

## Depolarization of Negative Muons in Water and in Aqueous Solutions

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The dependence of negative-muon depolarization on the temperature and on the hydrogen-peroxide concentration was measured in various aqueous solutions placed in longitudinal and transverse magnetic field. It is shown that the experimental data are not inconsistent with the familiar concepts regarding the behavior of free radicals in aqueous solutions. The residual polarization in an aqueous solution of hydrogen peroxide is found to be 1.8 times higher than in pure water. This is interpreted as being the result of chemical interaction between mesic nitrogen and hydrogen-peroxide molecules, leading to the formation of diamagnetic compounds. It is shown that the degree of depolarization does not depend on the magnetic field strength. According to the depolarization model in which mesic-atom chemical reactions are taken into account, this signifies that the mesic atom enters into chemical reactions within a time  $t \approx 10^{-11}$  sec at  $T = 300^\circ\text{K}$ ; the paramagnetic products of these primary reactions, which contain mesic nitrogen, then participate in secondary reactions within a time  $t_1 \leq 10^{-7}$  sec. The rate constants of the reactions leading to the formation of diamagnetic products can be obtained by treating the concentration and temperature dependences of the depolarization in an aqueous solution of hydrogen peroxide in accordance with a model in which chemical reactions are assumed between the mesic atom and the  $\text{H}_2\text{O}_2$  molecules. The order of magnitude of the constant  $k \approx 3 \times 10^{-11} \text{ sec}^{-1} \text{ cm}^3$  is the same as that of the constant for reactions between the free H and OH radicals in water. The temperature dependences of the residual depolarization in water and in a solution of hydrogen peroxide in water are the same and consistent with the concept that mesic-nitrogen reactions are chemical reactions limited by diffusion.

As already shown in<sup>[1,2]</sup>, capture of a muon by an atom results in a mesic radical whose atomic number is one less than that of the atom of the medium. Depolarization of the muon on the K shell of such a mesic radical depends on the chemical interaction of the latter with the medium. Owing to the paramagnetism of the produced mesic radical, measurement of the asymmetry coefficient  $a$  in the angular distribution of the  $\mu^-$ -decay electrons, at the precession frequency of the free spin of the muon, should yield zero residual polarization.

The fact that measurements on many substances yield a nonzero asymmetry coefficient  $a$  indicates that the mesic radical succeeds in entering in a chemical compound with formation of a diamagnetic electron shell within a time  $t_{\text{ch.r.}}$  smaller than or at least comparable with the time of rotation of the total magnetic moment of the mesic radical in the transverse magnetic field.

It is known that the value of  $a$  is smaller for all the investigated substances than that for conductors<sup>[1,3,4]</sup>. It can be assumed that this is connected with the fact that the mesic radical or its paramagnetic compound survives until the time of observation of the decay electrons. By measuring the asymmetry of the decay electrons by counting the electrons at a definite angle, i.e., by eliminating the apparent depolarization due to the precession of the summary magnetic moment in the transverse magnetic field, we can determine the contribution made by the process in question. By performing such measurements in longitudinal magnetic fields with different intensities it is possible to determine simultaneously also the possible role of the local magnetic fields in the investigated substance.

We have proposed in<sup>[2]</sup> a model for the calculation of the dependence of  $a$  on the time required for the mesic radical to enter into the chemical reaction. To verify the main premises of the model proposed in<sup>[1,2]</sup>

and to reveal new possible channels of chemical interaction of the mesic atom produced when muons are stopped in water, (i.e., the nitrogen mesic radical  $\mu\text{N}$ , since the electron shell has the structure  $1s^2 2s^2 2p^3$  when one oxygen charge is screened by the muon), we measured the residual polarization (which is proportional to the asymmetry coefficient  $a$  in the angular distribution of the  $\mu^-$ -decay electrons) for different aqueous solutions and for heavy water, and also for different values of the longitudinal ( $H_{\parallel}$ ) and transverse ( $H_{\perp}$ ) magnetic fields in water and in aqueous solutions of hydrogen peroxide. The experiments were performed with a pure muon beam<sup>[5]</sup> from the meson channel of the JINR synchrocyclotron. The setup and the apparatus are described in<sup>[6]</sup>. The value of  $a$  was measured (with the exception of the experiment with  $H_{\parallel}$ ) by the method of precession at the frequency of the free spin of the muon.

### AQUEOUS SOLUTIONS

The targets were aqueous solutions of ammonia (ammonium hydroxide  $\text{NH}_4\text{OH}$ ), ammonium chloride ( $\text{NH}_4\text{Cl}$ ), hydrogen chloride (hydrochloric acid  $\text{HCl}$ ), potassium hydroxide ( $\text{KOH}$ ), hydrazine ( $\text{N}_2\text{H}_4$ ), and hydrogen peroxide ( $\text{H}_2\text{O}_2$ ). All the substances were of analytically-pure grade, and the admixture of the light water in the heavy water did not exceed 0.3%. All the targets had standard dimensions  $120 \times 120$  mm and an approximate thickness  $4 \text{ g/cm}^2$  along the beam. The beam polarization was monitored with a graphite target of the same dimensions.

