Generation of Very Slow Polarized Positive Muons

E. Morenzoni,[1] F. Kottmann,[4] D. Maden,[1] B. Matthias,[2] M. Meyberg,[1]
Th. Prokscha,[2] Th. Wutzke,[1] and U. Zimmermann[3]
[1]*Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland*
[2]*Physics Institute, University of Heidelberg Philosophenweg 12, D-69120 Heidelberg, Germany*
[3]*Institute for Intermediate Energy Physics, Eidgenössische Technische Hochschule Zürich, CH-5232 Villigen PSI, Switzerland*
[4]*Institute for High Energy Physics, Eidgenössische Technische Hochschule Zürich, CH-5232 Villigen PSI, Switzerland*
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We have measured the polarization of very slow positive muons (kinetic energy ~10 eV) obtained from the moderation of a 100% polarized surface muon beam in rare gas solid layers. We find that the muons nearly retain their initial polarization, indicating that the moderation process is very fast compared to depolarizing mechanisms. This result opens the possibility of producing beams of very low energy polarized muons, which have a wide range of possible applications, e.g., as a magnetic microprobe of surfaces and thin films.

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Polarized positive muons (μ^+) and muonium atoms (μ^+e^-) have proven to be ideal microscopic probes of magnetic properties of matter as well as isotopic probes of proton and hydrogen diffusion mechanisms and chemical reactions in solids, liquids, and gases [1,2]. Such information is obtained by measuring the time evolution of the μ^+ spin polarization once they have thermalized in the sample [muon spin rotation technique (μSR) [3]]. For these studies the so-called surface μ^+ ($E \simeq 4.1$ MeV) originating from the two body decay of positive pions at rest are used [4]. These μ^+ are 100% longitudinally polarized with spin opposite to the direction of motion. The μSR technique is presently limited to the study of bulk characteristics of matter since the range and range straggling of surface μ^+ are approximately 150 mg/cm^2 and several tens of mg/cm^2, respectively. The availability of a beam of polarized very slow positive muons in the energy range of about 10 eV to a few tens of keV would extend the present range of application of μSR techniques to surface and thin film physics, thus offering a completely new spectroscopic method for these fields.

It has recently been shown that, if energetic μ^+ are injected into a thin moderator, very slow μ^+ are emitted from its downstream side [5,6]. The energy distribution of these μ^+ shows a maximum near 10 eV with a tail extending to higher energies. So far, the most efficient moderator appears to be a solid layer of Ar deposited on a metal substrate, giving an efficiency of 10^{-5} for converting an incoming surface μ^+ into a very slow one [6].

In this Letter we report a first measurement of the polarization of very slow μ^+ emitted from solid Ar and Kr. It is observed that the μ^+ retain most of the polarization of the incident high energy beam. This experimental finding demonstrates that a very low energy beam of polarized μ^+ for surface and thin film μSR can be realized by making use of the moderation technique.

This experiment was performed at the $\pi E1$ beam line of the Paul Scherrer Institute (PSI). The channel was tuned to deliver surface μ^+ at a momentum (p) of 27.3 MeV/c ($\Delta p/p = 3\%$). Positron background in the beam was suppressed by a static $\mathbf{E} \times \mathbf{B}$ separator adjusted to transmit the μ^+. Lead and copper collimators with an aperture 20 mm wide and 12 mm high defined the μ^+ beam spot on a plastic scintillator of 200 μm thickness which detected the incident μ^+ and showed a rate of $\sim 7\times10^5\ \mathrm{s}^{-1}$. After passing through a 50 μm stainless-steel vacuum window, the μ^+ entered the ultrahigh vacuum (UHV) chamber shown in Fig. 1.

