CONVERSION OF POSITRONIUM ATOMS ON O_2 *AND* NO *MOLECULES*

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Angular correlation curves for annihilation γ quanta are measured in a number of pure gases and in Ar + O_2 and Ar + NO mixtures at pressures on the order of 100 atm. A method is presented for separating the narrow component in the experimental curves for monatomic and polyatomic gases. The cross sections for the conversion of positronium atoms on O_2 and NO molecules are obtained from an analysis of the curves for the gas mixtures. The cross sections for the production of Ps atoms on the same molecules are also estimated.

IF the center of gravity of an annihilating electron-positron pair has a momentum p then in the case of two-photon annihilation, the difference between the γ-quantum scattering angle and π reaches a value $\theta = p/mc$, where m is the electron mass. In the case of positron annihilation in gases, the angular distribution of the γ quanta contains two components, a narrow component due to annihilation of singlet positronium, and a broad component corresponding to the annihilation of positrons with the orbital electrons of the atoms of the molecules (annihilation of free positrons, pick-off annihilation of Ps atoms, annihilation in the e^+m complex, etc.). The information that can be obtained from an analysis of the angular-correlation curves reduces primarily to qualitative conclusions concerning the character of the interaction of the positronium atoms. Thus, the conversion of the Ps atoms (i.e., the $^3S_1 \rightleftarrows {}^1S_0$ transition), on paramagnetic molecules (O_2, NO, NO_2), leads to an intensification of the narrow component; to the contrary, chemical interaction of the Ps atoms (for example, with Cl_2 molecules) causes an increase of the broader components at the expense of the narrower one. If the positronium is produced in the ortho and para states in the statistical ratio, that is, 3:1, then the intensity of the narrow component will be $I_N = f/4$ in the absence of conversion and chemical reactions (f is the fraction of the positrons making up the positronium). The presence of paramagnetic molecules in a gas that is inert with respect to Ps increases I_N compared with pure gas, and the solution of the kinetic equations yields the following relation for I_N[1]:

$$I_N = \frac{f}{4}\frac{\lambda_s(4\gamma+\lambda_p+\lambda_t)}{\gamma(3\lambda_t+4\lambda_p+\lambda_s)+(\lambda_t+\lambda_p)(\lambda_s+\lambda_p)}. \qquad (1)$$

Here $\lambda_s = 8 \times 10^9$ sec^{-1} and $\lambda_t = 7.2 \times 10^6$ sec^{-1} are respectively the rates of Ps decay in vacuum in the para and ortho states, $\gamma = n\sigma_c v$ is the conversion rate (v--velocity of Ps atom, n--number of converter molecules in 1 cm^3), and λ_p is the rate of pick-off annihilation of the positronium. By comparing the values of I_N obtained from the experimental angular-correlation curves with the theoretical relation (1) we can, knowing λ_p, determine the cross section for the conversion of the Ps atoms.

Measurements of the angular distribution of the annihilation quanta were made with the setup illustrated in Fig. 1. The angular resolution of the setup was about 1 mrad, the coincidence resolution time of the circuitry was 0.5 μsec. The positron source used in most measurements was the isotope Na^{22}, and in some measurements Cu^{64}. We determined the counting rate of the annihilation-quantum coincidences (energy 0.5 MeV) as a function of the angle θ between the detectors (NaI(Tl) crystals with FÉU-13 photomultipliers). The random-coincidence background was measured at θ = 20 mrad and subtracted from all the experimental curves. The working chamber was evacuated to a pressure 5×10^{-2} mm Hg before it was filled with the gas.

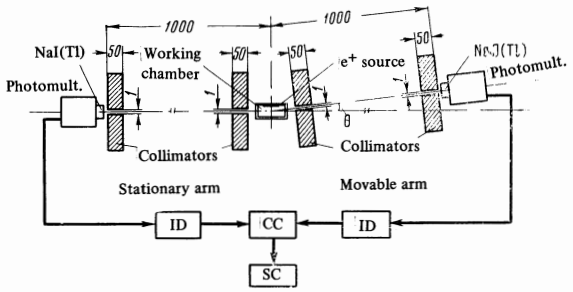

FIG. 1. Diagram of experimental setup. ID – integral discriminator, CC – coincidence circuit, SC – scaler.

