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Positron Annihilation in Aqueous Solutions

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The angular correlation of 2-quantum emission from the annihilation of positrons in different materials has been measured in an apparatus with 8 photon counters providing coincidences for 16 output channels. As positron targets were chosen indium, water and aqueous solutions of 5 paramagnetic salts and 10 other substances. It was confirmed that the amount of singlet positronium formed is influenced by two processes: a reduction of positronium due to electron capture by oxidizing substances and an increase of the triplet → singlet conversion due to the electron exchange with paramagnetic ions.

The oxidation potential of positronium is found to be very nearly zero. The conversion rate seems to be proportional to the number of unpaired electrons on the dissolved ions. A discrepancy with the interpretation of Green and Bell for their lifetime experiments is discussed.

I. INTRODUCTION

IN the study of the angular correlation of two-quantum annihilation of positrons it has been observed by several authors[1–5] that part of the annihilation events give rise to a "narrow component" in the angular correlation curve. This component is due to quanta emitted with a relative angle differing very little from 180°, meaning that the center of mass of the annihilating particles has a small momentum. Since it is reasonable to assume that the positronium atom has a small linear momentum, it is now generally agreed that the narrow component is due to self-annihilation of singlet positronium.

Also in positron lifetime experiments evidence for two-quantum annihilation of positronium is found in the form of a long component τ_2 in the lifetime. Experimental evidence for a relationship between the τ_2 and the narrow components is provided by a comparison of the lifetime measurements by Bell and Graham[6] and the angular correlation studies of Page et al.[1,2] both using crystalline and fused quartz as targets. Similar evidence is given by the experiments of Green and Bell[7] and of de Zafra[5] on paramagnetic ions in water.

The present investigation was started with the purpose of establishing this relationship between the two methods in more detail. It was considered that more insight into the mechanism of annihilation might be gained by such a procedure. As the most thorough studies on the τ_2 component have been performed by Green and Bell[8] on ions in aqueous solutions, the present work was devoted to angular correlation studies on similar targets. Besides, aqueous solutions have the advantage that a great number of substances can be dissolved within a wide range of concentrations, forming convenient targets for positron annihilation. Solids may be equally convenient, but the formation of positronium is known to be dependent upon the crystalline state of the target.

This works shows the expected result, that the presence of paramagnetic ions tend to increase the narrow component due to the conversion triplet → singlet positronium. In addition was also discovered the surprising effect of narrow component reduction by a number of substances. However, this can be explained in a general way by the recent suggestion of McGervey and DeBenedetti[9] that oxidizing substances can break up the positronium atom.

II. EXPERIMENTAL METHOD

A. Principle

When an electron-positron pair at rest annihilates with the formation of two photons the conservation of momentum requires that these photons are emitted in exactly opposite directions. An initial center of mass momentum might therefore be detected as a deviation from the angle π between the emission directions.

The experiment indicated in Fig. 1 includes two photon counters which are essentially line detectors due to the very low and very broad horizontal slits that are placed in front of them. An annihilation source is placed between the counters, so that photons are detected in two planes, each plane through the source and through one counter slit. When photon coincidences are observed as a function of the angle θ between the two planes this experiment measures the distribution of the vertical momentum component k_z as shown in the figure. This component is given by

$$k_z = \theta mc, \tag{1}$$

provided $k \ll mc$ where k is the total center-of mass momentum of the annihilating system.

Stewart[10] has shown that in this experiment the

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[1] L. A. Page, M. Heinberg, J. Wallace, and T. Trout, Phys. Rev. 98, 206 (1955).

[2] L. A. Page and M. Heinberg, Phys. Rev. 102, 1545 (1956).

[3] A. T. Stewart, Phys. Rev. 99, 594 (1955).

[4] R. L. de Zafra and W. T. Joyner, Phys. Rev. 112, 19 (1958).

[5] R. L. de Zafra, Phys. Rev. 113, 1547 (1959).

[6] R. E. Bell and R. L. Graham, Phys. Rev. 90, 644 (1953).

[7] R. E. Green and R. E. Bell, Can. J. Phys. 36, 1684 (1958).

[8] R. E. Green and R. E. Bell, Can. J. Phys. 35, 398 (1957).

[9] J. McGervey and S. DeBenedetti, Phys. Rev. 114, 495 (1959).

