Geant4 simulation of the PSI LEM beam line: energy loss and muonium formation in thin foils and the impact of unmoderated muons on the $\mu$SR spectrometer

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ABSTRACT: The PSI low-energy $\mu$SR spectrometer is an instrument dedicated to muon spin rotation and relaxation measurements. Knowledge of the muon beam parameters such as spatial, kinetic energy and arrival-time distributions at the sample position are important ingredients to analyze the $\mu$SR spectra. We present here the measured energy losses in the thin carbon foil of the muon start detector deduced from time-of-flight measurements. Muonium formation in the thin carbon foil (10 nm thickness) of the muon start detector also affect the measurable decay asymmetry and therefore need to be accounted for. Muonium formation and energy losses in the start detector, whose relevance increase with decreasing muon implantation energy ($<10$ keV), have been implemented in Geant4 Monte Carlo simulation to reproduce the measured time-of-flight spectra. Simulated and measured time-of-flight and beam spot agrees only if a small fraction of so called “unmoderated” muons which contaminate the mono-energetic muon beam of the $\mu$SR spectrometer is introduced. Moreover the sensitivity of the beam size and related upstream-downstream asymmetry for a specially shaped “nose” sample plate has been studied for various beam line settings, which is of relevance for the study of thermal muonium emission into vacuum from mesoporous silica at cryogenic temperatures.

KEYWORDS: Muon spectrometers; Instrumentation for particle accelerators and storage rings - low energy (linear accelerators, cyclotrons, electrostatic accelerators); Targets (spallation source targets, radioisotope production, neutrino and muon sources); Instrumentation and methods for time-of-flight (TOF) spectroscopy

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1 Introduction

Polarized positive muons $\mu^+$ can be used to investigate structural properties and dynamical processes of solid states via so called $\mu$SR technique [1] which stands for a collection of methods as Muon Spin Rotation, Relaxation and Resonance. A low-energy $\mu^+$ beam with tunable energy in the keV regime [2] plays an important role in the $\mu$SR field because these muons can be used to investigate thin films. The Low Energy Muon (LEM) beam line at PSI shown in figure 1 is delivering $\mu^+$ with tunable kinetic energy between 0.5 and 30 keV allowing the study of thin films and multilayers as a function of the implantation depth ranging from 0.5 nm to a few hundred nm [3, 4]. The low-energy $\mu^+$ are obtained by moderating a surface $\mu^+$ beam (4 MeV energy) from the $\mu$E4 beam line [5] with a 125 $\mu$m thick Ag foil coated with a 200-300 nm thick layer of solid Ar-N$_2$ [6]. The moderated $\mu^+$ leaving the solid Ar-N$_2$ surface have mean kinetic energy of 15 eV. As the moderator is placed at high voltage (typically $V_{\text{mod}} = 15$ kV), after leaving the moderator the $\mu^+$ are accelerated to about 15 keV kinetic energy. Using various electrostatic elements, the $\mu^+$ are transported from the moderator to the sample region. Neglecting the various focusing elements (einzel lenses and conical lenses), the $\mu^+$ are first bent by an electrostatic mirror $45^\circ$ to the muon beam axis and then transported through a spin rotator. Subsequently, they cross a thin carbon foil (C-foil) which acts as a start detector before being implanted into the sample. The nominal density and thickness of the C-foil are $\sim 2 \mu$g/cm$^2$ and 10 nm, respectively.

While crossing the C-foil the $\mu^+$ is ejecting several electrons (on average 3) of few eV energy from the foils surface whose detection define the $\mu^+$ implantation time in the sample and the start of the event in the data acquisition. The initially mono-energetic $\mu^+$ arriving at the start detector undergoes energy and angular straggling which degrade the beam quality and affect the measured $\mu$SR time spectra. Moreover, by traversing the thin C-foil, a fraction of the $\mu^+$ can undergo charge-exchange and leave the foil as muonium (Mu) or negatively charged muonium (Mu$^-$) which decrease the measurable total muon decay asymmetry.

Another complication is represented by the high-energy tail of the $\mu^+$ leaving the Ar-N$_2$ moderator not as epithermal $\mu^+$ at the eV energy but as only partially moderated muons whose energy
Figure 1. Schematic of the new LEM beam line [7]. The 4 MeV surface \( \mu^+ \) is moderated at an efficiency of about 0.01\% to an energy of about 15 eV before being re-accelerated again to energies up to 20 keV. The \( \mu^+ \) beam is then bent by a 45° electrostatic mirror before going through the spin rotator and the start detector and arriving at the sample plate mounted on the cold finger of the cryostat.

is sufficiently low to be deflected by the 45° electrostatic mirror and transported to the sample region. We term these muons as “unmoderated” muons.

All these processes, at the moderator and at the C-foil, affect the kinetic energy distribution of the \( \mu^+ \) leaving the C-foil and consequently the \( \mu^+ \) arrival time distribution at the sample position (relative to the signal in the start detector). This arrival time distribution needs to be known to understand the detailed shape of the decay positron time spectra at early times.