Since the investigated solutions contain different atoms in which the muon has greatly differing life-times, the distribution with respect to the time (after stopping the muon in the target) of the number  $N_1$  of the registered decay electrons is given by

$$N_i(t_i) = N_0 \exp\{-t_i/\tau_0\} \{1 - a_0 \cos(\omega_0 t_i + \delta_0)\} + N_1 \exp\{-t_i/\tau_1\} \{1 - a_1 \cos(\omega_1 t_i + \delta_1)\} + C, \quad (1)$$

where  $t_i$  is the center of the  $i$ -th channel of the analyzer,  $\tau_0$  and  $\tau_1$  are the lifetimes of the muon in the oxygen of the water and in the atom of the dissolved substance,  $a_0$  and  $a_1$  are the measured asymmetry coefficients for oxygen and the impurity, respectively,  $N_0$  and  $N_1$  are the numbers of the electrons registered at zero time for the oxygen and impurity components, respectively,  $\omega_0$  and  $\omega_1$  are the precession frequencies, in a magnetic field  $H_1 \approx 50$  G, of the free spin of the muon and of the total magnetic moment of the muon and the nucleus respectively<sup>[7]</sup>, if the spin of the latter differs from zero,  $\delta_1$  is the initial phase of the precession and is determined by the geometry of the setup, and  $C$  is the random-coincidence background.

The parameters of this expression were determined by least-squares variation with a computer. The maximum number of the parameters ( $N_0$ ,  $N_1$ ,  $\tau_0$ ,  $\tau_1$ ,  $\alpha_0$ ,  $\alpha_1$ ,  $C$ ) was varied for the solutions of  $\text{NH}_4\text{Cl}$ ,  $\text{KOH}$  and  $\text{HCl}$ . In view of the fact that the exponentials for the  $\mu^-$  decay in potassium and in chlorine decrease much more rapidly than in oxygen, they amount to a negligible fraction of the investigated 5- $\mu$ sec time interval. The errors in the determination of  $a_1$  were large and the parameter  $a_1$  was subsequently disregarded. The obtained values of  $\tau_1$  for potassium and chlorine do not contradict the tabulated data<sup>[8]</sup>. For all the remaining substances we took into account only one exponential in the data reduction, since it was practically impossible to differentiate, by means of the lifetime, between the muon stopping by the nitrogen and oxygen atoms (this pertains also to the case of the  $\text{NH}_4\text{Cl}$  solution, when the  $\mu^-$  decay in the nitrogen and in the oxygen is described by one exponential), and the parameters  $N_0$ ,  $\tau_0$ ,  $a_0$ , and  $C$  were varied.

Since the precession frequencies of the muon-plus-nucleus system differ significantly from  $\omega_0$  in the case of atomic capture of a muon by nitrogen<sup>[7]</sup>, the contribution from  $\mu^-$  decay in nitrogen will decrease the value of  $a_0$  at the frequency  $\omega_0$ . Therefore the values of  $a_0$  obtained for aqueous solutions of  $\text{NH}_4\text{OH}$ ,  $\text{NH}_4\text{Cl}$ , and  $\text{N}_2\text{H}_4$  have been increased by an amount equal to the relative contribution of the stopping of the muon by the nitrogen, estimated in accordance with the Z-law<sup>[9]</sup>. Taking the contribution of the dissolved substance into account, the error of such an estimate can be neglected in comparison with the experimental errors. The values of  $C/N_0$  did not exceed 0.05.

Table I lists the values of  $a_0$  relative to  $a_C$  for graphite, referred to the same decay-electron registration threshold. The numbers in the parentheses indicate the concentrations of the dissolved substances in molar fractions. All the quantities were measured at room temperature. For comparison, Table I gives the value of  $a_0$  for  $\text{KOH}$  from<sup>[3]</sup>.

All the investigated solutions can be grouped in two categories. The solutions of  $\text{KOH}$  and  $\text{HCl}$  in water are strong electrolytes, since the dissociation constant of these compounds in water is  $K \approx 10^7$ <sup>[10]</sup>. The purpose of measuring the depolarization in such solutions was to determine the character of the interaction of the mesic atom  $\mu\text{N}$  with the ion-radicals  $\text{K}^+$ ,  $\text{H}^+$ ,  $\text{OH}^-$ , and  $\text{Cl}^-$ .

Table I.

Substance (concentration)	$a_0/a_C$	Substance (concentration)	$a_0/a_C$
$\text{H}_2\text{O}$	$0,403 \pm 0,018$	$\text{KOH}$ (0,15)	$0,328 \pm 0,034$
$\text{NH}_4\text{OH}$ (0,24)	$0,427 \pm 0,028$	$\text{KOH}$ (granules)	$0,170 \pm 0,050$
$\text{NH}_4\text{Cl}$ (0,11)	$0,396 \pm 0,064$	$\text{HCl}$ (0,07)	$0,429 \pm 0,078$
$\text{N}_2\text{H}_4$ (0,15)	$0,343 \pm 0,041$	$\text{HCl}$ (0,23)	$0,443 \pm 0,056$
$\text{H}_2\text{O}_2$ (0,14)	$0,691 \pm 0,029$	$\text{D}_2\text{O}$	$0,424 \pm 0,044$

According to present-day concepts<sup>[11,12]</sup>, the reactions between the neutral radicals  $\text{H}$  and  $\text{OH}$  and the ion-radicals  $\text{H}^+$ ,  $\text{OH}^-$ , and  $\text{Cl}^-$  in aqueous solutions are strongly suppressed in comparison with reactions between neutral radicals. This is attributed to the presence of hydrate shells in the ion-radicals. The mechanism governing the reactions in this case has not been clearly determined, although it is assumed that the neutral radical can react with the ion via a stage in which the ligand is replaced in the internal coordination sphere (hydrate shell), or an electron may be transported through the medium<sup>[12]</sup>.