Once in the UHV chamber, they traversed two 30 μm Al cold shields before impinging on a cold target which acted as a moderator. This moderator consists of an Al substrate (250 μm thick, 99.999% pure, and cooled by a

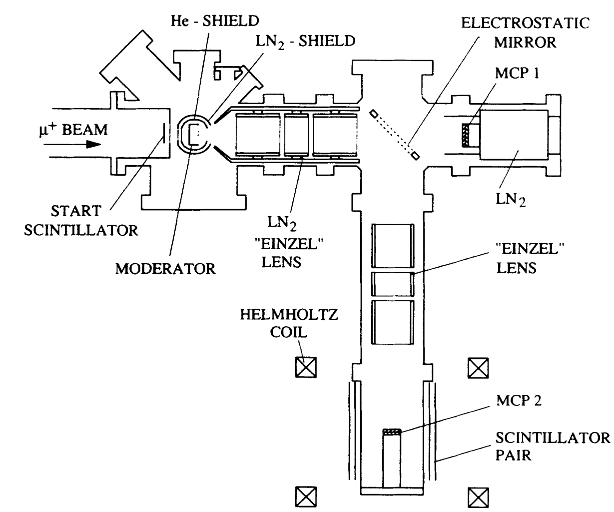

FIG. 1. UHV apparatus with cryostat used for the moderation studies. For details see text.

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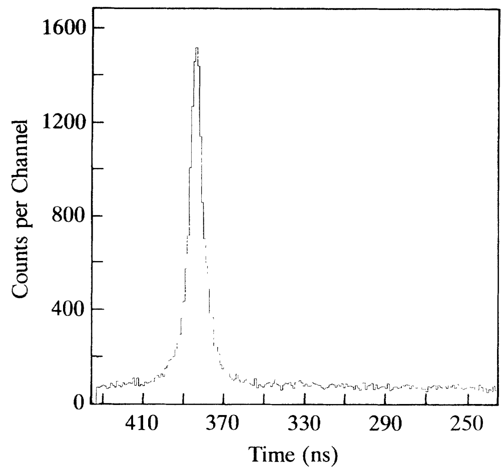

FIG. 2. Time-of-flight distribution between start scintillator and MCP 2. The peak is given by the very slow μ^+ emitted from the Ar moderator.

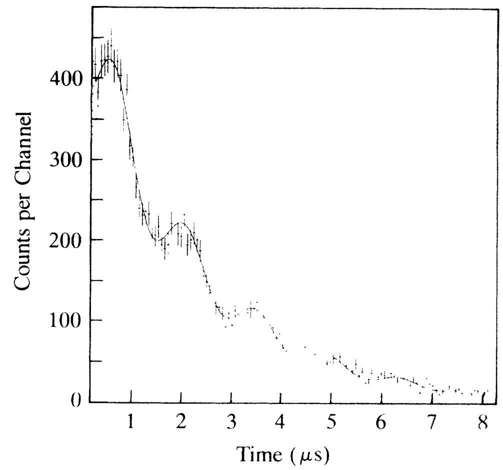

FIG. 3. Decay time distribution (left scintillator telescope) of the very slow μ^+ emitted from a solid Ar layer and precessing in a 50 G transverse magnetic field.

helium cryostat) with a solid layer of Ar or Kr deposited on it. The solid gas layer (thickness between 200 and 300 nm) is obtained by condensing onto the Al substrate research grade gas at a partial pressure of typically 10^{-5} mbar for 120 s. The substrate is held at a temperature of 20 K during deposition and subsequent measurements. An oscillating quartz crystal microbalance mounted just above the moderator target is also coated by a solid gas layer during deposition and is used as a monitor of the film thickness. The chamber base vacuum is typically 10^{-9} mbar to minimize surface contamination from the residual gas. The total thickness traversed by the μ^+ and the incident beam momentum are chosen such that the stopping density distribution of the μ^+ is centered at the downstream surface of the target, i.e., in the solid rare gas layer. The moderation efficiency in our experiment is between 10^{-4} and 10^{-5} depending on the moderator and the preparation parameters of the layer (deposition rate, substrate temperature, thickness, and age) [7]. The best value is obtained with solid Ar.

The Al substrate is electrically insulated and held at a positive voltage of 8 kV to extract the very slow muons from the production region. They are accelerated in a two stage system formed by the substrate and two wire grids. The extracted particles are focused onto a microchannel plate detector (MCP 2 in Fig. 1) with two electrostatic "einzel" lenses and an electrostatic mirror. Muons which are not completely moderated are not deflected appreciably by the electrostatic mirror and are detected by MCP 1, which is used for monitoring and beam tuning purposes.