The measurement objects were chosen primarily from among those gases for which the fractions of the positrons making up the positronium were known,

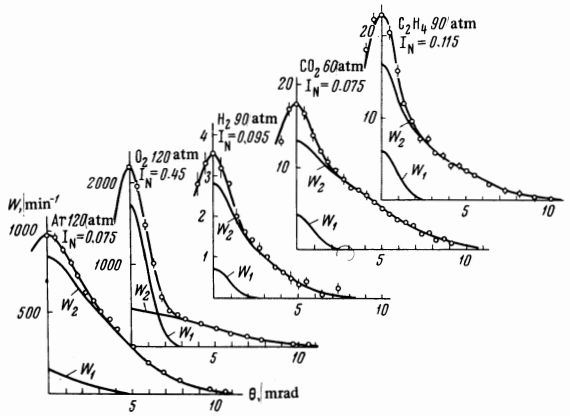

FIG. 2. Angular-correlation curves in pure gases.

namely inert (argon), diatomic (oxygen, which acts as a converter gas, and hydrogen), polyatomic (carbon dioxide, ethylene). Figure 2 shows the experimental plots of the angular correlation for the indicated gases at pressures in the 100 atm range. Each curve can be represented by the sum $W(\theta) = W_1(\theta) + W_2(\theta)$, where $W_1(\theta)$ corresponds to a distribution of quanta from the p-Ps annihilation (narrow component) and $W_2(\theta)$ is due to the annihilation of the free positrons and the o-Ps pick-off annihilation (broad component). The form of the function $W_1(\theta)$ can be obtained from the formula (see, for example, [2])

$$W_1(\theta) = C \int_{\theta = p/mc}^{\infty} \rho(p)\, p\, dp, \qquad (2)$$

since the criterion of the applicability of this formula[3] is satisfied for the narrow component under the conditions of the geometry of the employed setup.

To determine $W_1(\theta)$ it is necessary to know the density of the momentum distribution $\rho(p)$ of the p-Ps atoms at the instant of annihilation; this dependence differs greatly in inert and polyatomic gases. If we assume, as is customarily done, that initially the atoms of the positronium are uniformly distributed in energy from zero to E_0 (in accordance with the representations of Ore[4], E_0 is given by the relation $E_0/(V_i - 6.8 + E_0) = f$, where V_i is the ionization potential of the gas; all the quantities are in eV), then it is easy to show that in an inert gas, in which the positronium can lose energy only by elastic collisions, the functions $\rho(p)$ (at the instant of decay) and $W_1(\theta)$ take the form

$$\rho(p) = \frac{\text{const}}{p_0^2(1 - a/\lambda_s)}\left[1 - \left(\frac{p}{p_0}\right)^{2(\lambda_s/a - 1)}\right]\frac{1}{p} \qquad (3)$$

$$W_1(\theta) = \text{const}\left\{\theta_0\left(1 - \frac{1}{2\lambda_s/a - 1}\right) - \theta\left[1 - \frac{1}{2\lambda_s/a - 1}\left(\frac{\theta}{\theta_0}\right)^{2(\lambda_s/a - 1)}\right]\right\}.$$

Here $\theta_0 = p_0/mc = 2 \times 10^{-3}\sqrt{2E_0}$, $a = N\sigma_S v\xi$, N is the number of gas atoms in 1 cm^3, $\xi = 2m_{Ps}/M$ is the average logarithmic decrement of the energy per collision, and σ_S is the cross section for the elastic scattering of Ps atoms by the gas atoms, it being assumed that $\sigma_S \sim 1/v$, where v is the velocity of the positronium atom.

There are no published experimental data on the effective cross sections for the elastic scattering of Ps atoms by inert-gas atoms. Calculated values of the cross sections for the elastic scattering of Ps atoms by He atoms are given in[5], where it is shown that the energy changes from 4.5 to 10^{-16} cm^2 to 1.3×10^{-15} cm^2 when the energy of the Ps atom changes from 6.8 to thermal. Taking into account the ratio of the geometric dimensions of the He and Ar atoms, we have assumed that the cross section for the elastic scattering of thermal-energy Ps atoms by Ar atoms is 2×10^{-15} cm^2, which yields for the parameter λ_S/a a value ~2 at an argon pressure 120 atm. We note, incidentally, that the cross section for the elastic scattering of hydrogen atoms by Ar is also of the order of 10^{-15} cm^2 at a velocity equal to the thermal velocity of the Ps atom[6].