The reaction of the mesic atom  $\mu\text{N}$  with the ions in aqueous solutions of an electrolyte might be revealed by the decrease of  $a_0$  in comparison with the case of pure water, since the fraction of the paramagnetic product of the interaction of  $\mu\text{N}$  increases. The fact that the values of  $a_0$  are the same (within 10%) for aqueous solutions of  $\text{KOH}$  and  $\text{HCl}$  and for pure water indicates that the reactions of  $\mu\text{N}$  with nonradicals make a negligible contribution (not larger than the statistical error).

The solutions of  $\text{NH}_4\text{OH}$ ,  $\text{NH}_4\text{Cl}$ , and  $\text{N}_2\text{H}_4$  are rather weak electrolytes (dissociation constants  $K \approx 10^{-5}$ – $10^{-6}$ <sup>[10]</sup>) and they can be regarded as molecular solutions, as is the aqueous solution of  $\text{H}_2\text{O}_2$ . It is known (cf., e.g.,<sup>[13]</sup>) that in aqueous solutions of non-electrolytes at concentrations  $\lesssim 0.10$ – $0.15$  molar fractions, regardless of the nature of the dissolved substance, the lattice of the hydrogen bonds becomes stronger as the result of the increase of the energy of the hydrogen bonds and of their number. The molecules of the non-electrolyte occupy in this case the voids in the structure of the water. The equality (within the limits of statistical errors) of the values of  $a_0/a_C$  for water and for aqueous solutions of  $\text{NH}_4\text{OH}$ ,  $\text{NH}_4\text{Cl}$ , and  $\text{N}_2\text{H}_4$  means that at a non-electrolyte concentration 0.1–0.2 molar fractions the value of  $a_0$  is insensitive, within 10%, to changes of the ordering of the water. The structure effects for aqueous solutions of the electrolytes  $\text{KOH}$  and  $\text{HCl}$ , at the concentrations used by us, should also be negligible.<sup>[14]</sup>

The strong increase of  $a_0$  (by 1.8 times) for the weak aqueous solution of hydrogen peroxide should most probably be attributed to the appearance of new channels (in comparison with pure water) for the reaction of the mesic atom  $\mu\text{N}$  directly with the  $\text{H}_2\text{O}_2$  molecules, leading to the formation of diamagnetic compounds. The interaction of  $\mu\text{N}$  with the neutral radicals  $\text{H}$ ,  $\text{OH}$ , and  $\text{HO}_2$  in the aqueous solution of  $\text{H}_2\text{O}_2$ , produced when the medium near the mesic atom is disintegrated<sup>[2]</sup>, can apparently be neglected, since the contribution of such a set of radicals is proportional to the molar fraction of  $\text{H}_2\text{O}_2$  and does not exceed 2%. The same seems to pertain also to the re-

maining solutions of the electrolytes and non-electrolytes. The lower value of  $a_0$  for the  $N_2H_4$  solution can be attributed to the increased content of atomic hydrogen in the radical zone, which should lead<sup>[2]</sup> to an increased fraction of the produced paramagnetic  $\mu N$  compounds. Naturally, more definite conclusions can be drawn by measuring  $a_0$  with better statistical accuracy and at larger concentrations of the dissolved substances.

The value of  $a_0$  for heavy water does not differ from that obtained for ordinary water. From the point of view of the general picture of the behavior of the mesic atom<sup>[1,2]</sup>, this result is reasonable, since isotropic effects produced upon disintegration of the medium around the mesic atom, and influencing the density of the radicals, or in the rate constants of the reactions of the radicals with the mesic atom<sup>[15]</sup>, should not be large.

### DEPENDENCE OF RESIDUAL POLARIZATION ON THE MAGNETIC FIELDS

The experimental setup is shown schematically in Fig. 1. The solenoid 5 made it possible to obtain a longitudinal (along the direction of the mesons in the beam) field up to  $H_{\parallel} \approx 2000$  G. The lead insert 8 prevented the muons from stopping and decaying in the aluminum form of the solenoid, and by the same token excluded the second component in the time distribution of the decay electrons, which was close in magnitude to the oxygen value. The exponential connected with the stopping in lead could be readily eliminated by shifting the start of the electron registration by several tenths of a microsecond. We used a water target of 80 mm diameter and 70 mm thickness (along the beam). No effect of the focusing of the decay electrons by the strong field  $H_{\parallel}$  was observed within the limits of the experimental accuracy ( $\pm 5\%$ ) (the experiment was performed with  $\mu^+$  mesons and a graphite target). The longitudinal field was measured accurate to  $\pm 5\%$ . The large error is due to the appreciable inhomogeneity  $H_{\parallel}$  in the solenoid over the volume of the target.

Helmholtz coils 6 produced a transverse magnetic field up to  $H_{\perp} \approx 200$  G accurate to within a fraction of 1%.

