

## OBSERVATION OF POSITRONIUM REACTIONS IN AQUEOUS SOLUTIONS

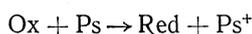
V. I. GOL'DANSKIĬ, O. A. KARPUKHIN, and G. G. PETROV

P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R. and Institute of Chemical Physics, Academy of Sciences, U.S.S.R.

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SEVERAL reports have recently been published on the annihilation of positrons in aqueous solutions, or on the effect of different additives on this annihilation,<sup>1-4</sup> and the proposed treatment of the observed results is based on attempts to introduce the thermodynamic oxidation-reduction potential of positronium.<sup>3,4</sup> Such a treatment, however, is not completely correct for the following reasons: a) in connection with the fast annihilation of the positrons, chemical processes of the type



(Ox — oxidizer, Red — reducer,  $\text{Ps} = e^+e^-$ ,  $\text{Ps}^+ = e^+$ ) are essentially irreversible; b) for the same reason and because of the exceedingly small stationary concentrations of Ps and  $\text{Ps}^+$ , one cannot use for the analysis of the positronium reactions the potentials of multi-electron transitions of the type  $\text{Me}^{++} + 2\text{Ps} \rightarrow \text{Me} + 2\text{Ps}^+$  (Me — metal); c) no macroscopic metallic phase is produced in reactions in which positronium participates, so that there is no justification for operating with standard metal-ion potentials. We note furthermore that the experiments described in references 1-4 were carried out essentially in highly concentrated (2M) solutions of multi-valent ions, in which the activity of the ions is, as is well known, a highly indeterminate quantity. In order to demonstrate the essentially kinetic character of the action of different substances on the annihilation of positronium in aqueous solutions, and in order to arrive at a systematic comparison of such an action with oxidation-reduction and magnetic characteristics of different ions, we have carried out a series of experiments to investigate the rate of  $3\gamma$  annihilation of positrons from a  $\text{Na}^{22}$  source (0.1 mC) in aqueous solutions. Without going into details of the apparatus, which is similar to that described by McGervey and de Benedetti,<sup>3</sup> we shall indicate only the basic results.

1) The table indicates the decrease in the rate of  $3\gamma$  annihilation compared with pure water under

the influence of various additives (essentially, different cations with a  $\text{Cl}^-$  anion inert to positronium). In the presence of a general tendency towards reduction of the counting rate  $C_{3\gamma}$  on going over to stronger oxidizers, there are, however, also serious deviations from such a sequence.

Substance	Concentration, mole/liter	Standard oxidation-reduction potential for the oxidizer-reducer pair indicated in parentheses	No. of unpaired electrons	Difference in $C_{3\gamma}$ ( $\text{min}^{-1}$ ) relative to water
KOH	1	+2.92 (K <sup>+</sup> /K)		-0.02±0.31
BaCl <sub>2</sub>	2	+2.92 (Ba <sup>++</sup> /Ba)		-0.55±0.39
NaCl	2	+2.71 (Na <sup>+</sup> /Na)		+0.07±0.25
MnCl <sub>2</sub>	2	+1.10 (Mn <sup>++</sup> /Mn)	5	-0.57±0.26
ZnCl <sub>2</sub>	2	+0.76 (Zn <sup>++</sup> /Zn)		+0.24±0.34
FeCl <sub>2</sub>	0.1	+0.44 (Fe <sup>++</sup> /Fe)	4	-0.99±0.27
CrCl <sub>3</sub>	2	+0.41 (Cr <sup>+++</sup> /Cr <sup>++</sup> )	3	-1.70±0.28
TlNO <sub>3</sub>	saturation	+0.34 (Tl <sup>+</sup> /Tl)		-1.24±0.29
CoCl <sub>2</sub>	2	+0.27 (Co <sup>++</sup> /Co)	3	-1.17±0.40
NiSO <sub>4</sub>	2	+0.23 (Ni <sup>++</sup> /Ni)	2	-1.03±0.36
CuCl <sub>2</sub>	2	-0.34 (Cu <sup>++</sup> /Cu)	1	-1.85±0.31
FeCl <sub>3</sub>	2	-0.77 (Fe <sup>+++</sup> /Fe <sup>++</sup> )	5	-2.62±0.33
FeCl <sub>3</sub>	0.1	-0.77 (Fe <sup>+++</sup> /Fe <sup>++</sup> )	5	-1.41±0.28
KMnO <sub>4</sub>	saturation	-1.63 (MnO <sub>4</sub> <sup>-</sup> + 3e/MnO <sub>2</sub> )		-2.44±0.42
H <sub>2</sub> O <sub>2</sub>	30% by weight	-1.78 (H <sub>2</sub> O <sub>2</sub> + 2e/H <sub>2</sub> O)		-1.55±0.28

These deviations can be partially due to the  $^3\text{S}_1 \rightarrow ^1\text{S}_0$  conversion on unpaired electrons of paramagnetic ions, but there is likewise no unambiguous connection between the magnetic properties of the ions and the value of  $C_{3\gamma}$ . Thus, in spite of the similarity of the data in the Table with the results of McGervey and de Benedetti,<sup>3</sup> and also with the results of Trumpy<sup>4</sup> (in those cases where we investigated the same additives), we do not consider it established that a systematic variation of  $C_{3\gamma}$  takes place with standard oxidation-reduction potentials of the additives.