In this paper, measurements of the energy loss in the thin C-foil at various \( \mu^+ \) energies which have been done using a time-of-flight (TOF) technique are presented. Energy losses and Mu formation in the 10 nm thick C-foil of the start detector as well as “unmoderated” fraction of \( \mu^+ \) have been implemented in the musrSim [8] simulation package which is based on Geant4 [9] to match the measured TOF spectra. The Geant4 simulation has been performed starting from the downstream of the moderator till the sample region.
The LEM beam line was upgraded in 2012 to allow for longitudinal μSR measurements, to have a better suppression of the proton/ion background from the moderator and to have a better time resolution. The first two items have been achieved by the installation of a spin rotator [10] as shown in figure 1 after the electrostatic mirror, whereas the improved time resolution has been achieved by reducing the distance between the start detector and the sample region (from 1164 mm to 563 mm distance, cf. figure 1 of [3]).

The beam sizes at the sample position for variations of the beam line settings have been studied and compared with determinations obtained from the upstream-downstream asymmetry measurements.

2 Energy losses in the thin C-foil

To determine the energy loss in the C-foil of the start detector a TOF technique was applied. A muon beam with well defined kinetic energy given by the moderator high voltage (HV) is focused into the C-foil of the start detector which is set to a negative HV ($V_C = -3.38 \text{ kV}$). The secondary electrons knocked out in the downstream direction by the muon crossing the foil are first accelerated and then deflected by a system of grids towards a micro-channel-plate (MCP3) as shown in the inset of figure 1. The MCP3 signal provides the “start” signal of the TOF measurement. Another micro-channel-plate (MCP2) is placed at the position usually taken by the sample holder while performing the μSR measurements. A signal from the MCP2 delivers the stop time of the TOF measurement. In addition, the detection of an $e^+$ from muon decay in the positron counters of the μSR spectrometer is required. The measured time spectra obtained in this way are shown in figure 2. Understanding these time spectra requires some knowledge of the processes occurring in the C-foil. The muons can leave the foil at various charge states: $\mu^+$, Mu and $\mu^-$, as depicted in figure 3(a). The equilibrium yield for these various charge states scaled from proton data [11] are shown in figure 3(b). As the model of H formation (via overlap of the atomic states and solid electron states, and electron tunneling given in [11]) when a low-energy proton beam crosses a thin C-foil depends only on the velocity, velocity scaling of the proton data has been assumed to calculate the Mu charge state yield.

The first peak at early times ($\gamma$ peak) is ensued in the following way: the electrons knocked out by a muon crossing the C-foil are transported and detected in the MCP3 delivering the start time of the event. The avalanche process occurring in the channel walls of the MCP3 generates about $10^6$ electrons impinging with energies of a few hundred eV on the anode of MCP3, where they generate UV photons by ionization/recombination processes or Bremsstrahlung. Some of these photons could be detected (non-zero solid-angle acceptance and non-zero detection efficiency) in the MCP2 providing the stop time of the event. The width of the $\gamma$ peak is about 0.7 ns and it corresponds to the timing resolution of the TOF system (MCPs and electronics). It was demonstrated that the position of this peak is not affected by HV variations at the RA conical lens or at the L3 einzel lens or in the start detector grids confirming that it is related with photon emission in the MCP3.

The second peak is caused by Mu atoms traveling from the C-foil to the MCP2 while the third (largest) peak is caused by $\mu^+$. Also for these peaks the start signal of the events is given by the electrons emitted from the C-foil. Since the Mu motion is not affected by the electric fields in the start detector, the Mu peak position (relative to the $\gamma$ peak) can be related in a simple way to the
Figure 2. TOF spectrum from the start detector to a micro-channel plate (MCP2) detector placed at the sample plate position for 12 keV $\mu^+$ beam transport energy. The peaks are corresponding to the prompt photons, the C-foil Mu atoms and the $\mu^+$, respectively. The time axis is set with an arbitrary zero. The HV at the RA and L3 lenses are switched off.

Figure 3. (a) Formation of Mu and $\mu^-$ in the carbon foil due to the charge exchange process. (b) The charge state yields of the $\mu^+$ exiting the carbon foil as a function of incoming muon energy at the foil surface, according to a velocity scaling of the proton data parameterizations [11].
kinetic energy of the Mu after the C-foil. The delayed timing of the $\mu^+$ peak compared to the Mu peak is mainly due to the fact that $\mu^+$ has to overcome the negative electrostatic potential in the start detector. Thus, $\mu^+$ leave the start detector at a smaller kinetic energy compared to Mu atoms producing the observed delay.