In the case of measurements in a transverse field,

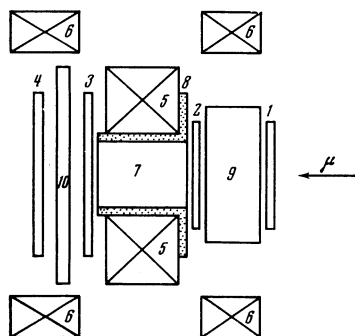


FIG. 1. Experimental setup (side view): 1 and 2—scintillators of muon telescope counters, 3 and 4—scintillators of decay-electron counters, 5—solenoid of longitudinal field, 6—Helmholtz coils for the transverse field, 7—targets surrounded by a lead insert, 8 and 9—muon moderator, 10—electron-telescope absorber.

the asymmetry coefficient was identified with the parameter  $a_0$  of the investigated Eq. (1), and in the case of measurements in a longitudinal field, the asymmetry coefficient  $a'$  was obtained by comparing the count  $N'$  at zero time in a longitudinal field with the count  $N_0$  at the zero instant in a transverse field, using the formula  $a' = -(N_0 - N')/N'$ . The asymmetry coefficients obtained in this manner are listed in Table II. We see that at room temperature the coefficient  $a'$  remains unchanged in the investigated range of  $H_{\parallel}$  and is equal to the coefficient  $a_0$  measured in a transverse field<sup>[4]</sup>. This means that at room temperature all the paramagnetic compounds containing the mesic atom  $\mu N$  enter in the subsequent chemical reactions prior to the start of the observation. An attempt to "freeze out" the chemical activity of the radicals containing the mesic atoms by lowering the temperature to 77°K has shown that even at this temperature the chemical interactions of such radicals terminate within a time much shorter than several times  $10^{-7}$  sec. This agrees with the general ideas concerning the behavior of radicals in condensed media<sup>[16,17]</sup>. The free radicals containing mesic atoms ( $\mu NH$ ,  $\mu NH_2$ , etc.) vanish apparently as the result of the relatively slow secondary reactions with impurities within a time on the order of  $10^{-8}$  sec<sup>[17]</sup>.

The independence of  $a'$  of  $H_{\parallel}$  shows that the local magnetic fields that can cause a decrease of the coefficient  $a_0$  in comparison with the value characteristic of conductors<sup>[3]</sup> do not exist in water.

As already noted above, measurement of  $a_0$  as a function of the intensity of the transverse magnetic field  $H_{\perp}$  permits an independent determination of the time at which the mesic atom enters the first chemical reaction. If this time is of the order of  $t \approx 1/\Omega$ , then a decrease of  $a_0$  by an approximate factor of two should be observed when  $H_{\perp}$  is increased from 20 to 200 G; on the other hand, if there is no dependence on the field, then the time at which the mesic atom enters the chemical reaction is of the order of  $t \approx 1/\nu \approx 10^{-11}$  sec<sup>[2]</sup> (at room temperature). It is also shown in<sup>[2]</sup> that under certain conditions there should be observed a dependence of the initial phase  $\delta$  of the precession on  $H_{\perp}$ . Under the conditions of the present experiment, however, the problem of determining  $\delta$  with the required accuracy could not be solved.

Table II. Values of the asymmetry coefficient of the decay electrons in longitudinal and transverse magnetic fields

$T, ^\circ K$	$H_{\parallel}$	$a'$	$H_{\perp}$	$a_0$
Pure water				
300	1265	$0.0171 \pm 0.0035$	50	$0.0202 \pm 0.0018$
	1000	$0.0241 \pm 0.0058$	200	$0.0207 \pm 0.0025$
	800	$0.0146 \pm 0.0058$		
77	200	$0.0110 \pm 0.0019$	50	$0.0105 \pm 0.0013$
$H_2O + 15$ wt. % $H_2O_2$				
300			43	$0.0295 \pm 0.0022$
			190	$0.0262 \pm 0.0017$
$H_2O + 7.5$ wt. % $H_2O_2$				
300			24	$0.0308 \pm 0.0020$
			47	$0.0292 \pm 0.0016$
			70	$0.0285 \pm 0.0022$
			106	$0.0274 \pm 0.0021$
			197	$0.0290 \pm 0.0020$

The values of  $a_0$  measured as a function of  $H_{\perp}$  in the indicated range are listed in Table II. They were corrected for the difference in the number of points per precession period. The reduction of the results obtained for the solution of hydrogen peroxide by least squares, using the equation  $a_0 = a_0^0 + bH_{\perp}$ , yields  $a_0^0 = 0.0298 \pm 0.0015$  and  $b = -(1.28 \pm 1.45) \times 10^{-5} \text{ G}^{-1}$ . Thus, there is no dependence of  $a_0$  on  $H_{\perp}$  within the limits of errors. A similar result is obtained also for pure water (see Table I). We can therefore conclude that the mesic atom enters into chemical reactions in water and in aqueous solution of hydrogen peroxide within a time on the order of  $10^{-11}$  sec (at room temperature), i.e., the temperature dependence of the residual polarization in the water is due to the competition between the fast chemical reactions of the mesic atom with the depolarization due to the hyperfine interaction of the magnetic moment of the muon with the magnetic moment of the electron shell.

