

Dependence of the asymmetry of negative muon decay electrons on the length of the hydrocarbon chain of alcohols and alkyl chlorides

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Relative (with respect to graphite) values of the asymmetry coefficients a/a_C for the angular distribution of $\mu^- \rightarrow e^-$ decay electrons from muons stopped in aliphatic alcohols $C_nH_{2n+1}OH$ and in chlorine-substituted paraffins $C_nH_{2n+1}Cl$ with various amounts of carbon atoms in the chain are obtained by the method of muon spin precession in a weak transverse magnetic field. With increasing n_C , the ratio a/a_C passes through a maximum and then approaches a limit, the values of which are determined for paraffin and polyethylene. The derived dependence is interpreted on the basis of the concept that chemical interactions of mesic atoms lead to the formation of diamagnetic compounds and the concept regarding energy migration along the carbon atom chain.

We have previously observed^[1-3], using the method of precession of the free spin of a muon in a weak transverse magnetic field, that the degree of depolarization of μ^- mesons stopped in different media depends on the parameters of these media (the temperature, the molecular weight, the chemical structure, the impurity concentration, etc.). These facts cannot be explained by the cascade-polarization theory developed for the case of a polarized mesic atom^[4] and without taking into account the effect of the electron shell^[5].

A hypothesis was advanced later^[6,7] to explain how the residual polarization of the μ^- mesons can depend on the properties of the medium surrounding the mesic atom. It follows from this hypothesis that to explain the experimental data it is necessary first to know the state of the electron shell of the mesic atom after its stabilization, and the stabilization time. If the shell is paramagnetic (as is observed when muons are stopped in substances whose atomic nuclei have zero spin), then the muon spin is connected with the magnetic moment of the electron shell. Under the conditions of the experiments in^[1-3], a noticeable residual polarization at the free-muon-spin precession frequency could have been observed only under one condition, namely that the electron shell of the mesic atom become diamagnetic within a time that does not exceed a certain characteristic value for the given mesic atom; this time is determined by the hyperfine-interaction constant between the magnetic moments of the muon and the electron shell.

The situation is quite similar to that obtaining when μ^+ mesons are stopped and atomic muonium is produced^[8]. In either case the cause of the rapid compensation of the paramagnetism of the mesic atom or of the muonium is considered to be some interaction with the medium, principally a fast chemical reaction. However, since the mesic atom is produced in highly excited states and reaches the ground state via Auger or radiative cascade transitions, its environment differs significantly from the environment of muonium.

It was noted earlier^[7] that for most compounds in which the negative-muon depolarization was measured, thermal chemical reactions of a mesic atom with neutral ones and with unexcited molecules of the medium are either difficult or energywise forbidden. It is therefore necessary to consider the reactions of a mesic atom with the decay products of the medium, mainly with the radicals produced near the mesic atom, as the result of the

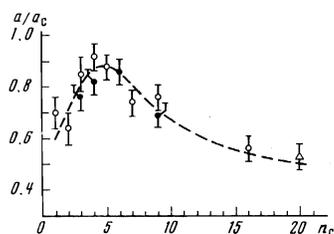
following three principal processes: a) disintegration of the medium molecules by the decelerating muon prior to atomic capture, b) disintegration of the medium by deceleration of the Auger electrons emitted in mesic-atom transitions; c) disintegration of the medium by fragments of the "Coulomb explosion" of the molecule^[9] which is ionized by the Auger transitions in the mesic atom. Unlike muonium, a mesic atom can be produced immediately in a bound state, and it is then necessary to consider the change in the paramagnetism of the radical that contains the mesic atom. The foregoing premises enable us to explain qualitatively (and in some cases also quantitatively), with the aid of the phenomenological theory developed for molecular media^[7], the experimentally-observed dependences of the depolarization and the characteristics of the medium. The parameters used in the theory have a definite physical meaning, and their values obtained by reduction of the experimental data do not contradict each other and the notion that the mesic atom can be regarded as a rather active radical. Within the framework of these premises, the depolarization of the μ^- mesons should depend on the density of the radicals near the mesic atom, and any phenomenon contributing to a change in the density of the radical should exert an influence on the residual polarization.

Let us see how a phenomenon like the intermolecular or intramolecular migration of excitation energy can influence the degree of depolarization. The term "energy migration" has been introduced to designate nonradiative replacement of the energy of a quantum of electronic excitation or the transport of vibrational or rotational excitation energy without losses, over distances exceeding interatomic distances^[10]. It is a sufficiently well established fact that energy migrates along a chain of carbon atoms in molecules of aliphatic alcohols^[11], in polymers^[12], in solutions of organic compounds containing the phenyl group^[13], etc. It is precisely to the energy migration processes that it is proposed to attribute the difference between the degrees of polarization of cyclic and aromatic hydrocarbons^[1] and of monomer and polymer organic compounds^[3]. The residual polarization decreases in all those cases (aromatic hydrocarbons, polymers with saturated bonds) when one can expect migration of part of the energy released upon production of the mesic atom from the region of its interaction, and by the same token a decrease of the radical density in this band.