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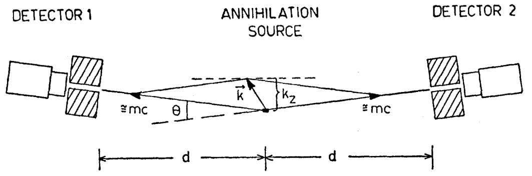

FIG. 1. Principle of the angular correlation experiment. The center-of-mass momentum of the annihilating system is indicated by the vector **k**.

density in momentum space is

$$\rho(k)=(a_1/\theta)\,dC(\theta)/d\theta, \tag{2}$$

and the momentum distribution is given by

$$N(k)=a_2\theta\,dC(\theta)/d\theta. \tag{3}$$

In these relations a_1 and a_2 are constant factors and $C(\theta)$ is the measured photon coincidence rate. Conditions for their validity are that $\rho(k)$ is varying slowly within the region Δk_z corresponding to the counter slit height, and secondly that the maximum momentum contributing to $\rho(k)$ corresponds to less than the width of the slits, i.e.,

$$k_{\max}/mc<b/d \tag{4}$$

where b is the effective slit width and d is the distance between the source and each counter.

B. Apparatus

The study of the angular correlation of annihilation quanta is always a rather time-consuming experiment since the solid angles utilized by the photon counters have to be very small, of the order of 10^{-5} steradian. Therefore the apparatus used in this work was constructed with the aim of obtaining data relatively fast. This setup is sketched in Fig. 2, which for the sake of clearness, is not drawn to scale.

A 5-cm diameter reactor irradiated copper disk provides the Cu^{64} positron source, having a half-life of 12.8 hours and an intensity of roughly 300 millicuries when extracted from the reactor. This source is mounted horizontally in a lead cassette which is open downward. The liquid target is 0.7 mm high, 6 cm wide and 4.5 cm long, resting on a 0.1-mm mica sheet which is supported 2 cm below the source. In this way a distant counter at approximately the same level can see the target and not the source. The lower part of the source-and-target unit is surrounded by a thin plastic bag keeping a moist atmosphere to prevent evaporation of the liquid. The photon counters are $1\frac{1}{2}$-in. diameter by $\frac{1}{4}$-in. high NaI(Tl) crystals mounted directly on photomultiplier cathodes. Instead of the customary slits a rectangular piece of lead is placed in front of each counter in such a way that only a lower crystal slice, the thickness of which represents 0.4 milliradian of θ, can see the target. Thus it is possible to mount the counters behind each other in approximately the same direction with respect to the target. Their relative angle is 0.6 milliradian for the counters $A\ B\ C\ D$ and 2.4 milliradians for $E\ F\ G\ H$. The series of counter pairs $A\ E, B\ E, C\ E, D\ E, A\ F, \cdots, D\ H$ therefore represents 16 values of θ covering 9.6 milliradians in steps of 0.6 milliradian. The steel beam holding counters $E\ F\ G\ H$ also can be turned about a horizontal axis through the target, making it possible to observe a complete angular correlation curve for each of the 16 counter pairs.

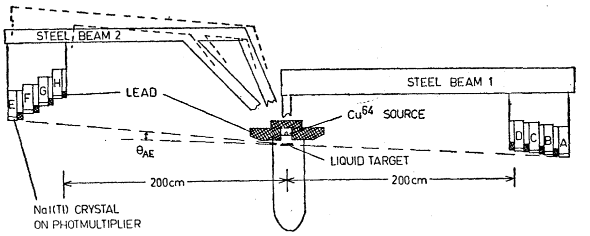

FIG. 2. Sketch of the apparatus made for the measurement of angular correlation of 2-quantum annihilation in liquids. Double coincidence rates are observed for pairs consisting of one counter from the set $A\ B\ C\ D$ and one from the set $E\ F\ G\ H$. The drawing is not to scale.

The sensitive slice of D which is the highest one of the stationary counters is located 4 cm below the plane of the liquid target surface. Measurements are predominantly taken with the counter set $E\ F\ G\ H$ above the straight line through counter D and the target. In this way the absorption of annihilation quanta in the liquid target is not varying appreciably with the angle θ. The electronic registering system is rather simple, consisting of 16 conventional germanium diode coincidence circuits of resolving time 2×10^{-7} sec, each connecting one pair of counters. Coincidences are fed to 16 output stages, each with a mechanical register. The circuits are set to accept pulses corresponding to energies above approximately 250 kev.