#### TEMPERATURE AND CONCENTRATION DEPENDENCES OF THE DEPOLARIZATION IN AQUEOUS SOLUTION OF HYDROGEN PEROXIDE

Within the framework of the model that takes into account the chemical interactions of the mesic atom, an increase of the residual polarization in an aqueous solution of  $\text{H}_2\text{O}_2$  can be the consequence of either an increase of the probabilities  $\lambda$  and  $\lambda_0$  of chemical reactions with respective formation of a diamagnetic and an arbitrary compound, or only of an increase of the contribution of  $\lambda/\lambda_0$  of reactions leading to the diamagnetic channel (at equal values of  $\lambda_0$ ). The choice between these two possibilities can be made by measuring the temperature dependence of the residual polarization in an aqueous solution of hydrogen peroxide. If  $\lambda_0$  for the solution of  $\text{H}_2\text{O}_2$  is much larger than for water, then the temperature dependence should be weak, since  $t_1$  is certainly smaller than  $t$  (see<sup>[3,41]</sup>). On the other hand, if  $\lambda_0$  for water and for the  $\text{H}_2\text{O}_2$  solution is the same, and only the ratio  $\lambda/\lambda_0$  changes, then the temperature dependence of the depolarization in  $\text{H}_2\text{O}_2$  should have the same form as for water. Measurement of the dependence of the depolarization on the  $\text{H}_2\text{O}_2$  concentration makes it possible to determine the rate constant of the corresponding chemical reaction.

Figure 2 shows the obtained dependence of the ratio  $a/a_C$  ( $a_C$  is the asymmetry coefficient for graphite) on the temperature at an  $\text{H}_2\text{O}_2$  concentration of 7.5 wt.%. The temperature was measured with several thermocouples inserted into the target. Prior to the start of the measurements, the target was placed in a container of foamed polystyrene and frozen to the temperature of liquid nitrogen (77°K). As the target was being heated, the information was recorded at different time intervals corresponding to different temperature intervals. During the first ten minutes the temperature was maintained close to 77°K and the corresponding experimental point has therefore a very small error in terms of the temperature scale. Subsequently the temperature started to rise at an average rate of 1 deg/min, and the succeeding three points (at 121, 165, and 204°K) are mean values obtained in an approximate tempera-

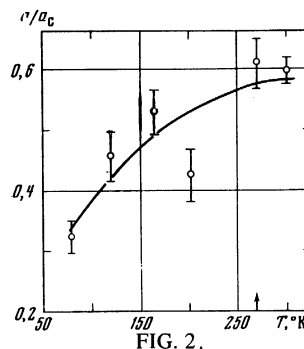


FIG. 2.

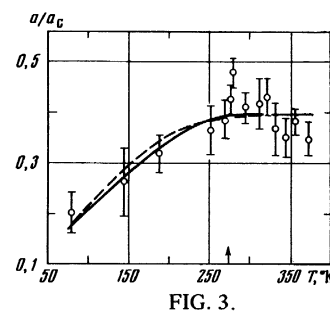


FIG. 3.

FIG. 2. Temperature dependence of the residual polarization of muons in an aqueous solution (7.5 wt. %) of hydrogen peroxide. The solid line corresponds to the value  $F = 1/3$ . The arrow indicates the temperature of the phase transition.

FIG. 3. Temperature dependence of the residual polarization in water. The experimental data are taken from [6]. The solid and dashed curves are the result of calculation for  $F = 0.2$  and  $0.4$ , respectively.

ture interval  $\pm 15^\circ$  (these limits are not indicated in the figure). The points at 272 and 300°K were obtained under stationary conditions with a small temperature spread, just as the point at liquid-nitrogen temperature. The indicated procedure with freezing followed by heating the target was repeated several times.

According to<sup>[2]</sup>, the temperature dependence of  $a_0/a_C$  can be described by the approximate expression

$$\frac{a_0}{a_C} = \frac{A_0}{a_C} \frac{\lambda}{\lambda_0} \left\{ F + (1-F) \text{th} \frac{\lambda_0}{2\nu} \right\}. \quad (2)$$

Here  $A_0$  is the value of the asymmetry coefficient at the end of the interatomic cascade (prior to the start of action of the paramagnetic-depolarization mechanism),  $F = \frac{1}{3} [1 + 2/(2J + 1)^2]$  (see<sup>[18]</sup>), and  $J$  is the total angular momentum of the electron shell of the mesic atom. If it is assumed that only the quantity  $\lambda_0$  depends on the temperature, then we can put  $\lambda_0 = \Lambda (T/300)^n$  ( $\Lambda$  is the value of  $\lambda_0$  at  $T = 300^\circ\text{K}$ ) and write down the expression for the temperature dependence of  $a_0/a_C$  in the form

$$\frac{a_0}{a_C} = \frac{A_0}{a_C} \frac{\lambda}{\lambda_0} \left\{ F + (1-F) \text{th} \left[ \frac{\Lambda}{2\nu} \left( \frac{T}{300} \right)^n \right] \right\}. \quad (3)$$

The experimental data on the temperature dependence of the depolarization in the  $\text{H}_2\text{O}_2$  solution were reduced by least squares with variation of the parameters  $A_0\lambda/a_C\lambda_0$ ,  $\Lambda/2\nu$ , and  $n$  at specified values of the parameter  $F$  in the range from 0.2 to 0.4. The result of such a reduction is shown in Fig. 2 by the solid line (variation of  $F$  between the limits indicated above does not affect the shape or the position of the curve). The values of the other parameters for  $F = \frac{1}{3}$  are given in Table III. For comparison, we have reduced in similar fashion the experimental data on the temperature dependence in water<sup>[4]</sup>. The results of the calculation for the case of water are shown in Fig. 3 and in Table III for  $F = \frac{1}{3}$ .

