6]

$$o\text{-Ps} + NO_2 \cdot \rightleftarrows p\text{-Ps} + NO_2 \ . \tag{7}$$

The dashed curve represents the theoretical quenching rate if the quenching is due to chemical reaction[6]

$$o\text{-Ps} + NO_2 \rightarrow PsNO_2, \tag{8}$$

where the annihilation rate of the Ps compound is assumed to be 2.0 nsec^{-1} or slightly more. Obviously, the experimental data, particularly for densities higher than 10^{-4} amagats, fit the solid curve much better than the dashed one. This indicates that the conversion reaction (7) is the responsible quenching process. This is a very reasonable explanation, because NO_2 is a known free radical with an unpaired electron. This also explains why the plot against the total density of both the monomers and dimers does not fit so well to the theoretical curve; the data for higher densities will be on the far right-hand side of the theoretical curve. The dimer N_2O_4 has no unpaired electron and is not expected to quench the Ps by conversion.

The quenching rate which is used to plot the solid line in Fig. 3 is

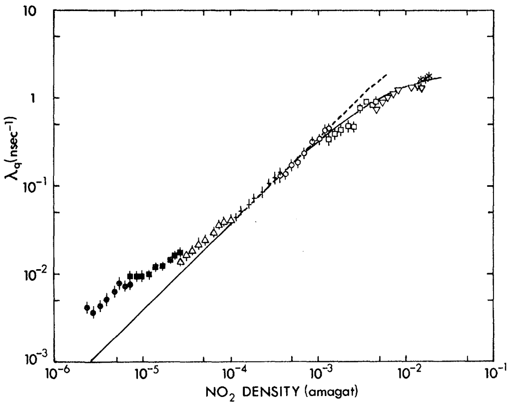

FIG. 3. Quenching rate of *o*-Ps for nitrogen dioxide in nitrogen dioxide and nitrogen mixtures plotted against the density of NO_2 only with N_2O_4 excluded; volume fraction of nitrogen dioxide in the mixtures: inclined crosses, 0.28%; inverted triangles, 0.074%; open squares, 0.020%; circles, 0.0055%; crosses, 0.0015%; triangles, 4.1 ppm; closed squares, 1.1 ppm; dots, 0.30 ppm; solid line, conversion quenching with rate $460D$ nsec^{-1} amagat^{-1}; dashed line, chemical quenching with same rate.

$$\lambda_q = 460D \ \mathrm{nsec}^{-1}\,\mathrm{amagat}^{-1}. \tag{9}$$

The errors involved are estimated to be less than 10%. If the Ps atoms are assumed to be thermalized during the conversion quenching, this rate is equivalent to a quenching cross section of 21×10^{-16} cm^2. This is the highest quenching rate or cross section ever determined for Ps atoms. The value of the cross section is about the same as the value calculated by assuming the NO_2 molecules to be hard balls. This implies that every collision is effective.

At lower densities of NO_2 ($< 10^{-4}$ amagats), the experimental data lie higher than the theoretical curve and the deviation is higher at lower densities. This can be explained as owing to the release of extra adsorbed NO_2 molecules on the walls of the vessel at lower densities. The total area of the vessel is calculated to be about 2000 cm^2 and the total volume 1200 cm^3. Assuming a NO_2 molecule to occupy an area of 10^{-15} cm^2 at the surface of the walls of the vessel, a monolayer adsorption of NO_2 molecules will account for 2×10^{18} NO_2 molecules. This is equivalent to 6×10^{-5} amagat, if all of them are desorbed. It seems that the desorption starts when the density of NO_2 in the gas phase is less than 10^{-4} amagat and NO_2 is preferably adsorbed on the walls (stainless steel) than N_2 molecules.

