A SEARCH FOR ATOMIC MUONIUM IN CHEMICALLY INERT SUBSTANCES

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The behavior of triplet muonium in several substances when temperature and other conditions are varied is investigated by observing its (spin) precession in a transverse magnetic field. The presence of atomic muonium in crystal and fused quartz, in solid carbon dioxide, and in ice is demonstrated. The results are discussed from the viewpoint of the physical and chemical interactions of muonium.

1. INTRODUCTION

WHEN (polarized) μ^+ mesons are slowed down to thermal velocities in matter they are in an energetically favorable condition for the formation of muonium (μ^+e^-) atoms by detaching outer electrons from the atoms of most moderating substances. Several workers have investigated questions concerning the formation and subsequent possible depolarization of muonium.[1-3] Attempts have also been made to observe free muonium experimentally. For example, the presence of muonium in gaseous argon has been demonstrated by the microwave method.[4] Since triplet muonium precesses in a transverse magnetic field at a frequency that is 103 times greater than that of a free μ^+ meson, the former can be identified by the conventional method of observing its decay time distribution in a transverse magnetic field of suitable strength.[5]

Muonium atoms, which resemble hydrogen atoms in many ways, can interact with nearby matter. The spin precession frequency of a μ^+ meson incorporated into a molecule will equal the precession frequency of a free meson when the (covalent) chemical bond is formed by an electron pair with mutually compensating spins. Therefore our present search for triplet muonium was performed under experimental conditions that largely prevented chemical reactions.

2. APPARATUS

To observe the decay time distribution in a transverse magnetic field (the precession curves) we used the same apparatus and electronic circuitry as in our earlier work (Fig. 1).[6] A few modifications improved mainly the resolving time of the coincidence schemes. Of importance also was the "stretching" of pulses from the synchrocyclotron of the JINR Nuclear Physics Laboratory[7] which we used for our experiments. By utilizing only the second (stretched) accelerator beam peak we completely eliminated modulation of the random coincidence background by the lower frequency of accelerating voltage (with ~70 nsec period). The precession curves were distorted by this modulation when the respective periods were close.

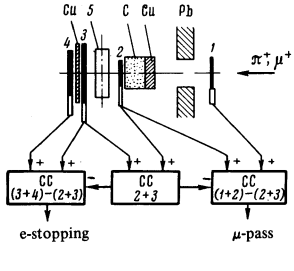

FIG. 1. Apparatus for observing time distribution of μe decay. 1,2,3, and 4 – scintillation counters; 5 – target; CC – coincidence circuits. The Hemholtz coils, compensating coils, and a portion of the electronics are not shown.

The mean pulse of beam particles was increased up to 190 MeV/c (by varying the conditions of μ production[8]). Increases were thus effected in both the μ^+ flux and the initial (~85%) beam polarization. The better positioning of the moderating filter (compared with[6]) narrowed the μ^+ stopping curve (5 g/cm^2 half-width). Practically no contamination (<2%) by $\pi \rightarrow \mu \rightarrow e$ decays was registered. The mean counting rate of the stoppings was 3000/sec in a ~5-g/cm^2 target. As a result of the "stretching" the accidental-coincidence background was considerably reduced, and in most of the experimental work did not exceed 3% of the level observed in the initial segment of the decay curve.

The transverse magnetic field was generated by a pair of Helmholtz coils; the field inhomogeneity within the target was under 1%. External magnetic fields were compensated along three mutually perpendicular axes by means of additional coils. Permalloy pickups measured the magnetic field strength with absolute accuracy ±0.05 Oe. The thickness of matter in the different targets was about 5 g/cm^2 (converted to carbon target thickness in accordance with their respective ionization losses: R_{eff}). For low temperature measurements we also used a thermally insulated foam plastic sheath (with ~0.07 g/cm^2 wall thickness in the beam direction).