If we assume the possibility of transitions in a transverse magnetic field between mesic-atom hyperfine structure states differing in the value with different mesic-atom spin projections on the direction of the magnetic field (see<sup>[18]</sup>), then the value of  $F$  for the ground state of the mesic atom can be smaller than  $\frac{1}{3}$ .

Table III.

Substances	$a_C \lambda_0 / A_0 \lambda$	$\Lambda / 2\nu$	$n$	$\chi^2$	$\Lambda \cdot 10^{-10} / \text{sec}^{-1}$
H <sub>2</sub> O + H <sub>2</sub> O <sub>2</sub> (7,5%)	1,7 ± 0,2	2,4 ± 1,5	1,4 ± 0,4	10,1	6,4 ± 4,8
H <sub>2</sub> O	2,5 ± 0,1	3,2 ± 2,4	2,3 ± 1,1	18,5	4,8 ± 3,0

If the rate of such transitions were much larger than  $\nu$ , then we would have  $F_{\min} = 0.2$ . It is precisely these considerations that dictated the choice of the values of  $F$  in the reduction of the data on the temperature dependences.

The values of the parameters and their errors, given in Table I for  $F = 1/3$ , differ insignificantly from those obtained at values of  $F$  from 0.2 to 0.4, but when  $F > 0.4$  the errors in the parameters become much larger than the quantities themselves. Since the interval 0.2–0.4 is reasonable for  $F$  from the most general point of view, the fact that the parameters are independent of  $F$  within these limits indicates that such a result is stable.

It follows from Table III that the values of the parameters  $\Lambda/2\nu$  and  $n$  for water and for the aqueous solution of H<sub>2</sub>O<sub>2</sub> coincide within the limits of errors, and the values of the parameter  $A_0 \lambda_0 / a_C \lambda_0$  differ appreciably. The temperature dependences have all the same form, meaning that the aqueous solution is H<sub>2</sub>O<sub>2</sub> differs from pure water only in the ratio  $\lambda/\lambda_0$  (we assume that  $a_0/a_C$  is the same for H<sub>2</sub>O and H<sub>2</sub>O<sub>2</sub>). The calculated curves describe quite well the experimental data, in spite of the fact that in the reduction of the data for water and the H<sub>2</sub>O<sub>2</sub> solution we did not take into account the possible jump in the value of  $a_0/a_C$  at the phase-transition point.

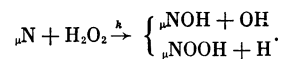
The exponent  $n$  in the temperature dependence of the depolarization has approximately the same value for water and for the H<sub>2</sub>O<sub>2</sub> solution. To obtain information on the temperature dependence of the rate constants of the chemical reactions from data on  $n$ , it is necessary to carry out special investigations of the temperature dependence of the concentrations of the different radicals, which can reveal also to a temperature dependence of the parameter  $\lambda/\lambda_0$ , for example as the result of the different temperature dependences of the rates of diffusion of the radicals H and OH. We can assume roughly, however, that  $\lambda/\lambda_0$  does not depend on the temperature, and ascribe the measured temperature dependence only to the rate constant of the chemical reaction. Then the obtained value  $n \approx 1.5$  can serve as evidence that the mesic-atom reactions are chemical reactions limited by diffusion. Indeed<sup>[20]</sup>, if the rate constant of the reaction in the aqueous solution becomes larger than  $10^{10}$ – $10^{11}$  liter-mole<sup>-1</sup> sec<sup>-1</sup>, then this means that it approaches the rate of molecular collisions resulting from the diffusion of the reagents, and takes place practically at each collision. According to the theory of diffusion-limited reactions, the rate constant should be proportional to  $T/\eta$ , where  $\eta$  is the viscosity of the solvent. Since  $\eta$  increases little with decreasing  $T$ , the experiment should reveal a temperature dependence with an exponent  $n$  somewhat larger than unity.

Using the value  $\nu = 10^{10}$  sec<sup>-1</sup> calculated on the basis of<sup>[21]</sup> for the mesic atom  $\mu\text{N}$ , we can obtain the values of  $\Lambda$  given in the last column of Table III. The

fact that the value of  $a_0/a_C$  for water and for the aqueous solution of H<sub>2</sub>O<sub>2</sub> is independent of the temperature at room temperature enables us to write for this temperature region a simple expression for the concentration dependence:

$$\frac{a_0}{a_C} = \frac{A_0}{a_C} \frac{\lambda^{(1)}(1-R) + \lambda^{(2)}R + k[\text{H}_2\text{O}_2]}{\Lambda^{(1)}(1-R) + \Lambda^{(2)}R + k[\text{H}_2\text{O}_2]} \quad (4)$$