We observe the polarization of the very slow μ^+ stopped in MCP 2 by using the standard transverse magnetic field μSR technique which relies on the correlation of the μ^+ spin direction with the direction of emission of the positrons from the μ^+ decay. While the very slow μ^+ arriving at MCP 2 produce pulses in this detector, the subsequent decay e^+ are observed by four plastic scintillator pairs surrounding MCP 2 providing a solid angle coverage of 60% of 4π sr. A pair of Helmholtz coils applies a magnetic field B of up to 100 G perpendicular to the expected spin direction of the polarized μ^+. To acquire the characteristic μ^+ spin precession signal, we measure the time between a pulse in MCP 2 and a signal in one of the four e^+ telescopes. A readout of the data acquisition system is triggered by a triple coincidence of MCP 2 preceded within 800 ns by a pulse in the start scintillator (S1) and followed within 10 μs by a pulse in one of the e^+ telescopes. From these events we derive histograms of the μ^+ time-of-flight distribution between S1 and MCP 2 and of the four decay time spectra started by MCP 2 and stopped by one of the e^+ detectors.

Figure 2 shows the μ^+ time-of-flight (TOF) spectrum between S1 and MCP 2. The peak corresponds to μ^+ emitted from the moderator with an initial energy of ~10 eV and with an energy spread of the same order of magnitude [7]. For the polarization measurement we select from the TOF spectrum only those events contained in the peak and we subtract the contribution to the μ^+ precession spectra produced by random coincidences between S1 and MCP 2. A resulting μSR spectrum showing the μ^+ spin precession signal in a transverse magnetic field of 50 G is plotted in Fig. 3. For each of the four scintillator telescopes,

$$N(t) = N_0^i e^{-t/\tau_\mu}[1 + A_\mu e^{-\lambda t}\cos(\omega_\mu t + \Phi_\mu^i)] + C^i. \tag{1}$$

In this equation the free muon precession frequency $\omega_\mu = 2\pi \times 13.55$ kHz/G, the μ^+ spin relaxation rate λ, and the decay asymmetry A_μ are fit parameters common to the four precession spectra, whereas the precession phases Φ_μ^i, the normalizations N_0^i, and the constant backgrounds C^i vary separately for each telescope (i =left, top, right, bottom). The muon lifetime is denoted by τ_μ. By fitting simultaneously Eq. (1) to the four preces-

TABLE I. Results of the polarization measurements.

Moderator	Asymmetry (%)	Polarization P(%)[c]	Polarization P'(%)[d]
Ar[a]	18.24 ± 0.56	87.0 ± 2.8	91.7 ± 7.5
Kr[a]	16.81 ± 4.30	80 ± 20	85 ± 23
Al[b]	19.89 ± 1.50	94.8 ± 7.2	100

[a] Very slow muons ($E \sim 10$ eV).
[b] Muons with keV energy.
[c] Normalized to asymmetry of surface muons in MCP.
[d] Normalized to asymmetry of keV muons emitted from the Al moderator.

sion spectra, we obtain the asymmetries $A_{\rm MCP}^{{\rm slow}\ \mu}$ given in Table I. Preliminary results give a comparable yield and asymmetry for very slow μ^+ produced by a solid layer of N_2. No relaxation of the slow μ^+ spin is observed in any of the moderators. The decay asymmetry is a direct measure of the polarization if the maximum observable asymmetry with our μSR apparatus and the behavior of a polarized μ^+ stopped in the microchannel plate detector are taken into account.