3. RESULTS

The absolute asymmetry parameters c′ were calculated (after computer processing of the experimental data) by averaging the values of c′ obtained for the actual test target and a bromoform ($CHBr_3$) target taken as a standard. Our earlier measurements[6] and those in[9] show that the decay asymmetry parameter in $CHBr_3$ for precession with meson frequency is extremely close to the possible maximum. The experimental result for bromoform is c′ = 0.280 ± 0.006.

Several control experiments performed with carbon bromoform, and cadmium targets of different thicknesses (Fig. 2) show that the corrections introduced in some cases for the thickness and atomic number Z of the target material are unimportant compared with the statistical accuracy of the experiments.

Table I.

Substance	T, °C	c_0	Damping, sec	c'
Fused SiO_2	+30°	0.161±0.012	1.3±0.2	0.050±0.006
	−196°	0.148±0.014	0.5±0.1	0.048±0.007
Crystalline SiO_2	+30°	0.167±0.013	0.4±0.1	0.056±0.006
Solid CO_2	−78°	0.070±0.015	0.4±0.1	0.038±0.013
	−196°	0.075±0.015	0.6±0.1	—
Solid H_2O	−196°	~0.16	~0.08	0.066±0.004

Table II.

Substance	T, °C	c_0	c'
Powdered SiO_2	+30°	0.02±0.02	0.110±0.005
Powdered Al_2O_3	+30°	0.04±0.05	0.111±0.007
Liquid N_2	−196°	0.01±0.02	0.037±0.006
Fused B_2O_3	+30°	0.02±0.03	0.127±0.006
	−196°	0.01±0.03	—
Benzene	−196°	0.02±0.03	—
Polyethylene	−196°	0.01±0.02	—
Paraffin	−196°	0.01±0.02	—

Tables I and II give the measured asymmetry parameters for spin precession at the precession frequency of triplet muonium in a magnetic field H = 7.2 Oe. Table I gives data derived from experiments in which the precession was observed; Table II gives data from experiments in which precession with the triplet muonium frequency was not detected. The tables also give the experimental temperatures and the asymmetry parameters of meson-frequency precession that were determined in separate experiments with H = 50 Oe and ~6 μsec observation time.

It should be noted that in all cases precession at muonium frequency is damped more or less rapidly. For each substance, therefore, the tables give the asymmetry parameter c_0 extrapolated to "zero" time (the instant of μ^+ stopping in the target) and the measured damping time (assuming exponential damping). The actual resolving time (±2.5 nsec) of the electronics and the distortions of the decay time distribution near the "zero" time that resulted from the finite duration of the anticoincidence signals (in counters 2 and 3) prevent observation of precession that is damped in less than 20—40 nsec. In the experiment with ice (T = −196°C) the field was increased to 29 Oe because it was necessary to observe several periods of rapidly damped precession. In this latter case the extrapolated initial asymmetry parameter can be subject to considerable error. In the computer processing of the measurements the experimental initial phases and observation periods of the precession agreed with the calculated values used.

Figures 3—5 show the precession curves of triplet

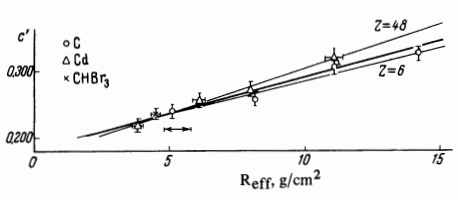

FIG. 2. Measured asymmetry parameter c'(at meson frequency) vs, target thickness. The double arrow shows the working size of the targets.

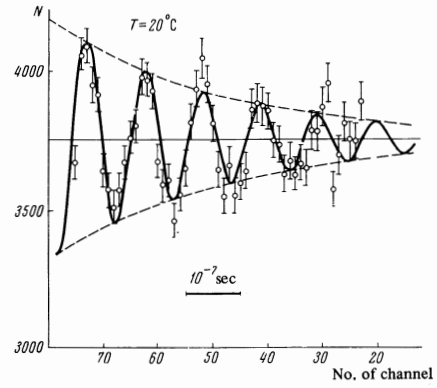

FIG. 3. Muonium precession curve in crystalline quartz. Horizontal axis – number of channel (10.1 nsec wide); vertical axis – counts corrected for exponential decay of meson.

muonium at different temperatures in the cases of crystalline and fused quartz, and of ice at −196°C. The very presence of muonium constitutes a convincing proof that these substances are chemically inert for the present purpose, i.e., there are no rapid chemical processes occurring that could yield molecular products constituted with the muonium.