Here  $\lambda^{(1)}$ ,  $\lambda^{(2)}$ ,  $\Lambda^{(1)}$ ,  $\Lambda^{(2)}$  are the reaction rates;  $K$  is rate constant of the reactions of  $\mu\text{N}$  with H<sub>2</sub>O<sub>2</sub>; the indices 1 and 2 correspond to reactions with the radicals produced upon disintegration of the molecules H<sub>2</sub>O and H<sub>2</sub>O<sub>2</sub>, respectively;  $[\text{H}_2\text{O}_2]$  is the concentration of the hydrogen-peroxide molecules per cm<sup>3</sup>;  $R = [\text{H}_2\text{O}_2] / ([\text{H}_2\text{O}_2] + [\text{H}_2\text{O}])$  is the relative molar concentration of H<sub>2</sub>O<sub>2</sub>;  $[\text{H}_2\text{O}_2] = A\rho/M_{\text{H}_2\text{O}_2}$ ,  $[\text{H}_2\text{O}] = (A/M_{\text{H}_2\text{O}}) \times (1 - \rho/d)$ ;  $A$  is Avogadro's number;  $M$  is the molecular weight and  $d$  is the density of pure hydrogen peroxide ( $d = 1.462$  g/cm<sup>3</sup>), and  $\rho$  is the concentration of the hydrogen peroxide in g/cm<sup>3</sup>. It is assumed in (4) that all the reactions of  $\mu\text{N}$  with H<sub>2</sub>O<sub>2</sub> lead to the diamagnetic channel:



The numerator of the right-hand side of (4) is the rate at which the mesic atom enters in the reaction in which diamagnetic products are formed, and the denominator is the rate of entry into any reaction at room temperature. Using the numerical values of the coefficients, we obtain

$$\frac{a_0}{a_C} = \frac{A_0}{a_C} \frac{\lambda^{(1)}(1-\tilde{\rho}) + \lambda^{(2)}\tilde{\rho} + 1.8 \cdot 10^{22} \rho k}{\Lambda^{(1)}(1-\tilde{\rho}) + \Lambda^{(2)}\tilde{\rho} + 1.8 \cdot 10^{22} \rho k}, \quad \tilde{\rho} = \frac{\rho}{1.9 - 0.3\rho} \quad (5)$$

The maximum concentration of hydrogen peroxide in our experiment was  $\rho_{\max} = 0.33$  g/cm<sup>3</sup> or 18% in terms of the number of molecules. Since  $\tilde{\rho}$  does not exceed 0.18, we can simplify (5) and rewrite it in the form

$$\frac{a_0}{a_C} = b \frac{1 + c\rho}{1 + f\rho}, \quad (6)$$

$$b = \frac{A_0 \lambda}{a_C \Lambda}, \quad c = \frac{1.8 \cdot 10^{22} k}{\lambda}, \quad f = \frac{1.8 \cdot 10^{22} k}{\Lambda}$$

The points and error bars in Fig. 4 show the experimental dependence of  $a_0/a_C$  on the H<sub>2</sub>O<sub>2</sub> concentration in units of  $10^{20}$  cm<sup>-3</sup> (at  $T = 300^\circ\text{K}$ ). The smooth curve is the result of a least-squares reduction with variation of the parameters  $c$  and  $f$ . We see that the curve describes the experimental data quite satisfactorily ( $\chi^2 = 5.5$ ). The value of the parameter  $b = 0.384 \pm 0.18$

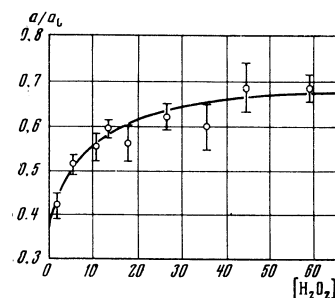


FIG. 4. Dependence of the residual polarization in aqueous solution of hydrogen peroxide on the concentration of the latter, expressed in units of  $10^{20}$  cm<sup>-3</sup>. The solid curve was calculated by least squares.

corresponds to the asymmetry coefficient in pure water. The two remaining parameters are found to be  $c = 20.0 \pm 6.3$  and  $f = 10.5 \pm 3.3$ .

Using the same value  $\Lambda \approx 5 \times 10^{10} \text{ sec}^{-1}$  for water and for the aqueous solution of  $\text{H}_2\text{O}_2$  (at  $T = 300^\circ\text{K}$ ) and the obtained value of the parameter  $f$ , we obtain the value  $k \approx 3 \times 10^{-11} \text{ sec}^{-1} \text{ cm}^3$  for the rate constant of the reaction  $\mu\text{N}$  with  $\text{H}_2\text{O}_2$ .

We note one more important circumstance. A noticeable change of  $a/a_C$  occurs for the solution of  $\text{H}_2\text{O}_2$  in water at an  $\text{H}_2\text{O}_2$  concentration of the order of  $10^{21} \text{ cm}^{-3}$ . This means that to explain the value of  $a/a_C$  measured in water it is necessary to assume approximately the same absolute concentration of the radicals  $\text{H}$  and  $\text{OH}$ .