2) In accordance with the results of Green and Bell<sup>1</sup> and of Trumpy,<sup>4</sup> we have also observed a strong quenching of the counting rate  $C_{3\gamma}$  by additions of the oxidizer anion  $\text{NO}_3^-$ , and found in addition that  $\text{MnO}_4^-$  has an even stronger action. But the action of  $\text{MnO}_4^-$  (0.01 M) was found to be perfectly identical in strongly acid (pH = 2), neutral (pH = 7) and alkali (pH = 12) solutions: the respective values of  $C_{3\gamma}$  were  $5.13 \pm 0.16$ ,  $5.01 \pm 0.16$ , and  $5.15 \pm 0.27 \text{ min}^{-1}$  (the value for water being  $6.04 \pm 0.09$ ). This demonstrates convincingly the decisive role of the concentration of  $\text{MnO}_4^-$ , and not of the oxidation-reduction potential of the system. We can arrive at an analogous conclusion from data on the quenching of  $C_{3\gamma}$  by means of additives of  $\text{Fe}^{+++}$  and  $\text{Fe}^{++}$ , taken at different concentrations.

3) The counting rate  $C_{3\gamma}$  as a function of the concentration of the  $\text{MnO}_4^-$  additive (in neutral solution) is given by the following figures:

Concentration of $MnO_4$ , mole/liter $C_{3\gamma}$ , $min^{-1}$	Saturated solution	0.1	0.01	0.001	0 (water)
	$3.6 \pm 0.42$	$5.08 \pm 0.45$	$5.08 \pm 0.12$	$5.50 \pm 0.30$	$6.04 \pm 0.09$

It is obvious that the quenching of  $C_{3\gamma}$  appears quite strongly even at low concentrations. In experiments with high concentrations of highly active additives, the value of  $C_{3\gamma}$  tends to a limit corresponding to the contribution of free  $3\gamma$  annihilation. Therefore a quantitative comparison of the effects of different additives on the formation and conversion of positronium makes it obligatory to perform the experiments at small concentrations. At the present time we continue systematic investigations of the kinetic characteristics of different reactions of positronium in aqueous solutions.

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<sup>1</sup>R. Green and R. Bell, *Can. J. Phys.* **35**, 398 (1957); **36**, 1684 (1958).

<sup>2</sup>R. de Zafra, *Phys. Rev.* **113**, 1457 (1959).

<sup>3</sup>J. McGervey and S. de Benedetti, *Phys. Rev.* **114**, 495 (1959).

<sup>4</sup>G. Trumpy, *Phys. Rev.* **118**, 668 (1960).

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### ASYMMETRY OF MOTT DOUBLE SCATTERING AND ABSOLUTE VALUES OF LONGITUDINAL POLARIZATION OF BETA PARTICLES

P. E. SPIVAK, L. A. MIKAÉLYAN, I. E. KUTIKOV,  
and V. F. APALIN

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AS a result of relative measurements of longitudinal polarization of the  $\beta$  particles emitted in the decay of  $P^{32}$ ,  $Sm^{153}$ ,  $Lu^{177}$ ,  $Ho^{166}$ ,  $In^{114}$ , and  $Au^{198}$ , differences up to 12% have been observed in the magnitude of the longitudinal polarization. This result showed the presence of deviations of the magnitude of the polarization from the predicted value, which for electrons is equal to  $v/c$ . The size of these deviations was obtained from results of absolute measurements of the polariza-

tion of the electrons from  $Sm^{153}$ .<sup>2</sup> The longitudinal polarization of the electrons was converted to a transverse polarization in crossed magnetic and electric fields and was found from the asymmetry of the scattering by gold from the relation

$$\langle \sigma \rangle = (1 + J_l / J_r) / (1 - J_l / J_r) S,$$

where  $\langle \sigma \rangle$  is the degree of polarization,  $J_l$  and  $J_r$  are the scattered intensities to left and right, and  $S$  is a function of angle, energy and charge. Using the values of  $S$  computed by Sherman,<sup>3</sup> we found that in the case of  $Ho^{166}$  and  $In^{114}$  the differences of the values of the polarization from the value of  $v/c$  reached 12–15%.

As we have noted,<sup>1,2</sup> for a final judgment concerning the magnitude of the deviation of the polarization from the value  $v/c$  we must have trustworthy values of the function  $S$ . The existing experimental data concerning this question are in most cases not sufficiently exact, while for the angles and energies which are of interest to us there are no data at all.

In this connection, we undertook a measurement of the quantity  $S$  in an experiment on double scattering of unpolarized electrons by gold. In the following we give a brief description of the apparatus and the results of measurements for energies of 245 and 290 keV and scattering angles  $\theta_1 = \theta_2 = 120^\circ$ . At present, we are carrying out measurements over an energy range 50–250 keV and scattering angles 90–150°. The detailed description of the work will be published soon.

The geometry of the experiment is shown in the figure. A beam of accelerated electrons 2, after passing through a system of diaphragms, entered a chamber in which there was placed the first scatterer 1. The second scatterer 4 was in a separate chamber at a distance of  $\sim 1$  meter from the first. The twice-scattered electrons were recorded by ring Geiger counters 3 placed at a distance of about 20 cm from the second scatterer. To reduce scattering from the walls of the appa-