B. Nitric Oxide

Since both the free-positron-annihilation rate and Ps quenching rate for the nitrogen oxide gas is very high, a small amount of contamination of nitrogen dioxide in the nitric oxide gas will affect the outcome of the experimental result considerably, particularly when both these rates for nitric oxide are low. The contamination can also be caused by the inclusion of a small amount of O_2 in the gas owing to the reaction

$$2NO + O_2 \rightarrow 2NO_2 . \tag{10}$$

This has been found to be true. The values of the free-positron annihilation rate and Ps quenching rate for NO reported before[18] was for contaminated NO. A quite different result was obtained after the NO gas had been carefully purified.

The nitric oxide gas obtained may contain typically 0.2% of nitrogen dioxide and 0.45% of nitrous oxide and 0.2% nitrogen as major impurities.[17] Since nitrous oxide[8] and nitrogen[4] are not strong quenching agents for Ps and free positrons, no attempts were made to remove them. Only nitrogen dioxide was to be removed. The nitric oxide gas under pressure was passed slowly through a coil cooled at dry-ice temperature, −78 °C, before it was used to fill the evacuated gas system. Nitric oxide, nitrous oxide, and nitrogen would pass through as gases. However, nitrogen dioxide would be trapped as a solid (in the form of the dimer) in the coil because of its high melting point of −11 °C. This method has been found to be very effective, because much lower annihilation rates were measured after the purification.

In pure nitric oxide there appears only one very long-lifetime component in positron-annihilation-lifetime spectra. There is also a middle long-lifetime component with a mean life of about 2 nsec. However, the exact values of the meanlife of the middle (second) long-lifetime component are very hard to precisely determine. This second long-lifetime component appears more definitely when nitrogen is added. By the change of the annihilation rates at various gas densities, the first long-lifetime component is attributed to free-positron annihilation and the second to Ps annihilation.

1. Free-Positron Annihilation

The free-positron-annihilation rate for both pure NO and $NO + N_2$ mixtures is plotted against the total density of the gas in Fig. 4. The rate in pure NO is linearly dependent on the density of NO and is calculated to be

$$\lambda_{e+} = (6.9 \pm 0.2)D \ \mu\text{sec}^{-1}\,\text{amagat}^{-1} . \tag{11}$$

The equivalent value of Z_{eff} is only 34 which is only slightly higher than the number of electrons in a NO molecule, 30. This value $Z_{eff} = 34$ is much lower than the one determined by Abid and Paul.[7]

In nitric oxide and nitrogen mixtures with higher concentrations of nitric oxide, the free-positron-annihilation rate seems to be higher than in either pure nitric oxide or pure nitrogen, as shown in Fig. 4. However, for these data the statistical deviations are high. This is apparently due to the interference of the second long-lifetime component with an annihilation rate very close to the rate of the first long-lifetime component. As the concentration of NO decreases, the interference in turn decreases; then the free-annihilation rate approaches that of pure nitrogen and its statistical deviations also decrease.

2. Positronium Annihilation

The mean life of the second long-lifetime component in the positron-annihilation-lifetime spectra in purified NO gas is only about 2 nsec or less. However, a second long-lifetime component with a much longer mean life, about 10 nsec or more, appears when N_2 is added to the NO gas as a diluent. This phenomenon is very similar to the cases of positron annihilation in nitric acid,[19] and other nitro compounds.[13,14] The quenching of *o*-Ps is observed primarily in the reduction of the intensity, but is not observed much in the reduction of the mean life of the component attributed to *o*-Ps. The addition of an inert diluent increases both the intensity and mean life of this component.

Tao and Green have proposed a theory to explain this phenomenon.[19] They proposed that *o*-Ps atoms are formed, but only the *o*-Ps atoms with energies