The time $t_0$ at which a $\mu^+$ is crossing the C-foil is given by:

$$t_0 = t_\gamma - \Delta t_{FE} - \Delta t_c,$$

where $t_\gamma$ is the position of the $\gamma$ peak, $\Delta t_{FE}$ is the TOF of the knocked out electrons from the C-foil to MCP3 and $\Delta t_c = 1.67$ ns is the TOF of a particle with the speed of light from the MCP3 to the MCP2. A $\Delta t_{FE} = 13.5(5)$ ns has been determined from the time difference of two prompt peaks in the $\mu^+$ decay time spectra in regular $\mu$SR measurements. The earlier peak is caused by positrons hitting the C-foil and producing foil electrons successively detected in the $e^+$ counters surrounding the sample region and the later peak is caused by positrons hitting the MCP3 directly and then detected in the $e^+$ counters. The TOF of foil electrons to the MCP3 is independent of the $e^-$ emission position at the C-foil but has a small dependence on the emission angle. However, this effect and the possible variation of the $e^-$ emission energy are included in the uncertainty of $\Delta t_{FE}$. The TOF of $\mu^+$ through TD was simulated for various initial position on the C-foil. The resulting TOF spread is about 0.1 ns. Since the motion of the Mu atom, being a neutral system, is not affected by the electric field in the start detector or other electric fields downstream of the start detector, the Mu atom will move from the C-foil to the MCP2 with uniform velocity ($v_{Mu}$). Its TOF from the C-foil to the MCP2 ($\Delta t_{Mu}^{meas}$) determined from the TOF spectra is given by

$$\Delta t_{Mu}^{meas} = t_{Mu} - t_0,$$

where $t_{Mu}$ is the position (most probable value) of the Mu peak. Therefore, the Mu kinetic energy right after crossing the C-foil is given by

$$E_{CFoil}^{Mu} = \frac{m_{Mu} v_{Mu}^2}{2} = \frac{m_{Mu}}{2} \left( \frac{L}{\Delta t_{Mu}^{meas}} \right)^2,$$

where $m_{Mu}$ is the Mu mass and $L$ the distance between the C-foil and MCP2. It can be assumed that both the $\mu^+$ and the Mu leaving the C-foil have the same kinetic energy $E_{CF} = E_{CFoil}^{Mu}$. With this assumption, the $\mu^+$ energy loss in the C-foil (independent on the muon charge state when leaving the foil) is simply given by

$$E_{loss} = eV_{mod} - eV_C - E_{CF},$$

where $V_{mod}$ is the HV at the moderator and $V_C$ the HV at the C-foil.

The energy loss in the $\sim 2 \mu g/cm^2$ C-foil for various $\mu^+$ energies are summarized in table 1 and are plotted in figure 4. From the energy loss, a stopping power of $S = 0.52(6)$ keVcm$^2$/μg for a muon energy of 12 keV is obtained which has to be compared with the value of 0.70(1) keVcm$^2$/μg in [12]. The uncertainty of the extracted stopping power and its deviation from the value in [12] is related to the uncertainty of the C-foil area density (0.2 μg/cm$^2$) originating from the non-uniformity of the foil and the uncertainty in the thickness from production to production.

To check for the correctness of the assumed distance $L = 563$ mm used in eq. (2.3) we calculate the TOF of the $\mu^+$ from the C-foil to the MCP2 ($\Delta t_{\mu^+}^{calc}$) using simple kinematic calculations and
compare it with the TOF of $\mu^+$ peak determined directly from the measured TOF spectra ($\Delta t_{\mu^+}^{\text{meas}}$), e.g. figure 2. In order to calculate the $\mu^+$ TOF from the C-foil to the MCP2, we need to consider the various regions defined by the distances $d_i$ as shown in figure 5. Since the TOF measurements have been accomplished without any HV at the RA conical lens, L3 einzel lens and no electric fields in the sample region the $\mu^+$ traveling from the C-foil till the MCP2 experience only regions of constant electric potential and two small regions in the start detector of constant electric field. The non-relativistic equations of motions for the various regions $d_i$ are given by:

\begin{align}
    d_1 &= \frac{1}{2}a_1t_1^2 + v_1t_1, \quad v_1 = \sqrt{\frac{2E_1}{m}}, \quad \Delta V_1 = (V_C - V_F), \quad a_1 = \frac{q}{m} \frac{\Delta V_1}{d_1}, \\
    d_2 &= v_2t_2, \quad v_2 = \sqrt{\frac{2E_2}{m}}, \quad E_2 = E_1 + \Delta V_1, \quad a_2 = 0, \\
    d_3 &= \frac{1}{2}a_3t_3^2 + v_3t_3, \quad v_3 = v_2, \quad \Delta V_3 = (V_F - V_B), \quad a_3 = \frac{q}{m} \frac{\Delta V_3}{d_3}, \\
    d_4 &= v_4t_4, \quad v_4 = \sqrt{\frac{2E_4}{m}}, \quad E_4 = E_2 + \Delta V_3, \quad a_4 = 0, \\
    d_5 &= \frac{1}{2}a_5t_5^2 + v_5t_5, \quad v_5 = v_4, \quad \Delta V_5 = (V_B - 0), \quad a_5 = \frac{q}{m} \frac{\Delta V_5}{d_5}, \\
    d_6 &= v_6t_6, \quad v_6 = \sqrt{\frac{2E_6}{m}}, \quad E_6 = E_4 + \Delta V_5, \quad a_6 = 0
\end{align}

where $m$ is the $\mu^+$ mass, $E_i$ the $\mu^+$ kinetic energy at the entrance of the $i$-region ($E_1 = E_{CF}$), $q$ the charge of the particle, and $V_C = -3.38$ kV, $V_F = -3.19$ kV and $V_B = -3.56$ kV the various HVs applied at the grids of the start detector. The TOF $t_i$ in these various $i$-regions can be calculated.