C. Measurements

In spite of the advantage that coincidences are observed in 16 channels simultaneously, corresponding to 16 values of the angle θ, data must be taken with frequent change of the angle between the two counter sets. The reason for this is that the response of a single channel can vary slightly due to electronic drift and temperature changes, so that the sensitivity factors cannot safely be considered as constant from one day to another. By changing the position of the movable counters $E\ F\ G\ H$ stepwise up and down again, part of a coincidence curve is obtained for each counter pair. Independently of single channel sensitivities these 16 curves can then be joined together to make one curve.

The background, which is mainly due to random coincidences can be measured by setting the smallest angle θ observed by any counter pair equal to 14 milliradians. In this position practically no 2-quantum annihilation coincidences are observed, while the single counting rates are unchanged. Maximum total single channel counting rates were about 25 coincidences per minute and about 1 coincidence per minute as background. The counting periods were from ½ to 2 hours, compensated for the decay of the 12.8-hour half-life source.

The results given below were obtained by about 3 days of effective counting time on each target.

III. RESULTS

A. Treatment of Data

Two-photon angular correlation measurements were performed for metallic indium, for distilled water and for solutions in water of the following chemicals: $MnCl_2$, $CoCl_2$, $NiCl_2$, $CuCl_2$, $FeCl_3$, NaCl, $ZnCl_2$, $CdCl_2$, $SnCl_4$, $NaSO_4$, $SbCl_3$, $NaNO_3$, $NaClO_3$, $KMnO_4$, and H_2O_2. All these substances were dissolved as 2 moles per liter of solution, except $KMnO_4$ which was dissolved to saturation. When added to water $SbCl_3$ forms SbOCl which was dissolved with dilute hydrochloric acid.

Green and Bell[7,8] have studied the lifetime of positrons as a function of concentration in a number of these solutions. The quenching effect that they observe is generally very near saturation at a concentration of 2 mole/liter, i.e., no greater change in the positron annihilation mechanism would be expected by increasing the amount of solute above this value. Similar saturation effects have been observed in angular correlation measurements by De Zafra.[5]

The results of the present experiment are given in Figs. 3, 4 and 5, where the curves on the left side show the measured coincidence intensities C as a function of the angle θ between the counter pairs. Background due to random coincidences has been subtracted and the curves have been normalized to the same area.

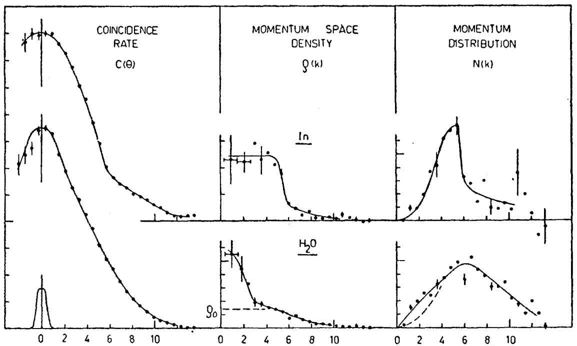

FIG. 3. Experimental results for indium metal and water. Relations (5) and (6) were used to obtain $\rho(k)$ and $N(k)$ from $C(\theta)$. The solid curves are just drawn to illustrate the general trend of the data, and are not based on any theory. The broken lines indicate the separation between the "broad" and "narrow" components, as explained in the text.

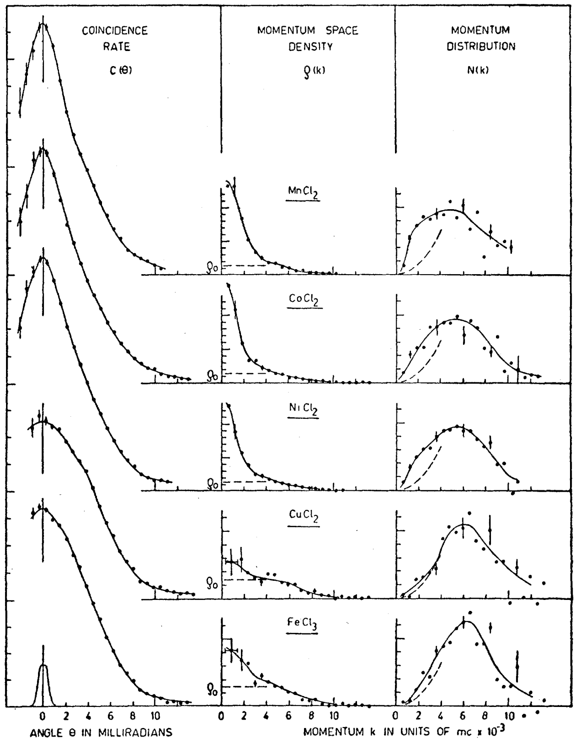

FIG. 4. Experimental results for paramagnetic ions, all dissolved in water as 2 moles per liter. See also text to Fig. 3.

