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CHEMICAL REACTIONS OF MUONIUM. IDENTIFICATION OF THE PRODUCTS OF INTERACTION WITH MATTER

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The dependence of the μ^+-meson spin precession on the chemical compound into which the meson enters is considered for different magnetic fields. It is shown that an identification by classes of compounds is possible, as is a finer differentiation with respect to the precession frequencies of the meson-nuclear spin system.

It was shown by Byakov and the author [1] that the coefficient of spatial asymmetry in the decay of μ^+ mesons in a medium is connected with the rate constant of the reaction of the hydrogen-like muonium atom (Mu) with the substance. The depolarization of the μ^+ meson when the latter enters into the composition of the molecule stops.[2]

A kinetic analysis of the aggregate of the processes which occur shows that the connection between the chemical reaction-rate constant of the chemical reaction between muonium and the substance RH and the experimentally observed asymmetry coefficient c' is determined by the expression

$$\frac{2k_5+2(k_1+k_2)[\mathrm{RH}]}{k_4} + \left(1+\frac{2k_3[\mathrm{RH}]}{(k_1+k_2)[\mathrm{RH}]+k_5}\right)^{-1} = \frac{c'}{c-c'}. \qquad (1)$$

Here k_1 is the rate constant of the reaction that leads to the formation of molecular products containing muonium, for example MuH; k_2 is the rate constant of the reaction producing radicals of the type $\mathrm{RMuH}^{\cdot}$ (the dot denotes the unpaired electron); k_3 is the rate constant for the triplet-singlet conversion (conversion insures a decrease in c' below half the maximum possible value of c, which is equal to $1/3$ for fully polarized mesons); k_4 is the depolarization constant of singlet muonium Mu^S due to the spin flip; k_5 is the μ^+-meson decay constant; [RH] is the concentration of the substance. The constants k_1, k_2 and k_3 can depend on the energy of the muonium atom. However, inasmuch as the time of slowing down of the epithermal (3—10 eV) muonium atom is considerably smaller than either the μ^+-meson lifetime or the characteristic time of the chemical reaction, we can assume that the prevailing contribution is made by the reactions of muonium with thermal energy.

In the derivation of formula (1), the polarized products are assumed to be Mu, the molecules MuH, and the radicals $\mathrm{RMuH}^{\cdot}$, which holds true in the absence of external magnetic fields. Inasmuch as in the experiment one customarily observes the spin precession in a constant magnetic field, it is expedient to examine the behavior of the products in this case.

In order for the electron energy to exceed the energy of interaction between the magnetic moments of the electron and the meson in the muonium, it is necessary to apply an external magnetic field exceeding the critical value $H'_{\mathrm{cr}} = 1580$ G.[3] In smaller fields, the spin system of the meson and of the electron in the triplet muonium (Mu^T) precesses at a frequency which is ~ 103 times larger than the precession frequency of the free meson (muonium frequency). In a diamagnetic molecule having closed electron shells and pairwise compensated electron spins, the field necessary to break the bonds of the magnetic moments of the meson and of the nucleus must exceed $H''_{\mathrm{cr}} \lesssim 20$ G. The latter depends on the magnetic moment of the nucleus m_{nuc} and the meson-nucleus distance r, since the field produced by the nucleus at the point where the μ^+ meson is located is proportional to m_{nuc}/r^3. When $H > H''_{\mathrm{cr}}$, the precession will take place with a frequency corresponding to the frequency of the free meson; when $H < H''_{\mathrm{cr}}$, the frequency is determined by the total magnetic moment and spin of the system. By virtue of the fact that H_{cr} is quite sensitive to r, we can in most cases take into account only the nucleus closest to the meson (when $r > 3$ Å we have $H''_{\mathrm{cr}} < 0.2$—0.5 G).

For radical products, taking into account the interaction of the magnetic moments of the meson and of the unpaired electron of the neighboring atom (in organic radicals $r \approx 1-1.5$ Å if the electron does not migrate over the chain of carbon atoms), analogous estimates yield $H'''_{cr} \approx 60-200$ G. In larger fields, precession takes place with meson frequency; when $H''_{cr} < H < H'''_{cr}$ the precession takes place with muonic frequency.