**Figure 4.** Energy loss in the 10 nm C-foil determined from the Mu peak in the TOF spectra for various incoming muon energy at the foil surface.
Figure 5. (a) Schematic view of the TOF measurement. (b) Schematic of the start detector with various grids to deflect the electrons and defined regions of constant potential or constant electric field. Distances \(d_i\) \((i = 1 - 6)\) are defined for \(\mu^+\) on the beam axis.

Using these simple relations:

\[
t_i = \frac{d_i}{v_i} \quad \text{for} \quad a_i = 0 \tag{2.11}
\]

\[
t_i = -\frac{v_i}{a_i} + \text{sign}(a_i) \sqrt{\left(\frac{v_i}{a_i}\right)^2 + \frac{2d_i}{a_i}} \tag{2.12}
\]

Where \(v_i\) is the \(\mu^+\) velocity when entering the region \(d_i\), and \(a_i\) the \(\mu^+\) acceleration in the region \(d_i\) of constant electric field. The total TOF \(\Delta t_{\mu^+}^{\text{calc}}\) is eventually given by the sum

\[
\Delta t_{\mu^+}^{\text{calc}} = \sum_{i=1}^{6} t_i . \tag{2.13}
\]
Table 1. Moderator potential $V_{\text{mod}}$, incoming muon energy at the C-foil surface $E_{\mu^+}$, measured and calculated TOF (for the peak maximum) of $\mu^+$ and Mu from the start detector to MCP2. The energy of the particle after the carbon foil $E_{\text{CF}}$ is determined from $\Delta t_{\text{meas} \mu}$. Knowing the $E_{\text{CF}}$, the energy loss $E_{\text{loss}}$ in the carbon foil can be calculated using eq. (2.4). The $\Delta t_{\text{calc} \mu^+}$ assumes the distances $d_1$ to $d_6$ given by mechanical construction and also assumes mono-energetic $\mu^+$ hitting the C-foil with energy given by the moderator and C-foil electric potentials, $E_{\mu^+} = eV_{\text{mod}} + 3.38$ keV.

| $V_{\text{mod}}$ (kV) | 8.5 | 10.0 | 12.0 | 13.5 | 15.0 | 16.5 | 18.0 |
|-----------------------|-----|------|------|------|------|------|------|
| $E_{\mu^+}$ (keV)     | 11.88 | 13.38 | 15.38 | 16.88 | 18.38 | 19.88 | 21.38 |
| $E_{\text{CF}}$ (keV) | 10.84(6) | 12.37(7) | 14.49(9) | 16.11(10) | 17.67(12) | 19.17(14) | 20.78(15) |
| $E_{\text{loss}}$ (keV) | 1.04(6) | 1.01(7) | 0.89(9) | 0.77(10) | 0.71(12) | 0.71(14) | 0.60(15) |
| $\Delta t_{\text{meas} \mu}$ (ns) | 131.4(5) | 123.0(5) | 113.7(5) | 107.8(5) | 102.9(5) | 98.8(5) | 94.9(5) |
| $\Delta t_{\text{meas} \mu^+}$ (ns) | 152.7(5) | 139.8(5) | 126.4(5) | 118.6(5) | 112.1(5) | 106.8(5) | 101.6(5) |
| $\Delta t_{\text{calc} \mu^+}$ (ns) | 152.9(3) | 140.0(3) | 126.5(3) | 118.4(3) | 112.0(3) | 106.7(3) | 101.8(3) |

As can be seen by comparing the 5-th with the 6-th rows of table 1, there is a very good agreement between $\Delta t_{\text{calc} \mu^+}$ and $\Delta t_{\text{meas} \mu^+}$, confirming the consistency of our TOF analysis, including the correctness of all distances and the assumption that Mu and $\mu^+$ exiting the C-foil have the same kinetic energies $E_{\text{CF}}$.

3 Geant4 simulations of the energy losses in a thin carbon foil

A Geant4-based (version 9.4 patch 04) simulation of the beam propagation in the LEM beam line has been accomplished. Figure 6 shows the geometry implemented in the Geant4 simulation. Beam line components relevant to this study are included, from the L1 einzel lens, spin rotator (SR) to the sample chamber. The detailed geometry including radiation shield, sample holder and cold finger of the cryostat are also implemented.

