# Formation of the muonic HF molecule during the stopping of negative muons in a neon-hydrogen mixture

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(Submitted 15 April 1993)
Pis'ma Zh. Eksp. Teor. Fiz. **57**, No. 12, 741–745 (25 June 1993)

The residual polarization of negative muons has been measured by the $\mu$SR method in an apparatus with a gaseous target, either pure neon or a $Ne+H_2$ mixture, at room temperature in a transverse magnetic field of 200 G. In pure neon (24 atm) a precession of the polarization of the muon frequency was not observed. The measured asymmetry coefficient in the mixture Ne (24 atm) $+H_2$ (16 atm) is $0.59\pm0.09\%$. The results are explained in terms of the formation of a muonic HF molecule. The results show that it is possible to use polarized negative muons to study fast physical and chemical processes in the gas phase.

Muon spin relaxation ($\mu$SR) is used widely and successfully to study the properties of materials and the dynamics of chemical interactions (see, for example, Ref. 1 and the bibliography there). Essentially all the experiments, however, have used positively charged muons, so there have been only limited opportunities for using the method to study interactions with a medium consisting of a light analog of the hydrogen atom or its ion. The use of negative muons would add substantially to the list of elements which could be studied, since the capture of a $\mu^-$ by an atom with atomic number $Z$ would result in the formation of a single muonic atom or ion in the medium which would be a chemical analog of an atom with an atomic number $Z-1$.[2] Attempts have been made to use the $\mu^-$SR method to study chemical reactions in condensed media.[3] However, the uncontrollable rate of the processes which lead to depolarization of the muon introduces such a large uncertainty and such a large ambiguity in the interpretation of the data that this direction has not been widely developed.

In the study which we are reporting here we have attempted to develop a method for carrying out experiments of this type in the gas phase. Specifically, we studied the reaction of muonic fluorine with hydrogen. The idea underlying the method is as follows: At the time at which a diamagnetic molecule forms, the hyperfine coupling of the magnetic moment of the muon with the paramagnetic electron cloud of the muonic atom or ion is disrupted. If the muon has not become completely depolarized by this instant, the $\mu$SR spectrum in an external magnetic field $B$ directed transverse with respect to the muon spin should exhibit a precession signal at the frequency $\omega_\mu=\gamma_\mu B$ of the free muon ($\gamma_\mu=8.52\times10^4\ s^{-1}\cdot G^{-1}$). There is accordingly the practical possibility of determining the rate at which the molecule forms and of working from the evolution of the polarization to identify the channels which lead to depolarization, by varying the composition of the medium, the total pressure of the gas mixture, and the strength of the external magnetic field.

  

We selected for study the HF molecule, which forms in an $Ne+H_2$ mixture when a negative muon is stopped in it. The Coulomb capture of the muon by the Ne atom leads to the formation of a muonic atom (in the first step, it is a multiply charged ion) with an effective atomic number $Z_{eff}=9$ and the electronic structure of fluorine.[2] The particular components of the mixture were chosen because (first) neon is chemically inert, (second) the reaction $F+H_2 \rightarrow HF+H$ has a large rate constant ($K > 10^{-11}$ cm$^3$/s; Refs. 4 and 5) {because of this large rate constant, the HF molecule is formed in a time comparable to the time scale of the hyperfine interaction of the muon with the electron cloud of fluorine[6] ($10^{-10}$ s)}, and (third) the probability for the capture of a muon by an Ne atom is five times that for capture by an $H_2$ molecule. Furthermore, when the HF molecule is observed, there is the possibility of determining the residual polarization of the muon in the $K$ shell of Ne, i.e., $P_K$. This is an important point, since measurements of $P_K$ through observation of a free muonic fluorine atom yield unstable results.[2,7]

The measurements were carried out in the $\mu$ line of the phasotron of a laboratory at the Joint Institute for Nuclear Research with the help of the $\mu$SR apparatus.[8] We had used the apparatus previously to measure the residual polarization of $\mu^-$ muons in gaseous deuterium.[9] The gaseous target, designed for pressures up to 50 atm, is a cylinder made of nonmagnetic stainless steel with a volume $\approx$ 9 liters. The axis of the cylinder coincides with the beam direction. Two silicon semiconductor detectors, one behind the other, are positioned in the target on the beam side. The stopping of muons in the gas volume is detected by selecting threshold values of the energy evolution in the semiconductor detectors as muons pass through them. The heights of the signals from the semiconductor detectors are stored on magnetic tape. During subsequent processing, it is possible to select muons stopped both in the gas and in the detectors themselves quite accurately. As a result, the background of $\mu \rightarrow e$ decays from the stopping of muons in the detectors was eliminated essentially completely, and the background from stopping events in the target housing was suppressed to $< 10\%$. A magnetic field of 200 G at the target, directed transverse with respect to the beam and having a value constant within $< 1\%$ over the target volume, was generated by means of Helmholtz coils. The electrons from $\mu \rightarrow e$ decay were detected by three telescopes, each consisting of two scintillation counters connected in coincidence. Two of the telescopes were positioned beside the target, parallel to the beam; the third was behind the target, transverse with respect to the beam. The gaseous Ne and $H_2$ used in the experiments were of a high-purity commercial grade. The purity of the Ne was raised during the filling of the target by passing the gas through a zirconium getter (at 650 °C). Since the primary problem at this stage of the research was to determine whether it is possible to observe the formation of an HF molecule, we exposed the target to the $\mu^-$ beam twice: Once in pure Ne and once in a mixture Ne (24 atm) + $H_2$ (16 atm). The time-resolved spectra from the three telescopes were processed jointly by the least-squares method (we used the FUMILI program) with the help of a fitting function