Thus, determination of the frequency and amplitude of the precession in different magnetic fields makes it possible to identify the products of the chemical reactions by classes of compounds and to determine their contribution. Indeed when $H'_{cr} > H > H'''_{cr}$, the precession with mesonic frequency is due to mesons which are contained in the molecules and the radicals, whereas the muonium, by virtue of the high precession frequency, can be regarded as depolarized under the experimental conditions. When $H''_{cr} < H < H'''_{cr}$, the molecular products cause precession with mesonic frequency, and the radicals and Mu—with muonic frequency. Inasmuch as simultaneous observation of these frequencies is difficult to attain in experiment, the bulk of the experimental data have been obtained on the amplitude of the mesonic frequency at the indicated limits of the external magnetic field. For $H < H''_{cr}$, precession of the spin system will be observed for the molecular products.

The groups of products can be separated into electronic and mesonic components, the composition of the components depending on the magnitude of the external magnetic field. The electronic component includes Mu or Mu with RMuH$^\cdot$, while the mesonic component includes MuH or MuH with RMuH$^\cdot$ at the external-field limits indicated above. Naturally, the sum of the components is equal to the total sum of the products.

If the rate of the chemical reaction is determined by the condition $V = k[RH] \gg k_5$, then the stationary concentration of the muonium is small compared with the stationary concentrations of the other products (a confirmation of this fact for several compounds follows from a series of experimental papers[4,5]), and the problem of separating the components greatly simplifies: when $H < H'''_{cr}$ one determines the radicals (muonic frequency) and the molecular products (mesonic frequency), and when $H > H'''_{cr}$ one determines their sum.

In general form, when taking into account the influence of the reaction of formation of radicals on the rate of formation of molecular products, we obtain an expression for the mesonic component

$$\frac{k_1[\mathrm{RH}]}{k_2[\mathrm{RH}]+k_5+k_4/A}=\frac{c'}{c-c'} \tag{2}$$

and analogously for the electronic component

$$\frac{k_2[\mathrm{RH}]+k_5}{k_1[\mathrm{RH}]+k_4/A}=\frac{c'}{c-c'}, \tag{3}$$

where

$$A=2+\frac{k_4}{(k_1+k_2)[\mathrm{RH}]+2k_3[\mathrm{RH}]+k_5}.$$

When reducing the right sides of formulas (1)—(3) to the form c'/c [it is obvious that the c' in formulas (2) and (3) are different], the sum of (2) and (3) yields formula (1). The corresponding plots of c' against the chemical reaction-rate constant are shown in Fig. 1. The continuous lines for the different concentrations of the substance—the muonium acceptor—shows the summary values of the asymmetry coefficient in the absence of magnetic fields (polarized products Mu, MuH, RMuH$^\cdot$), while the dashed lines represent the variation of the mesonic component (MuH or MuH with RMuH$^\cdot$), and the dash-dot lines show the electronic components. We see that when $k_1 \geq 10^6$ l/mole sec ([Ac] = 10) the amplitude of the muonic frequency is negligibly small if, of course, k_2 is small.

An interesting object, from this point of view, is benzene, for which, according to [6] and [7], $k_1 \sim 0$ and $k_2 = 5.9 \times 10^8$ liter/mole-sec at room temperature. This should correspond (depending on the value of k_3) to $c' \sim 0$ at $H < H'''_{cr}$ and $c' \sim 25\%$ when $H > H'''_{cr}$. At 50 G ($H < H'''_{cr}$) the experimental value is $c' = (4.6 \pm 1.2)\%$.[4] The agreement should apparently improve if we recognize that the presence, for example, of air oxygen diluted in the benzene can yield $c' = 3.6\%$ even in the absence of a contribution from the main substance. Since in this case $H'''_{cr} \sim 80-100$ G, we should expect an appreciable increase of c' with increasing H.

An investigation of the amplitude and frequency

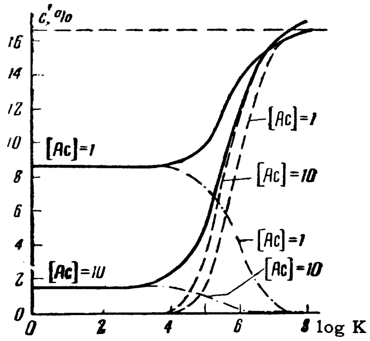

FIG. 1. Dependence of the coefficient c′ on the chemical reaction-rate constant k, liters/mole-sec.