Precise electric and magnetic field maps are inputs to the Geant4 Monte Carlo simulation. The magnetic field map of the SR was measured in a volume $10 \times 10 \times 150$ cm$^3$ around the origin of the SR coordinate system. The electric field of the SR was calculated using the commercial OPERA finite element programs (TOSCA/OPERA-3D) [26], and the electric field maps were calculated with the finite element software COMSOL [27]. Due to the modified sample plate setup in this work described in section 4, the electric field maps of the conical lens (RA) and the copper sample plate with cylindrical nose were re-calculated. The electrostatic module of COMSOL was used and a fine mesh was applied for higher accuracy calculations. A 2 mm spacing of grid points are used for the electric field maps. An example of the electric potential map in the sample region is shown in figure 10. Initial conditions of the $\mu^+$ beam are summarized in table 2. The typical number of events generated is ranging from $10^6$ to $10^7$, such that the statistical uncertainty is reduced to less than 1%.

The low-energy physics processes currently not available in Geant4 have been implemented to describe the energy losses and Mu formation processes in the thin C-foil of the start detector. When a $\mu^+$ is impinging on the C-foil our Geant4 simulation performs the following operations:
Figure 6. Schematic view of the LEM beam line as implemented in the Geant4 simulation. The beam which is given by the black solid line is simulated starting after the moderator. The arrows indicate spin (blue) and momentum (red) directions.

- Determine the charge state of the exiting muon, between $\mu^+$, Mu and $\mu^-$ using the yields given in figure 3.
- Calculate the energy loss (same for all particle charge states) assuming a Landau distribution with most probable value (MPV) given by the energy loss determined from the TOF measurements.
- Compute angular scattering using the standard Geant4 package for multiple-scattering.

More details regarding these three operations are given in the following subsections.

**Formation of muonium in the carbon foil.** Formation of “foil” Mu at the thin C-foil is implemented by velocity scaling of existing data from proton-C-foil data [11, 13, 14] as shown in figure 3. These “foil” Mu will be stopped when they reach a material interface.

**Muon energy loss in the carbon foil.** In Geant4.9.4, models simulating the $\mu^+$ energy loss and its fluctuation are implemented in the C++ class $G4\text{Muonisation}$. By default, for $\mu^+$ energy below 200 keV, $G4\text{BraggModel}$ is used where the energy losses are derived from the tabulated stopping power for proton using velocity scaling [15]. Energy loss fluctuation of $\mu^+$ is simulated by means of the $G4\text{IonFluctuation}$ model. For a thin absorber, the energy fluctuation is based on a very simple two energy-level atom model and the particle-atom interaction give rise either to an atomic excitation or an atomic ionization with energy loss distributed according to [16].

In our simulation, the $\mu^+$ energy loss is simulated based on the values determined from the TOF measurements and its fluctuation is implemented using a Landau random number generator based on CERNLIB [17]. It was found by L. Landau that a certain linear function of the energy loss
has, under certain assumptions, a universal (i.e. parameter free) density [18]. The generated random number from the universal Landau distribution $X$ is first shifted to have only positive energy losses $(X + 3.5)$ and then scaled linearly so that its MPV coincides with the measured $E_{\text{loss}}$ in table 1, i.e. the randomly generated energy loss distribution is

$$E_{\text{random loss}} = \left( \frac{X + 3.5}{3.5} \right) \cdot E_{\text{loss}}.$$  

(3.1)

Here, it is thus assumed that the energy loss distribution goes down to zero.

A comparison between energy losses extracted from the TOF spectrum and simulated energy losses using standard or our extended Geant4 version are shown in figure 7. A better agreement between simulations and measurements is achieved when parameterizing the energy losses using the Landau distribution of eq. (3.1). Interestingly, the Geant4 default simulation gives a better agreement below 0.5 keV. A cutoff approach was tried, however no improvement was achieved for the fitting of the TOF spectra. It is important to stress that the main attention is on the high-losses tail because it impacts the first few 100 ns of muSR measurements.

**Muon multiple scattering in the carbon foil.** In previous versions of Geant4, the measured transmissions of $\mu^+$ beam from the start detector till the sample were poorly reproduced due to the underestimation of the multiple-scattering process from C-foil [14]. However, recent versions of Geant4 have better physics models of multiple-scattering which reproduce correctly the Meyer scattering [13, 20]. In this paper, multiple Coulomb scattering is simulated by using *G4MuMultipleScattering* based on the model from Urban [21].
Figure 8. Measured and simulated TOF. The green curve represents the contribution arising from “unmoderated” muons which has been added to the simulated TOF as an empirical sum of two “Lorentzian” to achieve good matching with the data (see text for details).

Validation of the simulations. To validate the implementation of the low-energy processes, the simulated TOF spectra are compared to the measured TOF spectra. In figure 8 a comparison between simulated and measured TOF is presented. The green curve corresponds to the contribution of “unmoderated” muons which have to be assumed in order to match the measurements with the simulations. These are muons leaving the moderator not as epithermal muons at eV energy but as the non-fully moderated tail of the muon beam with keV energies.