The density in momentum space and the momentum distribution were calculated from the relations:

$$\rho(k)=a_1[2/(\theta_1+\theta_2)][C(\theta_2)-C(\theta_1)]/(\theta_2-\theta_1), \quad (5)$$

$$N(k)=a_2[(\theta_1+\theta_2)/2][C(\theta_2)-C(\theta_1)]/(\theta_2-\theta_1), \quad (6)$$

which is a reasonably good approximation to (2) and (3) since the changes in $C(\theta)$ are small compared to the difference $\theta_2-\theta_1$ between neighboring points.

Solid curves have been drawn as the seemingly best fit to the experimental points. These curves were not used for any of the calculations.

As the target and counters are 60 and 38 mm wide, respectively, the "effective slit width" b is about 100 mm. The maximum momenta contributing to $\rho(k)$ are about 0.015 mc so that the condition (4) for using the formulas (2) and (3) is clearly fulfilled. The experimental resolving power due to target and counter heights is something between a gaussian and a rectangular distribution. This curve is shown below the peaks of the coincidence curves. Its half-width is 1.1-milliradian corresponding to 0.0011 mc in momentum units. This "effective slit height" is a fairly large quantity compared to the variations in some of the curves for $\rho(k)$. A correction for this resolving power would move all the curves slightly against the origin, and particularly would the narrow peaks of $C(\theta)$ and $\rho(k)$ become narrower and higher. Such a correction was not applied, however, since it cannot be done quite unambiguously when the statistical errors are as large as here. Also, in this work we are particularly interested in studying the relative difference between the curves. The quantitative results on the narrow component which are given below are integrated magnitudes which are not influenced by geometric corrections applied to the curves.

An 0.7-mm thick plate of indium metal was used as a target in order to compare the obtained result with previous data. Stewart[10] has studied a number of metals with the angular correlation method. Our curves for indium are essentially the same as he obtained. As expected for a metal, one finds that the density in momentum space is uniform up to an abrupt cutoff, roughly corresponding to thermalized positrons annihilating in a free electron gas.

B. The Narrow Component

The sharp peak found in some of the curves $C(\theta)$ is called the "narrow component" which is generally interpreted as resulting from annihilation of singlet positronium. Only one clear-cut case of a narrow component superimposed upon a broader distribution is known from experiment. This is the case of ice as studied by De Zafra and Joyner.[4] For other targets one merely finds that the sharpness of the peak varies from one substance to another. Thus, from the curve $C(\theta)$ it is difficult to see clearly how the narrow component should be separated off.

A glance at Figs. 3, 4, and 5 shows that the narrow component is represented as a characteristic high density at low momenta in momentum space. The difference between various targets comes out more clearly in this representation, but the statistical uncertainties are large for the single points on the peaks since ρ is proportional to $1/k$.

Those curves for $\rho(k)$ which have the least narrow component can, within the statistical uncertainties, be represented by a momentum space density which has uniform height ρ_0 between 0 and about 0.005 mc. Also, several of the other curves show an indication of a step about the height of ρ_0. We shall take this momentum space density to represent the "broad component." This is indicated as a broken line in Figs. 3, 4, and 5. We shall now assume that the narrow component is given by the experimental points above this line in the region between 0 and 0.004 mc. In the momentum distribution representation the straight line $\rho=\rho_0$ is transformed into a parabola since $N(k)\propto\rho(k)k^2$. The area above this parabola, as given by the experimental points, is now defined as the narrow component intensity I_N, provided that the curve $N(k)$ is normalized to unit area. It is seen that I_N is not very sensitive to what form is chosen for the broad component at very low momenta.

In Table I the second column contains the experimental values of I_N. Also the value of I_N for the solution relative to the value for the solvent (water) is given. Statistical uncertainties for I_N are listed in the fourth column.