$$F_i(t)=A_{Nei}\exp(-\lambda_{Ne}t)[1+a\cos(\omega t+\varphi_i)]+A_{Fei}\exp(-\lambda_{Fe}t)+B_i. \quad (1)$$

Here $i$ specifies the telescope; $A_{Nei}$ is the number of electrons from $\mu \rightarrow e$ decay at $t=0$ in the Ne in the $i$th telescope; $\lambda_{Ne}$ is the muon consumption rate in Ne; $N_{Fei}$ and

TABLE I.

| Target | $a$ (%) | $\omega$ (rad/$\mu$s) | $\tau_\mu$($\mu$s) | $\chi^2/N$ | Statistics |
|---|---|---|---|---|---|
| 24Ne | 0.08(10) | 17.3 | 1.463(10) | 633/606 | $2.56\times 10^6$ Ne |
| | | | | | +11.5%Fe |
| Si | 2.93(68) | 16.8(41) | 0.792(30) | 549/530 | $96\times 10^3$ |
| 24Ne+16$H_2$ | 0.59(9) | 17.16(10) | 1.466(9) | 558/523 | $3.5\times 10^6$ Ne |
| | | | | | +7.7%Fe |
| Si | 3.68(69) | 17.24(33) | 0.769(20) | 525/522 | $96\times 10^3$ |
| 24Ne+1.8$H_2$ | 0.325(130) | 17.36(38) | 1.467(13) | 529/531 | $1.55\times 10^6$ Ne |
| | | | | | +9.9%Fe |
| Si* | 3.29(63) | 17.85(35) | 0.755(30) | 541/531 | $115\times 10^3$ |
| 24Ne+8$H_2$ | 0.33(14) | 16.97(38) | 1.463(13) | 523/531 | $1.62\times 10^6$ Ne |
| | | | | | +8.5%Fe |
| C | 4.32(15) | 17.01(2) | 2.015(5) | 574/594 | $960\times 10^3$ |

*Total over run on mixtures with +$H_2$ (1.8 atm) and +$H_2$ (8 atm).

$\lambda_{Fe}$ are the corresponding quantities for Fe; $B_i$ is the background; and $\varphi_i$ is the initial phase. The functional was minimized under the condition that the parameters $\lambda_{Ne}$, $\lambda_{Fe}$, $a$, and $\omega$ be the same for all the telescopes and under the condition that the phases $\varphi_i$ of the lateral telescopes be $\varphi_0 \pm 90°$, where $\varphi_0$ is the phase for the telescope in the muon beam.

The results of the analysis are shown in Table I. The asymmetry $a$ for pure neon was found from the frequency interval $17.0 \pm 0.5$ rad/$\mu$s. The parameters of the precession in silicon are results found from an analysis of spectra recorded by the method of detecting stopping events described above. They serve as a control of the parameters of the apparatus, including the magnetic field in the target. Also shown in Table I are data obtained with a carbon target, required for determining the polarization of the muons of the beam.

Figure 1 shows the precession amplitude $a$ and the value of $\chi^2$ versus the frequency $\omega$ as found from an analysis of the time-resolved $\mu$SR spectrum for the mixture Ne (24 atm) + $H_2$ (16 atm).

It follows from these results that a signal at the frequency of the free muon is observed in the neon–hydrogen mixture. This observation can serve as evidence that a diamagnetic HF molecule forms. With this result established, we undertook an effort to study the kinetics of the formation of this molecule. For this purpose we carried out two exposures of the target, with the mixtures Ne (24 atm) = $H_2$ (1.8 atm). The results of the analysis of these exposures are also listed in Table I.

The processes which occur in the Ne + $H_2$ mixture can be described schematically as the chain

$$\mu^- + \mathrm{Ne} \rightarrow {}_\mu\mathrm{F}^{n+} + [\mathrm{Ne} + \mathrm{H_2}] \rightarrow {}_\mu\mathrm{F}^+ + [\ldots] \rightarrow \mathrm{Ne}_\mu\mathrm{F}^+, {}_\mu\mathrm{F}^+, {}_\mu\mathrm{F}^0 + \mathrm{H}_\mu\mathrm{F}^+ + [\ldots] \rightarrow \mathrm{H}_\mu\mathrm{F}.$$