About 40% of the $\mu E4$ beam hits the moderator target where about one half is stopped [5, 6]. This means that about 20% of the incoming $\mu^+$ beam will go through the moderator as “unmoderated” $\mu^+$ with a mean energy of several hundred keV and a low-energy tail ranging down to few keV energies [22]. Even though a large fraction of the “unmoderated” $\mu^+$ will not be reflected by the electrostatic mirror, they still contribute about 10-15%, depending on the moderator HV, to the total $\mu^+$ which are impinging on the sample. It has been shown [23] that the electrostatic mirror which is set at the same HV as the moderator ($V_{\text{mod}}$) deflects particles with kinetic energies ($E_k$) in the range $eV_{\text{mod}} < E_k < 2eV_{\text{mod}}$ by 90°.

Hence, “unmoderated” muons with slightly larger kinetic energy compared with the “moderated” muons are deflected towards the sample region. This explains why the “unmoderated” fraction contribute to the time spectrum at slightly earlier times compared with the “moderated” muons as well visible in figure 8.

The existence of “unmoderated” muons has been experimentally verified by taking data without any Ar-$N_2$ layer at the moderator. The corresponding TOF spectrum is shown in figure 9. Differently from the situation in figure 8 this measurement was performed with non-zero voltage at the conical lens. The contribution of “unmoderated” muons to the TOF spectra can be empirically
Figure 9. (a) TOF spectra of the “unmoderated” $\mu^+$ determined from a measurement without any Ar-N$_2$ layer on the moderator for various moderator and electrostatic mirror HVs (solid black line $-12$ kV, dotted red line $-13.5$ kV and dashed blue line $-15$ kV). (b) Measured “unmoderated” TOF at $12$ kV beam transport (blue dotted line) fitted with the sum of two Lorentzian functions (red solid line). For comparison a TOF spectrum (black dashed line) of “moderated” muons measured with the Ar-N$_2$ moderator rescaled by a factor of 10 is also shown.

The “unmoderated” muon contribution given by the green curve in figure 8 results from the sum of two “Lorentzian”. The relative widths and amplitudes of this two “Lorentzian” peaks cannot be assumed from the “unmoderated” measurement because of the different HV settings of the conical lens focusing the beam on the MCP2 and therefore are free parameters. In conclusion, simulated and measured TOF spectra agree very well together if a Landau distributed energy straggling in the C-foil is used and a small fraction of “unmoderated” muons is accounted for. It is important to note that the TOF spectra of figure 8 at times around $t \in [108;112]$ ns and $t \in [118;122]$ ns cannot be reproduced simply by modifying the muon energy losses in the C-foil assumed in the Geant4 simulation. The data in these two regions can be reproduced only by the contamination of “unmoderated” muons.

Simulations can then be used to determine the $\mu^+$ and Mu kinetic energy distributions and related arrival time distributions at the sample plate for any beam line settings (moderator HV, sample HV, conical lens HV etc). Slow $\mu^+$ and Mu tails cause detrimental distortions of the measured $\mu$SR time spectra which need to be accounted for when considering the “early” part of the measured time spectra. The starting point of the time window where $\mu$SR fit can be reliably applied without being distorted is dictated by the low-energy $\mu^+$ tail.

The knowledge of the fraction of “foil” Mu entering the LEM spectrometer is also an essential input for the analysis of the $\mu$SR data. As the precession frequency of the muon spin in the Mu atoms is a factor of 100 larger than that for a free muon [24], the “foil” Mu produced at the C-foil give rise to depolarization effects and reduction of the observable total decay asymmetry. This is...
because the Mu eventually stop not only in the sample region with a well defined constant magnetic field but also on the thermal shield of the sample cryostat and other elements which may have depolarizing effects and are placed at various B-field values. In the simulation, we assume that once Mu has formed, depolarization occurs independently of its states (singlet or triplet).

The recent upgrade of the LEM beam line was characterized mainly by the insertion of the spin rotator and the moving of the trigger detector closer to the sample region. The closer placement of the trigger detector to the sample region has caused a larger fraction of the “foil” Mu to reach the sample region, enabling a better study of the process related with “foil” Mu production. The insertion of the spin rotator opened the way for longitudinal $\mu$SR measurements, broadening the spectrum of possibilities available at the PSI-LEM spectrometer. In addition, the spin rotator was designed also to reduce beam contamination into the sample region. However the insertion of the spin rotator changed in a yet not fully understood way the beam propagation, increasing the beam size at the sample position. It is probably the insufficient knowledge of the fringe fields of the newly inserted spin rotator which do not allow an exact simulation of the transport of the muons.