The somewhat arbitrary way of calculating I_N may be justified since the main point of the present investigation is to compare the effects of various targets by some standard procedure. A justification is also provided by a comparison with some of the results of Green and Bell.[8] They have measured the amount I_2 of triplet positronium decaying by two-photon annihilation in water and in 2 mole/liter $NaNO_3$ in water. For neither of these targets is there any reason to assume a conversion triplet → singlet positronium. As the ratio of

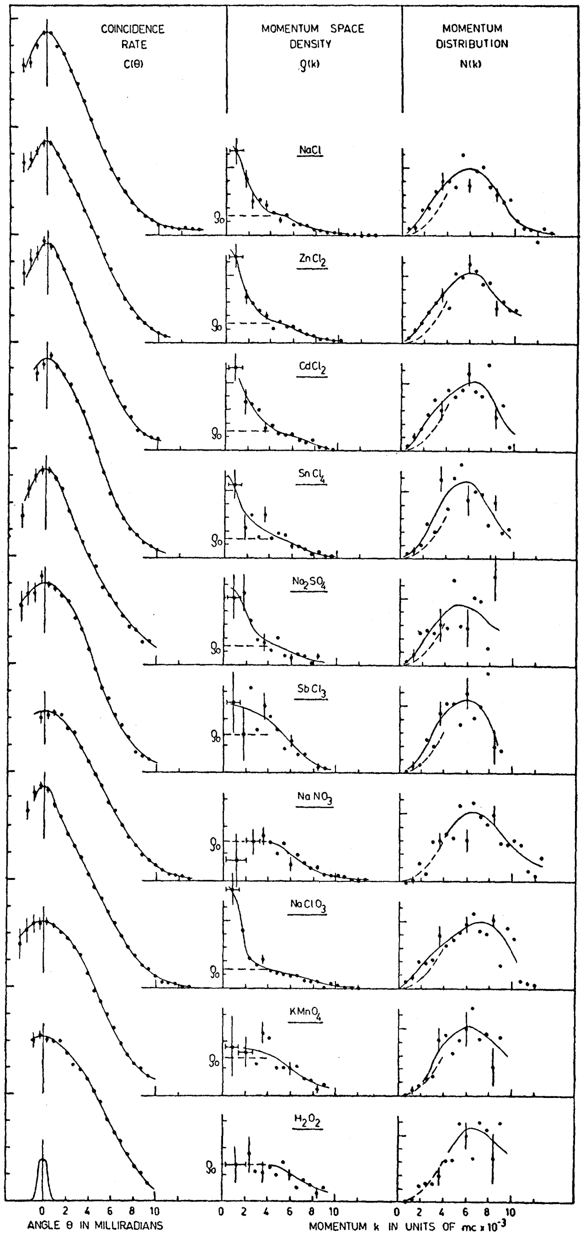

FIG. 5. Experimental results for nonmagnetic substances dissolved in water. $KMnO_4$ was dissolved to saturation, the other chemicals as 2 moles per liter. See also text to Fig. 3.

TABLE I. Experimental results together with other properties of the targets.

Target	Narrow component I_N (%)	I_N(target) $-I_N(H_2O)$ (%)	± (%)	No. of unpaired electrons	Oxidizing potential (V)	σ_{pc} from Green and Bell[a] (10^{-19} cm^2)
H_2O	8.7	0	1.0		...	
As solutes:						
$MnCl_2$	22.0	13.3	1.0	5	−1.1	2.5
$CoCl_2$	16.1	7.4	1.2	3	−0.29	5.3
$NiCl_2$	14.5	5.8	1.0	2	−0.22	5.6
$CuCl_2$	2.1	−6.6	1.0	1	0.34	11
$FeCl_3$	7.3	−1.4	1.0	5	0.74	38
NaCl	9.5	0.8	1.2		−2.71	
$ZnCl_2$	8.9	0.2	1.2		−0.76	
$CdCl_2$	10.1	1.4	1.2		−0.40	
$SnCl_4$	9.7	1.0	2.2		0.13	
Na_2SO_4	8.8	0.1	2.5		0.20	
$SbCl_3$	6.0	−2.2	2.0		0.21	3.7
$NaNO_3$	2.1	−6.6	1.2		0.94	
$NaClO_3$	7.6	−1.1	1.5		1.45	
$KMnO_4$	2.8	−5.9	2.0		1.63	
H_2O_2	−0.3	−9.0	1.5		1.78	

[a] See reference 7.

triplet to singlet positronium formed is as 3 to 1, we have $I_N = I_2/3$.