The μSR spectrometer (bottom part of Fig. 1) was calibrated by attaching it directly to the surface μ^+ beam line after the start scintillator and measuring the asymmetry signal produced by surface muons stopped in the MCP in a transverse magnetic field. For this measurement we started the time differential μSR measurement with the signal from the start scintillator. Unique correspondence between an incident muon and a decay positron was ensured by reducing the beam intensity to $\sim 2\times10^4$ μ^+/s and by recording only those events originating from a μ^+ which was neither preceded nor followed by another μ^+ within ± 10 μs. With this arrangement we performed two measurements. The maximum observable asymmetry, including the effects of the counter geometry, finite solid angle, target size, and absorption of low momentum e^+, was determined by measuring the asymmetry signal in a very pure Al target where no depolarization occurs [8]. From fits to the precession spectra at $B = 25$, 50, and 100 G we obtain an asymmetry of $A_{\rm Al}^{{\rm surface}\ \mu} = (28.22 \pm 0.15)\%$. From a Monte Carlo simulation of the μSR spectrometer and taking into account that the $\mathbf{E}\times\mathbf{B}$ field in the separator rotates the spin by 4° we expect that $A_{\rm Al}^{{\rm surface}\ \mu} = (28.06 \pm 0.17)\%$ for 100% beam polarization. The behavior of polarized μ^+ in the MCP [9] was then investigated by measuring the asymmetry signal with MCP 2 as the target. In this case we obtain $A_{\rm MCP}^{{\rm surface}\ \mu} = (20.97 \pm 0.18)\%$. A Fourier analysis of the asymmetry spectra shows that the missing asymmetry observed with the MCP target is due to muonium formation. The MCP consists mainly of a glass [10] and it is known that a significant fraction of μ^+ stopped in oxide insulators form muonium [8].

The values for the absolute (transverse) polarization P of the ~ 10 eV μ^+, given in Table I, are obtained from the expression $P = 100\% \times (A_{\rm MCP}^{{\rm slow}\ \mu}/A_{\rm MCP}^{{\rm surface}\ \mu})$.

The results show that the very slow μ^+ retain practically the full initial polarization. The fits to the Ar data give the phases $\Phi_\mu^L = (212\pm4)°$, $\Phi_\mu^T = (119\pm5)°$, $\Phi_\mu^R = (29.5\pm4)°$, and $\Phi_\mu^B = (297\pm4)°$. This corresponds to an initial rotation within the precession plane of Φ_μ^R and is in good agreement with the result of a Monte Carlo simulation of the very slow μ^+ transport from production target to detector including the action of the magnetic field on the μ^+ spin in flight. Rotation out of this plane, which would lead to a reduced transverse polarization, is negligible.

We also determined the polarization of the slow muons which are emitted from the blank Al substrate. Instead of a narrow peak as in Fig. 2, in this case, the spectrum of the TOF between S1 and MCP 2 shows a less pronounced, broader peak. About a factor of 60 fewer very slow muons with $E \sim 10$ eV are emitted from the pure Al substrate than from the frozen gas layer [7]. The broad distribution corresponds to muons of several keV energy which are emitted from the Al substrate acting as a degrader and which are accepted by the electrostatic transport system. The asymmetry observed for these muons ($A_{\rm MCP}^{{\rm keV}\ \mu}$) is given in Table I. Since even thermalized μ^+ in Al do not depolarize, one can expect that these keV μ^+ are also 100% polarized. We can therefore use the value of $A_{\rm MCP}^{{\rm keV}\ \mu}$ for an alternative determination of the polarization P' of the very slow μ^+ emitted from the rare gas layers according to the expression $P' = 100\% \times (A_{\rm MCP}^{{\rm slow}\ \mu}/A_{\rm MCP}^{{\rm keV}\ \mu})$. The results are summarized in Table I and are in agreement with the results for P.

Based on the analogy with e^+ emission from solid moderators, diffusion controlled [5] and hot μ^+ emission mechanisms have been proposed [6,11,12] to explain the moderation of μ^+ down a kinetic energy of a few eV, but the data available to date do not provide an unambiguous signature to distinguish between these or other emission mechanisms. According to the recombination-assisted diffusion mechanism of Ref. [5], the very slow μ^+ are produced from the breakup near the moderator surface of muonium which is formed inside the solid. Muonium formed inside the solid can acquire energy by catalyzing a first electron-hole recombination. A second recombination between the bound muonium electron and another hole near the surface causes the muonium to break up and a μ^+ with several eV of kinetic energy to be reemitted.