4 Upstream-downstream asymmetry and the muon beam size

The beam size at the sample position is needed to normalize and analyze the $\mu$SR data and also to understand the LEM beam line and validate the Geant4 transport simulation. For example knowledge of the beam size at the sample position is used to remove the contributions arising from muons not impinging on the sample of interest. The so-called upstream-downstream asymmetry $A_{\text{ud}}$ can be used to infer the beam spot size at the sample position:

$$A_{\text{ud}}(t) = \frac{N_u(t) - N_d(t)}{N_u(t) + N_d(t)},$$ (4.1)

where $N_{u,d}(t)$ are the total number of decay positron detected as a function of time, in the upstream and downstream detectors surrounding the sample region as shown in figure 10(b). The values of
Figure 11. (a+c) The black dots are the simulated upstream-downstream decay asymmetry $A_{ud}$ for various RMS values $x_{rms}$ of the muon beam spot. These plots are used to determine the experimental muon beam sizes at the sample position. In the simulation, Mu is generated at rest on top of (a) a 1 mm thick $20 \times 20$ mm$^2$ fused quartz disk on top of the nose sample plate, for year 2011 setup, without spin rotator and (c) the Ag coated nose sample plate, year 2012 setup, with spin rotator in the low-energy muon beam line. The horizontal dotted lines are the measured decay asymmetries. (b+d) Muon beam spot at 14 keV implantation energy measured with the MCP2. The top (bottom) panels are before (after) the LEM upgrade. Quartz and Ag samples were used for the determination of muon beam sizes because Mu emission into vacuum is absent in these materials and $A_{ud}$ are time independent.

$A_{ud}$ given in this paper were obtained by fitting $A_{ud}(t)$ with a constant function for times larger than 200 ns. The fitted $A_{ud}$ for various implantation energies are shown in figure 11(a) and (c). The upstream-downstream asymmetry and its time evolution are also the central ingredient of the longitudinal $\mu$SR technique.

A MCP plate at the sample position can be used to perform not only measurements of the muon TOF, but also measurements of the beam profiles which are shown in figure 11(b) and (d). The MCP measurements provide a 2-dimensional profile of the muon beam, but it can only be used when there is no HV applied to the sample plate. On the contrary, the upstream-downstream
asymmetry is strongly correlated with the beam size, and it can be measured on-line and for any sample-plate HV. This plays a crucial role especially at low energy, when a high positive HV has to be applied to the sample plate ($V_{\text{sample}}$) to tune the $\mu^+$ implantation energy $E_{\text{implant}}$ which is given by

$$E_{\text{implant}} = eV_{\text{mod}} - eV_{\text{sample}} - E_{\text{loss}}.$$ 

(4.2)

The electric potential ensued by the HV at the conductive sample plate of figure 10(a) shows a curvature of the equipotential lines which gives rise to a radial force causing a defocussing of the muon beam and thus an increase of the muon beam profile at the sample plate. Note that this effect is particularly relevant for the “nose sample plate” shown in figure 10 which was developed for a dedicated experiment, which looked for thermal Mu emission into vacuum from mesoporous silica targets [25]. The standard $\mu$SR sample holder, being a simple plate, do not show such a strong curvature and therefore the beam defocussing effect is smaller.

A large variation of the beam size at the sample position for small variation of the beam parameters when using this “nose sample plate” has been observed. Because of this sensitivity a study of the $A_{\text{ud}}$ asymmetry using this “nose” sample plate for various beam line settings was performed to investigate the validity of the Geant4 beam transport simulation, which is also relevant for the analysis of the experiment for thermal Mu emission into vacuum [25].

The black lines in figure 11(a) and (c) shows the correlation between the beam size and the asymmetry $A_{\text{ud}}$. They have been computed assuming muon decaying from the sample plate with a given transverse spatial distribution described by a 2D Gaussian function with a width $x_{\text{RMS}} = y_{\text{RMS}} = \sigma_{x,y}$. Note that $A_{\text{ud}}$ can be larger than the theoretical-maximum-decay asymmetry of 0.33 due to the shielding effect of the nose sample plate on the downstream detector, i.e. the positron has a lower probability of reaching the downstream detector (according to eq. (4.1), $A_{\text{ud}} \to 1$ when $N_u \gg N_d$).

By comparing the measured asymmetries (horizontal dotted lines) with the asymmetry versus beam size predicted from the simulations, the beam size can be extracted. Then this beam size can be compared with the beam size obtained from a transport simulation of the full LEM beam line starting from the moderator till the sample region, including the processes in the C-foil and the electric fields in the modified sample region shown in figure 10(a). The asymmetry measurement can be thus used to validate the Geant4 beam transport of the LEM beam line.

Figure 11(a) and (c) show a decrease of the asymmetry for decreasing implantation energy revealing that the beam size increases considerably with decreasing energy. This has to be related to a substantial defocussing effect when the HV at the sample is increased due to stronger electric fields and slower muon velocity.

From the correlation line deduced from Geant4 simulation as depicted in figure 11(a), the muon beam size at 14 keV implantation energy has a RMS value $\sigma_{x,y} = 6.9$ mm. This value compares well with the value measured with the MCP2 of 6.3 mm (The MCP2 is placed 18.5 mm downstream of the nose sample plate and hence a slightly smaller beam spot is expected) and with the value of 7.2 mm computed with Geant4 beam transport.