Green and Bell obtained the results

$$I_2(H_2O) = (21 \pm 5)\%,$$

$$I_2(2 \text{ mole/liter } NaNO_3) = (3.5 \pm 1.5)\%,$$

which gives for the narrow component

$$I_N(H_2O) = (7 \pm 1.7)\%,$$

$$I_N(2 \text{ mole/liter } NaNO_3) = (1.2 \pm 0.5)\%.$$

Both these results agree within experimental uncertainties with the data given in Table I.

De Zafra and Joyner[4] found the value $I_N = (8.8 \pm 0.9)\%$ for ice where the narrow and broad components are easily separated. This also fits well with our value, but it must not definitely be taken as the value for water since it is not known definitely whether I_N is changing or not when the phase changes.

The target NaCl dissolved in water shows the same behavior as water alone. This is also found for lifetime studies,[8] so we shall assume that neither Na^+ or Cl^- are influencing the annihilation mechanism. Other cations studied were therefore dissolved as chlorides, and the anions were sodium compounds, except for $KMnO_4$ which for the present purpose is supposed to have essentially the same properties as $NaMnO_4$.

IV. DISCUSSION

A. Effect of Oxidation

The first part of the experiment was performed as a study of some paramagnetic ions. One expects an enhancement of the narrow component in these cases corresponding to increased triplet → singlet conversion caused by the unpaired electrons of the ions. This mechanism was inferred from the observed quenching of the long lifetime τ_2 by such ions.[7] Previous angular correlation studies on Mn^{++} and Co^{++} in water showed the expected increase of the narrow component.[5] As seen from Fig. 4 and Table I, we find that Mn^{++}, Co^{++}, and Ni^{++} produce an enhancement, while Cu^{++} and Fe^{+++} give rather a reduction of the narrow component. These results can not be explained by a change in conversion rate only.

As the two latter ions are both oxidizing, it was found to be of considerable interest to study other oxidizing substances as well. Such a study might also give further evidence for the suggestion by McGervey and DeBenedetti[9] that oxidizing ions might capture the electron of the positronium atom according to the reaction

$$A + (e^+e^-) \rightarrow A^- + e^+ \qquad (7)$$

where A is the oxidizing agent. Figure 5 and Table I contain the results for a number of solutes without any unpaired electrons, i.e., these substances are not supposed to cause conversion. In the table is also listed the highest chemical oxidation potential for the substance dissolved, as quoted by Latimer and Hildebrand.[11] The values obtained for I_N are plotted in Figs. 6 and 7 as a function of the oxidation potential. It is seen that nonmagnetic substances exhibit no increase of I_N above the value for water, as expected when no conversion takes place. For the oxidizing targets the amount of singlet positronium is generally reduced.

The ions NO_3^-, Cu^{++}, Fe^{+++}, and SbO^+ performing such a reduction have previously been studied with the lifetime technique.[7,8] For the nitrate solution no change was observed in the decay rate τ_2 of the triplet positronium, but a strong reduction was found in the amount I_2 of positrons annihilating with this lifetime. The other three ions gave the same limiting lifetime τ_2 (min) at high concentration as all the other τ_2-quenching substances that were tried. Green and Bell[7]

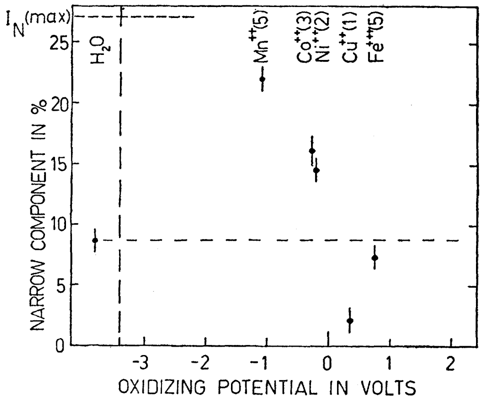

FIG. 6. Experimental values of the narrow component plotted as a function of the oxidizing potential for the dissolved paramagnetic ions. The narrow component for water is shown for comparison. Figures in parentheses indicate the number of unpaired electrons per ion.