Taking the concentration of the  $\text{H}_2\text{O}$  molecules ( $3.34 \times 10^{22} \text{ cm}^{-3}$ ) into account, we can state that to explain the short lifetime of the mesic atom in water ( $\sim 10^{-11} \text{ sec}$  at room temperature) one must have a rather small degree of disintegration of the matter in the region of the mesic atom. Since the disintegration of the medium at the end of the muon track and in the case of deceleration of the fragments of a "Coulomb explosion" are similar in character to those produced by the recoil atoms in nuclear transformations, the conclusion that the disintegration of the medium near the mesic atom is weak and the conclusion concerning the time of its chemical stabilization agree with the data obtained by different methods for the case of recoil atoms<sup>[22]</sup>.

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<sup>1</sup>V. S. Evseev, JINR Preprint R-14-4052, Dubna, 1968.

<sup>2</sup>A. A. Dzhuraev and V. S. Evseev, Zh. Eksp. Teor. Fiz. **62**, 1166 (1972) [Sov. Phys.-JETP **35**, 615 (1972)].

<sup>3</sup>A. A. Dzhuraev, V. S. Evseev, G. G. Myasishcheva, Yu. V. Obukhov, and V. S. Roganov, Zh. Eksp. Teor. Fiz. **62**, 1424 (1972) [Sov. Phys.-JETP **35**, 748 (1972)].

<sup>4</sup>V. S. Evseev, V. S. Roganov, V. A. Chernogorova, G. G. Myasishcheva, and Yu. V. Obukhov, Yad. Fiz. **8**, 741 (1968) [Sov. J.

Nucl. Phys. **8**, 431 (1969)].

<sup>5</sup>V. G. Varlamov, Yu. M. Grashin, A. V. Dem'yanov, B. A. Dolgoshein, and V. S. Roganov, JINR Preprint I-4084, Dubna, 1968.

<sup>6</sup>A. I. Babaev, M. Ya. Balats, G. G. Myasishcheva, Yu. V. Obukhov, V. S. Roganov, and V. G. Firsov, Zh. Eksp. Teor. Fiz. **50**, 877 (1966) [Sov. Phys.-JETP **23**, 583 (1966)].

<sup>7</sup>A. I. Babaev, V. S. Evseev, G. G. Myasishcheva, Yu. V. Obukhov, V. S. Roganov, and V. A. Chernogorova, Yad. Fiz. **10**, 964 (1969) [Sov. J. Nucl. Phys. **10**, 554 (1970)].

<sup>8</sup>A. O. Vaisenberg, Myu-mezon (The Mu Meson), Nauka, 1964.

<sup>9</sup>S. S. Gershtein, V. I. Petrukhin, L. I. Ponomarev, and Yu. D. Prokoshkin, Usp. Fiz. Nauk **97**, 31 (1969) [Sov. Phys.-Usp. **12**, 1 (1969)].

<sup>10</sup>Spravochnik khimika (Chemist's Handbook), Vol. 3, Khimiya, 1964.

<sup>11</sup>E. T. Denisov, Usp. Khim. **39**, 62 (1970).

<sup>12</sup>E. T. Denisov, Usp. Khim. **40**, 43 (1971).

<sup>13</sup>Yu. I. Naberukhin and V. A. Rogov, Usp. Khim. **40**, 369 (1971).

<sup>14</sup>O. Ya. Samoilov, Struktura vodnykh rastvorov élektrolitov i gidratsiya ionov (Structure of Aqueous Solutions of Electrolytes and Hydration of Ions), AN SSSR, 1957. A. M. Blokh, Struktura vody i geologicheskie protsessy (The Structure of Water and Geological Processes), Nedra, 1969.

<sup>15</sup>Energii razryva khimicheskikh svyazei. Potentsialy ionizatsii i srodstvo k elektronu (Chemical-bond Breaking Energies. Ionization Potentials and Electron Affinity), (Handbook), AN SSSR, 1962.

<sup>16</sup>I. V. Vereshchinskii and A. K. Pikaev, Vvedenie v radiatsionnyu khimiyu (Introduction to Radiation Chemistry), AN SSSR, 1963. A. K. Pikaev, Impul'snyi radioliz vody i vodnykh rastvorov (Pulsed Radiolysis of Water and Aqueous Solutions), Nauka, 1965.

<sup>17</sup>P. J. Dyne, Kennedy, Can. J. Chem. **36**, 1518 (1958).

<sup>18</sup>V. A. Dzhrbashyan, Zh. Eksp. Teor. Fiz. **36**, 277 (1959) [Sov. Phys.-JETP **9**, 188 (1959)].

<sup>19</sup>A. P. Bukhvostov, Yad. Fiz. **9**, 107 (1969) [Sov. J. Nucl. Phys. **9**, 65 (1969)]; I. I. Gurevich, I. G. Ivanter, E. A. Meleshko, B. A. Nikol'skii, V. S. Roganov, V. I. Selivanov, V. P. Smilga, B. V. Sokolov, and V. D. Shestakov, Zh. Eksp. Teor. Fiz. **60**, 471 (1971) [Sov. Phys.-JETP **33**, 253 (1971)].

<sup>20</sup>E. F. Caldin, Fast Reactions in Solution, Wiley, 1964 [Russ. transl., Mir, 1966, p. 20].

<sup>21</sup>S. Goudsmit, Phys. Rev. **37**, 663 (1931); M. A. Heald and R. Beringer, Phys. Rev. **96**, 645 (1954).

<sup>22</sup>S. I. Bondarevskii, A. N. Murin, and P. P. Seregin, Usp. Khim. **40**, 95 (1971).

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