4. DISCUSSION OF RESULTS

The observed extinction of the precession can be induced by spin-depolarizing processes (such as conversion, i.e., the transition of triplet muonium into the singlet state, followed by rapid depolarization), and also by the involvement of muonium in a "slow" chemical reaction. However, depolarization does not occur, strictly speaking, in the latter instance. Extinction is associated only with the method utilized here to observe spin precession in a transverse magnetic field. Indeed, at a sufficiently slow rate of chemical interaction a transition to precession with the meson frequency can occur at any instant. Even at the meson frequency pre-

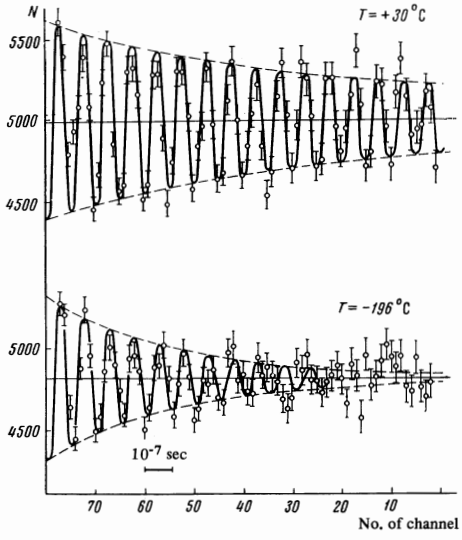

FIG. 4. Muonium pression curves in fused quartz at different temperatures, with 18.5-nsec channel width.

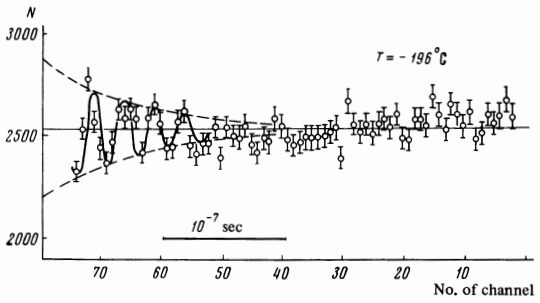

FIG. 5. Muonium precession curves in ice, with 4.8-nsec channel width.

cession averaged over all initial phases is not observed. A comparison of experimental results obtained with and without a transverse magnetic field will permit a clearer discrimination of processes that are associated with true depolarization of muonium.

In substances that exhibit both muonium- and meson-frequency precession at an identical temperature the latter appears to result either from free μ^+ mesons (if the probability of muonium formation is less than unity) or from muonium involved in a chemical reaction within the epithermal region (because the reaction rate of thermalized atoms is quite low). For fused quartz confirmation is found in the constancy of the asymmetry parameters measured at meson frequency for different temperatures.

It is interesting to compare the observation of muonium-frequency precession in fused quartz at both room and very low temperatures. Since both chemical and conversion processes (if we regard conversion as an exchange of electrons in different spin states when thermal collisions occur between muonium and atoms of the ambient) should be impeded at very low temperatures, the experimentally observed faster extinction of precession at -196°C is incomprehensible from this point of view. A role may possibly be played here by contraction of the "phase volume," a factor that was considered for the case of positronium atoms in [10].