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[11] W. M. Latimer and J. H. Hildebrand, *Reference Book in Inorganic Chemistry* (The Macmillan Company, New York, 1947).

interpret this result as an increase of the conversion rate against infinity, while no additional decay rate for positronium is occurring. These considerations indicate that the effect of the oxidizing substances is to reduce the formation of positronium rather than increasing the decay rate.

Ore[12] has discussed the formation of positronium in gases, and Ferrell[13] has applied his picture to the solid state. However, a similar discussion for the complicated liquid solution has to be a very rough approximation, at best. Figure 8(a) shows an electron energy diagram for the target, where V_i is the ionization potential and V_1 is the lowest excited electron state. After the last ionizing collision slowing down the positron, it is left in the energy region between V_i and zero, as shown by the total height of diagram 8(b). If now B is the binding energy of positronium, this state can only be obtained by positrons in the hatched area above V_i-B. Figure 8(c) is the region of kinetic energy of the positronium atom. Now this positronium may break up again in the subsequent collisions with atoms, in such a way that its electron is captured in some excited state of the target. This capture is illustrated by Eq. (7), which is the same as a chemical oxidation process. As seen from Fig. 8 electron capture can take place for kinetic positronium energies higher than

$$E_p=V_1-V_i+B. \qquad (8)$$

This is the width of the Ore gap, being equal to the region of positronium that is stable against break-up.

An increase in oxidation potential means increased electron affinity V_i-V_1. Thus, if the lowest excited electron state of the oxidizer is V_1' as in Fig. 8(a), the Ore gap is reduced to E_p' or it may vanish completely. This reduction is valid only for those positronium atoms that collide with the oxidizing atoms. The reduction in positronium formation is therefore dependent upon both concentration and the value of the chemical oxidation potential.

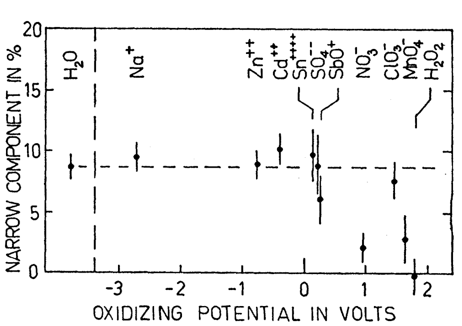

FIG. 7. Experimental values of the narrow component plotted as a function of the oxidizing potential for the dissolved nonmagnetic substances. The narrow component for water is shown for comparison.

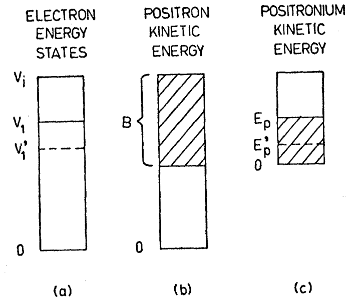

FIG. 8. Ore diagram for positronium formation. The symbols are explained in the text.

From Fig. 7 one may conclude that positronium formation is inhibited by electron capture in the solution of NO_3^-, MnO_4^-, and H_2O_2 in water. The ions ClO_3^- and SbO^+ are in this respect weak oxidizers. ClO_3^- has a high potential, but it may be that this ion is hydrated to such an extent that the surrounding water molecules prevent sufficient contact with positronium. The figure indicates that one may assign a small positive oxidation potential to the positronium atom, although more detailed studies would be required to establish its value with certainty.

B. Effect of Paramagnetic Ions

The conversion of triplet to singlet positronium can occur by means of electron exchange with the unpaired electrons of paramagnetic ions. Ferrell[14] has shown that this process can take place without spin-flip of the atomic electron. Therefore energy is not required and even positronium at rest can be converted in this way.

The ions Mn^{++}, Co^{++}, and Ni^{++} are not oxidizing, so their effect upon the narrow component is due to conversion only. It appears from Fig. 6 that the conversion due to these ions is roughly proportional to the number of unpaired electrons per ion. It can not be said conclusively whether this proportionality is accidental or not. But in any case, the observed varying degree of conversion is not in good agreement with the conclusion drawn from the lifetime measurements of Green and Bell,[7] who find that the slow component τ_2 approaches a common minimum value for all paramagnetic solutions as concentration increases. Assuming that the conversion rate is infinite and the pickoff annihilation rate λ_p is the same as for water they calculated the lifetime

$$\tau_2(\text{min})=4/[(\lambda_s+\lambda_p)+3(\lambda_t+\lambda_p)]$$
$$=3.9\times10^{-10}\text{ sec}, \qquad (9)$$

where λ_s an λ_t are spontaneous annihilation rates for singlet and triplet positronium, respectively. This agrees extremely well with the experimental value $\tau_2(\text{min})=4.0\times10^{-10}$ sec, which was very nearly obtained for all the targets at 2 moles per liter. Under these assumptions one easily finds that the narrow component would

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[12] A. Ore, Univ. Bergen Årbok Naturvitenskap. Rekke No. 9 and No. 12 (1949).