The hot μ^+ emission mechanism suggests that the observed very slow μ^+ are muons which have not thermalized (epithermal μ^+). Initially the surface μ^+ rapidly lose energy in matter by Coulomb collisions with electrons, ionizing and exciting the target atoms (electron-hole pair and exciton creation). When a μ^+ has lost most of its energy, at energies below $\sim$ 10 keV, charge changing cycles involving muonium formation and breakup also become important as an energy dissipating mechanism [13]. In wide band gap insulators such as solid Kr, Ar, and N_2, these processes have high threshold energies (band gap energy 11, 14, and 14 eV, respectively [14]). Once the μ^+ has reached a kinetic energy of the order of these threshold levels, the corresponding efficient energy loss mechanisms are strongly suppressed or even energetically impossible and the energy loss rate becomes low since the dominant remaining energy loss mechanism is via relatively inefficient phonon emission. Regarding phonon emission, van der Waals solids such as Kr, Ar, and N_2 are also particularly favorable to a low energy loss rate since they have soft phonon spectra with energies of the order of 5–10 meV [14]. The particularly efficient moderation to eV energies of μ^+ especially in Ar is therefore a consequence of an open energy window in the moderation process which significantly increases the escape depth of the $\sim$ 10 eV muons.

The observation that the very slow μ^+ retain almost their full initial polarization implies an upper limit to the time scale of the mechanism responsible for the moderation and is consistent with the hot muon emission mechanism. Diffusionlike processes, which assume thermalization at the onset, or delayed processes, where the μ^+ originates from a muonium atom living for a time scale characteristic of the period (t_{HF} =224 ps) of electron-muon spin exchange oscillation due to the hyperfine interaction, can be eliminated as a source of the majority of the observed very slow muons. In solid Ar and Kr, thermalization leads to practically 100% muonium formation [15]. Muonium survival for times non-negligible compared to t_{HF} would reduce the polarization of the very slow μ^+ by a factor of 2, at variance with our results. In solid N_2 also, thermalized μ^+ quickly depolarize and only retain one-third of the initial polarization [16]. On the other hand, epithermal μ^+ emission conserves the initial surface μ^+ polarization since depolarization via electron scattering and Coulomb scattering is negligible and the overall time for slowing down to 10 eV is very short ($\sim$ 10 ps). As a result even transient muonium formation does not give rise to a sizable depolarization. Our measurements also show that other depolarizing processes such as spin exchange interactions with free electrons or ions, which are produced in the wake of the slowing down μ^+, have no appreciable effect.

The discovery that the very slow μ^+ are polarized is an essential step towards their use in a number of new applications [17]. A solid Ar moderator in conjunction with a surface μ^+ beam can be used as a source of a tunable beam of polarized μ^+ with energies between $\sim$ 10 eV and a few tens of keV. Solid Ar, with conversion efficiencies up to 10^{-4} [7], combined with the new surface μ^+ beam line $\pi E5$ at PSI, which is designed to deliver more than 10^8 μ^+/s [18], can provide a source flux of about 10^4 very slow polarized μ^+ per s.

By varying the energy of the μ^+ between $\sim$ 10 eV and a few tens of keV, implantation depths between typically fractions of a nm and a few hundred nm can be achieved. The corresponding estimated range straggling varies between 80% and 20%. Thus thin films, vacuum-solid interfaces, and solid-solid interfaces would become accessible to μSR studies, which have provided valuable information about magnetism, defects, and diffusion in bulk matter, and the behavior of implanted hydrogen isotopes. The field of possible applications of very slow μ^+ is not restricted to solid state physics. The μ^+ beam can also be converted into a clean muonium beam by passing the μ^+ through a thin foil or a gas target which neutralizes it. Such a source of muonium may be expected to contain an admixture of the metastable $2s$ state and may be used for new spectroscopic studies of the $n = 2$ states, free from background due to the incident μ^+ beam.

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