We have attempted an experimental verification of one possible explanation for the foregoing situation, retaining the picture developed in [2]. It can be assumed that in fused quartz at -196°C the conversion rate is dominant over the chemical reaction rate, and that the damped precession of triplet atomic muonium is observed. At room temperature a relatively rapid chemical reaction occurs, resulting in the formation of both an electron pair with compensating spins and an unpaired electron whose magnetic moment is associated with the magnetic moment of the meson. This muonium-like radical [3] will precess like free muonium in a magnetic field of insufficient strength to break this bond ($H < H_{crit}$, where H_{crit} is the bond-disrupting field). Theoretical calculations yield $H_{crit} = 1580$ Oe for atomic muonium. [11] Since the electron-meson separation is larger in the radical than in the muonium atom, the critical field is considerably lower in this case. Based on the dimensional parameters of the quartz lattice, H_{crit} is found to be of the order of several tens of oersteds. Fused quartz was tested at room temperature and at the low temperature in a transverse magnetic field H = 37 Oe. If for the radical (assuming a correct hypothesis regarding its formation) $H_{crit} < 37$ Oe, the initial asymmetry parameter for precession with the muonium frequency should be considerably reduced. At the low temperature an increase of the field from 7 to 37 Oe should not seriously affect the experimental result because the critical field for the muonium atom is much higher than 37 Oe.

In these experiments the spin precession has a period of the order of 20 nsec; because of the finite resolving time of the electronics some "instrumental" reduction of the precession amplitude can occur. From this point of view the experiment at -196°C served as a control. In the two experiments the precession amplitude was identical within statistical error limits. The electronics limited the possibility of applying higher fields.

Muonium precession was observed in acoustic crystalline quartz for two cases of mutually perpendicular crystallographic axes (along and perpendicular to the beam); the results coincided. It must be emphasized, however, that the extinction time of precession in crystalline quartz at room temperature is notably shorter than in fused quartz under similar conditions and is close to the results obtained for fused quartz at -196°C. The presence of triplet muonium atoms in the crystal structure of ice is of fundamental importance. Since a closed coordination system of hydrogen bonds is formed at the low temperature, only conversion processes could yield a probability for the existence of muonium. The observation of muonium in the aforementioned system is interesting in connection with searches, by means of EPR, for atomic hydrogen formed by radiolysis of ice.

In the case of a fine SiO_2 powder (with $1-10\mu$ grains) the search for muonium precession yielded a negative result. The simultaneous steep increase of the asymmetry parameter measured at meson frequency provides evidence that surface defects have an appreciable effect (electron traps, for example). All of the foregoing can be applied to the results of an experiment with a target made of powdered aluminum oxide (Al_2O_3).

Since a muonium atom experiences about 10^6 collisions with neighboring atoms during a decay time of the order of 1 μsec, even a relatively insignificant amount of chemically active material can appreciably reduce the lifetime of polarized muonium. For example, we can expect muonium to exist in liquid nitrogen (at -196°C). It is known [12] that the presence of 0.01—0.001% of oxygen in gaseous argon can strongly reduce the muonium signal by the microwave method. If this last effect is associated with chemical reactions of O_2, then oxygen should produce no effect at low temperatures, since conversion processes are evidently temperature-independent. When nitrogen was used the oxygen impurity constituted 0.1—0.3%; thus the absence of polarized muonium in this case indicates the considerably greater influence of the paramagnetism of the excited-triplet O_2 molecule. Similar conclusions were derived from the results in recent work by Mobley et al. [13]

For several other substances such as polyethylene, paraffin etc. where muonium precession was not detected, it is unfortunately impossible to guarantee a sufficient degree of material purity or the absence of surface defects.

5. CONCLUSIONS

The results provide a basis for imposing certain requirements on the materials in which muonium atoms could be observed (under the experimental conditions that are achievable at present). Among such requirements are the absence or extremely slow course of chemical reactions leading to the formation of molecular products, a small contribution from lattice defects and from other circumstances leading to the formation of electron traps, the absence of chemically active or paramagnetic impurities etc.

The rate of conversion exchange, which depends implicitly on the electron configuration of the test molecules, also considerably affects the lifetime of polarized muonium. Investigations of muonium interaction processes, when muonium is observed directly, can elucidate the roles of different factors. An understanding of the physical picture presented by the formation and behavior of atomic muonium is extremely important when one considers chemical reactions in which muonium participates.

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