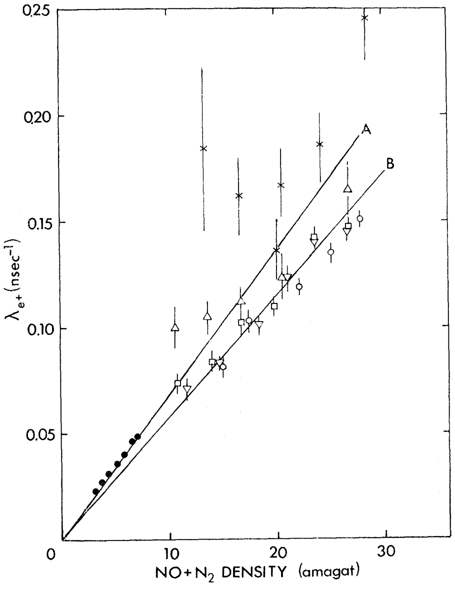

FIG. 4. Free-positron-annihilation rate in pure NO or NO and N_2 mixtures; dots, pure NO. Volume fraction of NO in the mixtures: inclined crosses, 11%; triangles, 5.0%; squares, 2.0%; inverted triangles, 0.80%; circles, 0.35%; line A, rate $6.9D$ μsec^{-1} amagat^{-1}; line B, rate $5.8D$ μsec^{-1} amagat^{-1}.

higher than a certain threshold energy are quenched by compound formation. It seems the quenching of o-Ps in NO follows a similar pattern. The reaction

$$\mathrm{Ps} + \mathrm{NO} \rightarrow \mathrm{PsNO}^* \rightarrow \gamma \text{ rays} \tag{12}$$

is suggested as an extra quenching process in addition to conversion. Here, we must propose that the compound PsNO formed is at an excited state and metastable. However, if the mean life of the excited state is greater than the annihilation mean life, about 0.5 nsec, this reaction will effectively quench the o-Ps. The threshold energy of reaction (12) is the energy required to form the PsNO* compound.

Certainly reaction (12) will compete with the slowing-down reaction

$$\mathrm{Ps} + \mathrm{NO} \rightarrow \mathrm{Ps}(\text{lower energy}) + \mathrm{NO}. \tag{13}$$

In pure NO reaction (12) dominates. Once N_2 is added to the NO gas, the slowing-down reaction will become dominant. Then a component in the positron-annihilation spectrum with a much longer mean life will appear. The quenching rate of o-Ps in NO or $NO + N_2$ mixtures is controlled by the reaction rates of reaction (12) and the slowing-down rate. Certainly, the inhibition of the formation of Ps atoms[20] is another plausible explanation. Since the extra slowing-down power contributed by the addition of nitrogen gas will compete with the inhibition reaction as well as the formation reaction of positronium, it requires a much more complicated process to describe this phenomenon. A more detailed investigation in NO with various diluents may clarify this point.

The quenching rate of o-Ps in the purified NO is estimated to be roughly

$$\lambda_q = 0.15D \text{ nsec}^{-1} \text{ amagat}^{-1}, \tag{14}$$

where D is the density in amagats. The quenching rate of o-Ps for NO in $NO + N_2$ mixtures is plotted against the density of NO in Fig. 5. The relationship is not linear. In view of the complicated nature of the formation and annihilation of Ps atoms in a mixture of NO and N_2, as mentioned above, this is not very surprising.

The quenching rate for the whole density range approximately fits the formula

$$\lambda_q = 0.038D^{0.6}, \tag{15}$$

with D being expressed in amagats and λ_q being expressed in nsec^{-1}. At a density range of 1–3 amagats, the rate is approximately

$$\lambda_q = 0.029D \text{ nsec}^{-1} \text{ amagat}^{-1}, \tag{16}$$

and at a very low density the rate is approximately

$$\lambda_q = 0.12D \text{ nsec}^{-1} \text{ amagat}^{-1}. \tag{17}$$

This rate is very close to the rate in purified NO. Both of these rates are of the same order of magnitude as the quenching rates of oxygen gas[21] and ions of the transition metals in solutions[22] but are far below the quenching rate of NO_2 ($460D$ nsec^{-1} amagat^{-1}).