In polyatomic gases, the positronium loses effective energy as a result of excitation of vibrational and rotational energy levels of the molecules, so that it can be assumed that at pressures ~100 atm the para positronium has time during its lifetime to enter into thermal

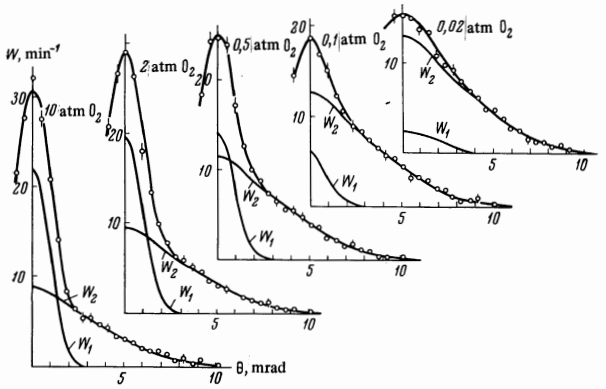

FIG. 3. Angular-correlation in Ar + O_2 gas mixture at total pressure 120 atm.

equilibrium with the gas. Indeed, the mean free path of the atom at such pressures is $\sim 10^{-7}$ cm and the para positronium experiences on the average 10^4 collisions during its lifetime ($\sim 10^{-10}$ sec). Even if the probability of excitation of the vibrational quantum of the molecule upon collision with the Ps atom were 10^{-2} (which corresponds to a cross section $\sim 10^{-18}$ cm^2), the positronium would have time to lose several dozens of eV (the energy of the vibrational quantum of the molecules lies in the region 0.15–0.3 eV), whereas the maximum energy of the Ps atom is only 6.8 eV. Consequently, the para positronium has a Maxwellian distribution at the instant of annihilation, that is,

$$\rho(p) = \text{const} \cdot \exp(-p^2/4mkT)$$

and the narrow component, assuming ideal geometric resolution, would have the form

$$\nu(\theta) = \text{const} \cdot \exp\left(-\frac{mc^2}{4kT}\theta^2\right).$$

If $R(\theta' - \theta)$ is the resolution function of the apparatus, then, the experimentally measured narrow component is given by the following formula:

$$W_1(\theta) = \int \nu(\theta') R(\theta' - \theta)\, d\theta'.$$

We chose for the resolution function the Gaussian curve

$$R(\theta' - \theta) = \text{const} \cdot \exp\{-(\theta' - \theta)^2/2\sigma^2\},$$

where $\sigma = 0.85$ mrad and is determined by the collimator slit width. Integrating, we get

$$W_1(\theta) = \text{const} \cdot \exp\left\{-\frac{\alpha\theta^2}{1 + 2\alpha\sigma^2}\right\}, \qquad (4)$$

where $\alpha = mc^2/4kT = 5 \times 10^6$.

The $W_1(\theta)$ curves of Fig. 2 were calculated from formulas (3) for Ar and (4) for the remaining gases, the constants being chosen such as to satisfy the condition:

$$I_N = \int_0^{\infty} W_1(\theta)\, d\theta \Big/ \int_0^{\infty} [W_1(\theta) + W_2(\theta)]\, d\theta$$

$$= \begin{cases} f - \text{ in the case of oxygen} \\ f/4 - \text{ in all other cases.} \end{cases}$$

The experimental values of f are given in[7]; most data were obtained at pressures not higher than 30 atm; however, according to Levin (private communication),

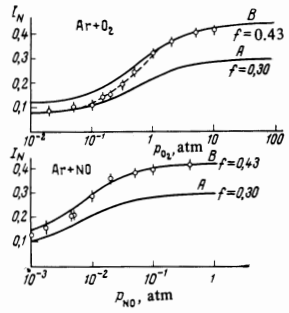

FIG. 4. Variation of intensity of narrow component as a function of the pressure of the additives in the gas mixtures Ar + O_2 and Ar + NO at a total pressure of 120 atm.

the intensity of the long-lived component of the temporal spectrum of positron annihilation in argon does not depend on the pressure, up to 150 atm, and f amounts to 30% in argon.

As already noted, an admixture of paramagnetic molecules in the inert gas causes an intensification of the narrow component. Figure 3 demonstrates this fact convincingly for the Ar + O_2 mixture at a total pressure of 120 atm. The relative content of O_2 in the Ar + O_2 mixture do not exceed 10%, so that it can be assumed that the form of the broad component for the mixture does not differ from the form for pure argon. Joining the curves of Fig. 3 with the $W_2(\theta)$ curve for Ar (Fig. 2), in the angle range in which there is no narrow component ($\theta > 3$ mrad) we obtain the broad component for the mixture, and then also the narrow component $W_1(\theta) = W(\theta) - W_2(\theta)$. The values of I_N calculated in this manner are plotted against the pressures of O_2 and NO in Fig. 4 together with the theoretical curves obtained from formula (1).