[13] R. A. Ferrell, Revs. Modern Phys. **28**, 308 (1956).

[14] R. A. Ferrell, Phys. Rev. **110**, 1355 (1958).

increase by the ratio

$$I_N(\max)/I_N(H_2O)=\lambda_s\tau_2(\min)=3.12.$$

In Fig. 6 the highest obtainable value $I_N(\max)$ is indicated as a broken line.

In the presence of Fe^{+++} and Cu^{++} the narrow component must be influenced by the two competing processes: reduction of positronium formation by the oxidation potential and triplet → singlet conversion due to the unpaired electrons of the ions. This is evidently in agreement with the results in Fig. 6. We note that the reduction of I_N is larger for Cu^{++} with only one unpaired electron as compared with Fe^{+++} having 5 unpaired electrons. Both processes might in principle have been observed in the lifetime experiment by Green and Bell,[7] but a reduction in I_2 is obviously very difficult to measure when τ_2 is small. As shown in Table I, these authors observe an anomalously large conversion cross section σ_{pc} for Fe^{+++} and Cu^{++}. One may ask whether this is incidentally so, or an example of reduced lifetimes τ_2 due to the oxidation potentials. In the latter case, the quenching of the τ_2 component found by Green and Bell for the weakly oxidizing $SbCl_3$ could also be explained without assuming paramagnetic properties for this substance. However, if one assumes that the decay rate of positronium is influenced by oxidation, this is obviously in disagreement with the idea of a constant $\tau_2(\min)$ as discussed above.

V. CONCLUSION

The experimental setup, which was equipped with 16 coincidence channels where ordinarily one channel was used, has been working fairly satisfactorily. For the present measurements the apparatus was operated manually, but it might well be made to work automatically. In that case it is clear that the angular correlation method need no longer be the slow and laborious way of obtaining data on positron annihilation.

The most easily obtained numerical data from these experiments are the values of the narrow component which equals the amount of singlet positronium that annihilates. The method that was developed to determine this quantity may not be the most correct one, but it is consistent and it gives good agreement with previously well-established data on water and $NaNO_3$ in water.

The main result of this work is the establishment of the fact that the variation in the amount of singlet positronium is determined by two main processes: a reduction of positronium formation due to electron capture by oxidizing substances and an increase of the triplet → singlet conversion due to electron exchange with paramagnetic ions. The study of the first effect showed that the oxidation potential of positronium is very near to zero, possibly slightly positive. For the second process a rough proportionality was found with the number of unpaired electrons, as might be expected.

As we do not find complete triplet → singlet conversion for any of the substances studied, our results are not in agreement with the conclusions of Green and Bell[7] who report a practically infinite conversion rate for the solutions studied here. It should be noted that these authors find an exceptionally high conversion cross section σ_{pc} for the oxidizing substances $FeCl_3$ and $CuCl_3$. In addition they assign a conversion effect to $SbCl_3$ which is not paramagnetic but also slightly oxidizing. It is therefore probable that the quenching of the τ_2 lifetime as given by σ_{pc} is not only due to electron exchange conversion, but is also dependent upon chemical effects including oxidation. If this is really the case, the questions arise: When $SbCl_3$ is not paramagnetic, why do lifetime experiments on solutions of this substance and on $NaNO_3$ give fundamentally different results? Would lifetime measurements on other oxidizing substances give quenching of τ_2 as by $SbCl_3$ or quenching of I_2 as by $NaNO_3$?

Obviously, it would be very valuable if various experimental techniques could be applied to annihilation in the same materials, measuring I_2, τ_2, I_N and possibly the 3-γ rate for different concentrations, different oxidation potentials and different numbers of unpaired electrons. This would certainly give a better understanding on many problems of the positron annihilation mechanism which are still not solved.

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