For a more detailed investigation of the connection between μ^- -meson polarization and the process of intramolecular migration of energy, we have measured in this study, by the method of precession of free spin of the muon (at room temperature), the relative values of the residual polarization (relative to graphite), a/a_C , in several series of aliphatic alcohols $C_nH_{2n+1}OH$ and chlorine-substitute paraffins $C_nH_{2n+1}Cl$. The experiments were performed with a pure muon beam from the meson channel on the synchrocyclotron of the Nuclear Problems Laboratory of JINR. The apparatus and the procedure have been described earlier (see, e.g., [1-3, 14]).

The experimental data are shown in the figure. The value of a/a_C in the compounds of both type first increases with increasing number n of the carbon atoms in the chain, goes through a maximum, and decreases to a limiting value approximately equal to 0.5, as determined by the value of the asymmetry coefficient of the limiting compound for these series, namely polyethylene [1]. For alcohol, the figure shows the measured values of a/a_C , which are the values, averaged over the probability of the atomic capture, for the mesic boron and mesic nitrogen produced when the muon lands on the carbon and oxygen atoms. In the case of the alkyl chlorides, the values of a/a_C pertain only to mesic boron. Since the points for the alcohols and the alkyl chlorides fit on one and the same curve, this indicates that the governing factor is indeed the length of the hydrocarbon chain.

The results can be interpreted in the following manner. So long as the molecule dimensions do not exceed the dimensions of the zone in which the chemical reactions of the mesic atom takes place, the intermolecular migration of the part of the energy released upon production of the mesic atom does not lead to a considerable decrease of the density of the radicals within the limits of this zone, inasmuch as the entire energy is ultimately released here. The growth of the quantity a/a_C for alcohols with $n = 1-5$ with increasing n [3] can be attributed here to a number of causes. First, an increase of the chain length is accompanied by an increase in the total number of valence electrons in the molecule. Because of their high "mobility," individual negative charge migration [15] occurs along the chain of carbon atoms to the particular atom that has turned into a mesic atom and in which Auger transitions with ionization of the surrounding parts of the molecule take place. The more valence electrons contained in the molecule, the more favorable the conditions for the Auger transitions, and consequently the more Auger electrons are emitted and the more radicals are produced in the mesic atom. Second, when the chain of the alcohol molecule with small n becomes longer, its radiative stability decreases [16], and consequently the radical density increases,



Dependence of the relative residual polarization in alcohols (○) and alkyl chlorides (●) on the number n of the carbon atoms in the chain. Data for alcohols with $n = 1-5$ were taken from [3], and for paraffin (△) from [1].

and with it the contribution of those radicals which form diamagnetic compounds with the mesic atom. With increasing molecular weight of the alcohol, its viscosity increases, and this also can contribute to an increase in the density of the large radicals in the interaction zone, via a decrease in their diffusion rate.

With increasing n , part of the energy leaves the zone of the chemical reactions of the mesic atom. This leads to a decrease in the radical density and to a decrease of a/a_C . The tendency of a/a_C to a limit at large values of it can be explained by assuming the presence of energy migration at a distance larger at least than 5-10 links of the chain, which does not contradict the concept advanced in [11]; in this case the radius of the chemical-interaction zone of the mesic atom is smaller than the dimensions of the molecule with such a long chain. As a consequence of specific conformational properties of chain molecules, their dimensions are much shorter than those of a chain stretched out in one line, the length of which at $n = 5-10$ would be 12-25 Å [17].

If we compare the average radius r of the zone of chemical reactions of the mesic atom with the average distance s between two ends of the molecule, then for $n = 10$ we have $s \approx 10$ Å [17], and consequently $r \lesssim 10$ Å. It is assumed here that the conformational properties of the molecules of the aliphatic alcohols and of the alkyl chlorides are the same as those of paraffins. On the other hand, one can estimate the value of r by using the expression $r \approx (2Dt)^{1/2}$ (where D is the diffusion coefficient of the mesic atom and t is the average time of entry of the mesic atom into the chemical reaction, determined by reducing the temperature dependence of a/a_C [2, 3]) and assuming [7] that the reactions of the mesic atom in condensed media are described by the theory of diffusion-limited chemical reactions [18].

The value of D for mesic boron (at $n = 10$, the contribution of mesic nitrogen can be neglected) in alcohols with $n \approx 10$ can be estimated in the following manner. Since $D \sim 1/\eta$ (η is the viscosity of the medium) [18] and at room temperature the diffusion coefficient of mesic boron in water is not larger than that of atomic hydrogen, i.e., $D_{\mu B} \lesssim 10^{-4}$ cm² sec⁻¹ [19], it follows that the viscosity in alcohol with $n \approx 10$ is larger by approximately one order of magnitude than in water, and consequently $D_{\mu B} \lesssim 10^{-5}$ cm² sec⁻¹. The average time required for the mesic atom to enter into the chemical reaction lies in the range $10^{-11}-10^{-10}$ sec [2, 3], and $r \lesssim 1.5-5$ Å. This value does not contradict the one obtained above, if account is taken also of the initial range (5-10 Å) of the "hot" mesic atom, which had acquired energy as a result of a "Coulomb explosion" or cascade transitions.

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