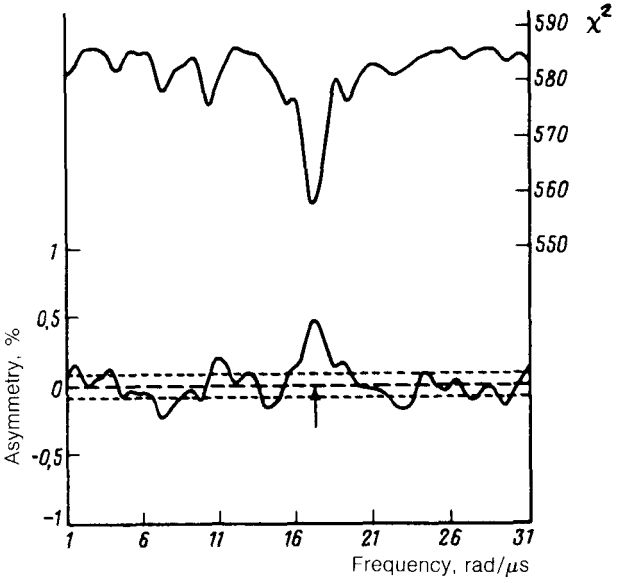


FIG. 1. $\chi^2$ (upper curve) and precession amplitude (lower curve) versus the precession frequency $\omega$ for a mixture of Ne (24 atm) + $H_2$ (16 atm). The dashed lines show the $\pm\ \sigma_a$ error corridor. Frequency, rad/$\mu$ s

Here $_\mu$F is the muonic atom (or ion) of fluorine. During observation of the frequency $\omega_\mu$ in a magnetic field, the paramagnetism of the intermediate states and the hyperfine interaction lead to the following behavior of the asymmetry coefficient $a$ as a function of the rate of the chemical reaction, $\lambda_H$, and the precession frequency of the paramagnetic state, $\omega_p$:[3]

$$a=A\frac{\lambda_{\rm H}}{(\lambda_{\rm H}^2+\omega_p^2)^{1/2}}. \qquad (2)$$

Here $A=a_0P_KD$, $a_0\approx 0.33$, and $P_K$ is the residual polarization of the muon in the $K$ shell. The quantity $D$ is determined by the depolarization due to hyperfine interactions in the $_\mu F^+$ ion ($D^+=0.447$),, in the $_\mu F^0$ atom ($D^0=0.417$),[10] and (possibly) in molecular ions. If the duration of the chemical interaction is smaller than or comparable to the time scales of these processes, $D$ also depends on $\lambda_{\rm H}$. Furthermore, there may be a contribution to this quantity from charge exchange during the formation of the electron cloud of $_\mu F^{n+}$ ion. However, according to estimates of Ref. 11, these processes would be of minor importance at a mixture pressure on the order of 20 atm.

Unfortunately, it is not possible to analyze in detail the time evolution of the formation of the HF molecule at the accuracy level of the measured asymmetry parameters at the $H_2$ pressures 1.8 and 8 atm. A fit of expression (2) to the experimental data with the substitution $\lambda_{\rm H}\to\lambda_1 X$, where $X$ is the $H_2$ pressure (in atmospheres), and $\lambda_1$ is the rate of the chemical reaction at $X=1$ atm $H_2$, yielded the

following estimates of the parameters A and $\omega_p/\lambda_1$: $A=0.54\pm0.09\%$ and $\omega_p/\lambda_1=2.8\pm2.0$. Taking $\omega_p\simeq100\omega_\mu$ to be the value for muonium, we find the rate of the chemical reaction to be $K\simeq(2.3\pm1.6)\times10^{-11}$ cm$^3$/s. The result agrees with the previous result $K=(2.5\pm0.8)\times10^{-11}$ cm$^3$/s (Ref. 4). Assuming that the residual polarizations $P_K$ in the $K$ shells of Ne and C are the same, and also assuming $a_C=a_0P_K$, we find an estimate of the depolarization factor: $D=A/a_C=0.13\pm0.02$. This value is smaller than expected, even if we assume that a neutral $_\mu F^0$ atom necessarily forms in an intermediate state in the cascade of reactions $_\mu F^+ + Ne + H_2 \rightarrow ... \rightarrow H_\mu F$. The lifetimes of both $_\mu F^+$ and $_\mu F^0$ are longer than the corresponding hyperfine-interaction times. Unfortunately, it does not seem productive to pursue the discussion in more detail because of the statistical errors involved here. Analysis of the experimental results which were obtained shows that the parameters $a$ and $\lambda_1$ are very sensitive to the magnitude of the asymmetry coefficients and the accuracy with which they are measured at low hydrogen concentrations, e.g., at 1.8 atm $H_2$. These problems can be overcome by building up a sufficient statistical base.

The fundamental conclusion which follows from the results of this study is that it has been demonstrated experimentally that it is possible to use the $\mu^-$SR method to study fast interactions of muonic atoms with a medium. Such studies have previously been carried out exclusively with positive muons.

We wish to thank V. P. Dzhelepov and V. G. Zinov for support of and interest in this study. We also thank V. S. Roganov and A. N. Rudenko for assistance in the measurements.

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Translated by D. Parsons