For the curves A, we have f = 30% (pure argon); for curves B, f corresponds to the fraction of positronium production in pure O_2 and NO. The value of λ_p was assumed to be 2.5×10^5 sec^{-1} atm^{-1} [8], and the values of the volume conversion probabilities $\gamma/n = \sigma_c v$ are listed in the table and are compared with the data obtained from an analysis of the annihilation time spectra [8,9].

It is seen from Fig. 4 that with increasing pressure of the O_2 in the Ar + O_2 mixture an increase takes place in the fraction of positronium formation[1)] and that at O_2 pressures higher than 1 atm f reaches the same value as in pure oxygen. This circumstance makes it possible to estimate the order of magnitude of the effective cross section for a Ps production on O_2 molecules. If V_i is the ionization potential of a gas, then it is obvious that the positronium can be produced only by those positrons whose energy is not lower than $V_i - 6.8$ eV. Then, in the Ar + O_2 mixture, the positrons having an energy lower than the Ore gap for Ar but higher than the energy threshold for reproduction of Ps on O_2, that is, in the interval from 5.4 to 9.0 eV, can produce Ps on O_2 molecules and this is apparently precisely the reason why

Values of $\sigma_c v$ in $cm^3 sec^{-1}$

Converter	Present work	Results of [8,9]
O_2	$(2,4 \pm 0,5) \cdot 10^{-12}$	$(0,8 \div 3,0) \cdot 10^{-12}$
NO	$(2,1 \pm 0,5) \cdot 10^{-10}$	$(1 \div 4) \cdot 10^{-10}$

a transition from curve A to curve B is observed in Fig. 4. With this, low oxygen concentrations inhibit the formation of Ps on argon weakly, but at concentrations higher than 1%, the Ore gap for Ar is effectively blocked by inelastic energy losses of the positrons on the O_2 molecules. Therefore the limiting value of the fraction of Ps production in the Ar + O_2 mixture is f_{O_2}, and we shall have for the intensity of the narrow component, in lieu of formula (1),

$$I_N = \frac{1}{4}[f_{Ar} + (f_{O_2} - f_{Ar})w] \frac{\lambda_s(4\gamma + \lambda_p + \lambda_t)}{\gamma(3\lambda_t + 4\lambda_p + \lambda_s) + (\lambda_p + \lambda_t)(\lambda_p + \lambda_s)}. \quad (1')$$

We assume here that the probability w of Ps production on O_2 by positrons with energy from 5.4 to 9 eV is equal to unity at O_2 concentrations exceeding 1%, and that at lower concentrations it is possible to neglect the inelastic energy loss of positronium, after which we readily obtain for w the expression

$$w = 1 - \frac{n_1\sigma_s}{n_2\sigma_f N}\left[1 - \exp\left\{-\frac{n_2\sigma_f N}{n_1\sigma_s}\right\}\right],$$

where σ_s and σ_f are the cross sections for the elastic scattering of positrons by Ar and the formation of Ps by O_2, averaged over the interval 5.4–9.0 eV; n_1 and n_2 are the numbers of Ar atoms and O_2 molecules per cm^3, and $N = \xi^{-1}\ln(E_{max}/E_{min})$ is the number of elastic collisions experienced by the positron on losing energy from E_{max} to E_{min} (for the Ar + O_2 mixture we have E_{max} = 9 eV, E_{min} = 5.4 eV).

The dashed curve of Fig. 4 was calculated from formula (1') with $\sigma_f/\sigma_s = 4 \times 10^{-2}$. If we use for σ_s the value $1.5\ \pi a_0^2$ obtained experimentally by Marder [11], then the average cross section for the production of Ps on O_2 is $\sim 5 \times 10^{-18}$ cm^2, which should be regarded as a lower limit of the cross section, since we have not taken into account the inelastic losses of positron energy on the O_2 molecules. A similar estimate shows that the cross section for the production of positronium on NO is approximately two orders of magnitude higher than on O_2. The same pertains also to the conversion cross section, as can be seen from the table.

In conclusion we note that we have also investigated the plots of the angular correlations in Ar + O_2 and Ar + NO mixtures at different argon pressures. Within the limits of experimental errors, no dependence of the cross sections for the conversion of the Ps atoms on O_2 or NO on the argon pressure was observed in the interval 30–120 atm.

1)The counting rate of 3γ coincidences in the annihilation of positrons in gases and gas mixtures was investigated in [10] and it was shown that when oxygen or nitrogen is added to argon, the fraction of positrons forming the positronium is appreciably increased compared with pure argon.

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