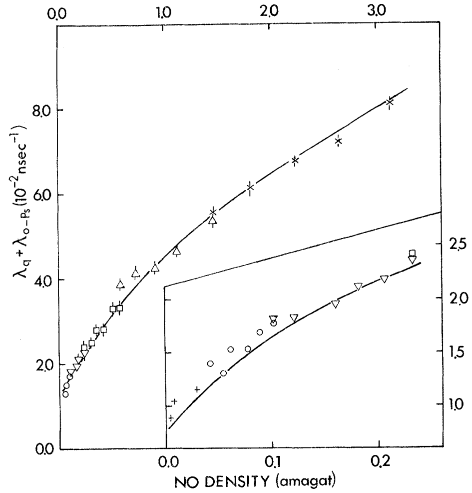

FIG. 5. Quenching rate of o-Ps for NO in NO and N_2 mixtures plotted with annihilation rate of o-Ps in free space included against density of NO (the insert is an enlarged portion at lower densities). See Fig. 4 for explanation of data points except those crosses which are for lower NO concentrations; solid line, quenching rate $0.038D^{0.6}$, with D in amagats and λ_q in nsec^{-1}.

IV. SUMMARY

The free-annihilation rate of positrons in nitrogen dioxide gas with both monomers and dimers in equilibrium is $(0.048 \pm 0.003)d$ nsec^{-1} g^{-1}. When nitrogen is added as a diluent the rate at higher gas densities (> 15 amagat of nitrogen) is higher, $(0.065 \pm 0.006)d$ nsec^{-1} g^{-1}. This can be explained as owing to the lower annihilation rate at energies higher than thermal. The equivalent values of Z_{eff}, 720 and 1090, respectively, are much higher than even the numbers of electrons in the dimers (46). The formation of a collision complex e^+NO_2 is suggested as an explanation of the high annihilation rate.

The monomer of the nitrogen dioxide (NO_2) has been found to be the strongest quenching agent for o-Ps ever observed. The quenching rate is found to be $460D$ nsec^{-1} amagat^{-1} which is equivalent to a cross section 21×10^{-16} cm^2, if the o-Ps atoms are assumed to be thermalized. The quenching mechanism is found to be caused by conversion.

For studying positron annihilation in nitric oxide, care must be taken to remove all the nitrogen dioxide in the gas. In the purified NO gas the free-annihilation rate is found to be $(6.9 \pm 0.2)D$ nsec^{-1}

amagat^{-1}, which is equivalent to $Z_{eff} = 34$.

The Ps quenching rate in pure NO gas is estimated to be roughly $0.15D$ nsec^{-1} amagat^{-1}. When N_2 is added to the NO gas, the quenching rate is changed to about $0.029D$ nsec^{-1} amagat^{-1}. This type of quenching is very similar to Ps in nitric acid. The formation of a metastable positronium compound PsNO* is proposed to account for the higher quenching rate for higher-energy *o*-Ps atoms The addition of N_2 increases the slowing-down rate. Therefore the quenching rate is reduced. However, the quenching rate for NO in $NO + N_2$ mixtures is not linearly dependent on the density of NO. At very low densities of NO the quenching rate increases to about $0.12D$ nsec^{-1} amagat^{-1}, which is the same as the rate in pure NO. It seems that in N_2 gas *o*-Ps atoms are not fully thermalized before their annihilation. A more detailed investigation in NO with various diluents may clarify this point. It is also surprising to find that the annihilation rates of both free positrons and Ps are much higher in nitrogen dioxide than those in NO.

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Evolution of Explosively Unstable Systems

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The present study concerns the evolution in time of explosively unstable systems under the influence of time-dependent dissipative and nonlinear coupling effects. Generalized criteria for instability as well as new mode solutions are constructed by introducing a suitable transformation in time.

I. INTRODUCTION

So far, the studies on explosive instability[1–12] have been devoted to systems where the parameters of the problem, i.e., the coefficients of nonlinear coupling and of dissipation, are considered as constants with respect to time. Accordingly the analysis of the associated problems needs further detailed consideration, taking time dependence of the above parameters into account. An extended theory is particularly motivated by the interesting possibilities of comparison offered by recent experi-