Compound	Spin	Magnetic moment	Precession frequency	ρ	Compound	Spin	Magnetic moment	Precession frequency	ρ
		in meson units					in meson units		
μ^+	$^1/_2$	1	1	1	o - $MuCl^{35}$	2	0.908	0.227	0,377
Mu^T	1	~206	~103	0,5	p - $MuCl^{35}$	1	1.092	0.546	0,377
Mu^S	0	~208	—	0.5	o - $MuCl^{37}$	2	0.923	0,231	0,123
o - MuH	1	0,686	0,343	0,5	p - $MuCl^{37}$	1	1.077	0,538	0,123
p - MuH	0	1,314	—	0,5	o - $MuBr^{79}$	2	0.763	0,191	0.253
o - MuD	$^3/_2$	0,904	0,301	0,5	p - $MuBr^{79}$	1	1,237	0,618	0.253
p - MuD	$^1/_2$	1,096	1.096	0,5	o - $MuBr^{81}$	2	0.745	0,186	0.247
o - MuF	1	0,704	0,352	0.5	p - $MuBr^{81}$	1	1.255	0,628	0.247
p - MuF	0	1,296	—	0.5	o - MuI	3	0.684	0,114	0,5
					p - MuI	2	1.316	0,329	0.5

of the precession when $H < H''_{cr}$ can lead to interesting conclusions. Let us consider, for example, the molecule MuH. For polarized mesons, the probabilities of formation of the molecule with parallel and antiparallel meson and proton spin orientations (ortho- and para-MuH) are equal. The magnetic moment of the meson, in nuclear magnetons, is equal to 8.8903[8] with $m_p = 2.79275$.[9] The total moment of o-MuH is 6.0976 or, in fractions of the meson magnetic moment, m = 0.686 (the magnetic moments of the electrons are compensated). Inasmuch as the spin of the system is S = 1, the precession frequency of o-MuH in fractions of the μ^+-meson frequency is equal to $\omega = 0.343$ with an amplitude equal to the initial value for the free meson at $H > H''_{cr}$ (the contribution corresponds to the fractional yield $\rho = ½$). For the p-MuH molecule, the spin is S = 0 and there is no precession ($\rho = ½$). The summary coefficient of the decay asymmetry for the MuH molecules is reduced to one-half. The table lists the corresponding values for several simple molecules.

For molecules in the same state, but different in their isotopic composition (for example o-$MuCl^{35}$ and o-$MuCl^{37}$), as can be seen from the table, the precession frequencies are close, whereas for molecules with different spin states the frequencies differ by a factor of 2—3 and can be readily separated by harmonic analysis. Figure 2 gives an example of the behavior of the molecules MuH. When H > H″ precession takes place with mesonic frequency, and when H < H″ a sharp change takes place in the frequency and in the amplitude of the precession. Since for diatomic molecules listed in the table the ground state is $^1\Sigma$, the contribution of the spin-orbit interaction can be disregarded. It is clear that the rotational quantum number must be small. Deviations from the simple relation considered can be estimated in more complicated cases from the experimental data.

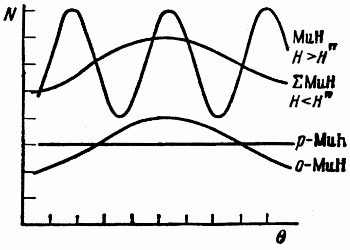

FIG. 2. Precession of the μ^+-meson spin in the MuH molecule at different magnetic field intensities.

The identification of the products of interaction between muonium and matter by this method is comparable, to some degree, with the hyperfine structure in nuclear magnetic resonance. The presence of the corresponding frequencies indicates unambiguously the form of the molecule containing muonium. Some estimates, in addition, can be obtained from H''_{cr} for various molecules which we list below:

Compound:	MuH	MuD	MuF	$MuCl^{35}$	$MuCl^{37}$
H''_{cr}, arb. un.:	10,00	3,07	4,97	0,58	0,48

Compound:	$MuBr^{79}$	$MuBr^{81}$	MuI
H''_{cr}, arb. un.:	1,10	1,19	0,99

Thus, the diatomic molecules considered can be arranged in the following series relative to the intensity of the external magnetic field necessary to break the meson-nuclear interaction:

$$MuH > MuF > MuD > MuBr > MuI > MuCl.$$

It is curious that, owing to the small value of the magnetic moment of chlorine, MuCl does not fit the general rule for the variation of the properties of hydrogen halides.

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