However for the 2012 measurements, after the LEM upgrade, the beam size values obtained from the MCP measurements and the values from the asymmetry measurement do not agree with the beam size obtained from the Geant4 simulation of the beam propagation in the LEM beam line. This calls for a verification and detailed investigation of the LEM beam line simulations. To study
Table 2. LEM beam line initial settings for the Geant4 simulations and initial beam parameters.

|                        | High voltage setting (kV) |
|------------------------|---------------------------|
| Moderator              | 12                        | 15                        |
| Einzel lens, L1        | 7.19                      | 8.99                      |
| Mirror                 | 12                        | 15                        |
| Spin rotator, $E^x_{SR}$ | 2.09                      | 2.29                      |
| Spin rotator, $B^z_{SR}$ | 62.3 G                    | 69.7 G                    |
| Einzel lens, L2        | 8.38                      | 10.484                    |
| Lens, L3               | 8.99                      | 11.483                    |
| Conical lens, RA       | 9.15                      | 11.9                      |

The sensitivity of $A_{ud}$ to various beam line settings the sample holder of figure 10 is used. The beam line settings of table 2 are for the SR setup, experimentally optimized.

The dependence of $A_{ud}$ on the various beam line parameters has been investigated and summarized in figure 12.

- (a) Muon implantation energy: $A_{ud}$ is decreasing with decreasing implantation energy due to defocussing effects caused by the electric potential applied at the sample holder (see figure 10(a)). The musrSim (new) represents the Geant4 simulation where the muon energy losses at the C-foil are parametrized using Landau distributions. From figure 13, it can be seen that for a lower implantation energy, there is a considerable amount of $\mu^+$ which do not hit the “nose sample plate” which has a radius of 15 mm. Thus at lower implantation energy, there is a higher probability of decay positron being detected by the downstream detector, resulting in a reduced $A_{ud}$ asymmetry.

- (b+c) Initial phase space of the muon beam: initial phase space $(x',y')$ after the acceleration section of the moderator is taken to be $\sigma_x = \sigma_y = 7.5$ mm for the beam size and $\sigma_{x'} = \sigma_{y'} = 2.0^\circ$ for the beam divergence from a recent simulation [13]. The initial polarization vector is chosen as $\mathbf{P}_\mu = (0.9848, 0, 0.17365)$ since the $\mu^+$ spin is rotated by $10^\circ$ clockwise after traversing the static separator of the $\mu$E4 beam line before the $\mu^+$ is focused on moderator (see figure 1).

- (d) Electric and magnetic field of the spin rotator: the magnetic field of the spin rotator was fixed to $B^z_{SR} = -62.3(-69.7)$ G for 12(15) keV transport energy to obtain the experimental $\mu^+$ spin rotation of $20^\circ$ counter-clockwise. The spin angle with respect to the $\mu$E4 beam direction (corresponding to $-x$ direction in the simulation) is then changed from $+10^\circ$ (the angle after the separator in the $\mu$E4 beam line) to $-10^\circ$. The electric field was varied
Figure 12. Measured and simulated $A_{ud}$ versus $\mu^+$ implantation energy. $\sigma_{xy}$ and $\sigma_{x'y'}$ are the standard deviation of the muon phase space after the acceleration section of the moderator, $E_{SR}^x$ the electric field in the spin rotator and $V_{RA}$ the HV at the conical lens. For each plot only one parameter is varied whereas the other one are given in table 2.
Figure 13. Simulated energy of the muon beam at sample-plate plane as a function of distance $R$ from the beam axis for 2 different muon implantation energies: (a) 11 keV and (b) 3 keV. Region 1 is mainly contributed by muons with kinetic energy above 9.15 keV before reaching the RA. Region 2 is mainly given by muons with energy around 9.15 keV ($= V_{RA}$) before reaching the RA. Region 3 is originated from Mu, which are not decelerated by the electric fields in TD and are not affected by the RA and sample plate electric potentials, and hence have higher energies than the $\mu^+$. Since Mu are not focused by the RA lens they have a distribution with larger transverse extension.

from $E^{SR}_x = 1.85$ to 2.25 kV/mm and is chosen such that the beam is centered on the original sample plate position (16 mm downstream of the nose sample plate) as shown in figure 14(a). From figure 12(d) it is visible that that $A_{ud}$ is maximal at around 2.09 kV/mm. This occurs when the beam is centered on the sample plate (see figure 14(a)) and the downstream detectors are shielded from the positron by the material of the “nose sample plate” itself.

The design value of 2.68 kV/mm which was expected from simply considering the relation $v = \frac{E}{B}$ has thus to be tuned to 2.09 kV/mm to center the beam on the axis. This indicates that there are not well understood mismatch between the electric and magnetic fields that cause the $\mu^+$ to deviate from its original path.

- (e) Electric potential of the conical lens (RA): if the focusing power is not optimal the $A_{ud}$ decreases since the beam becomes larger.

- (f) Z-offset of the sample plate position: it is obvious that there is a strong dependence of the $A_{ud}$ on the z-position of the “nose sample plate”. Moving the sample plate downstream, in z-direction, will decrease the $A_{ud}$ as the downstream detector will be less shielded by the sample plate from positrons.
Figure 14. (a) Simulated $\mu^+$ beam mean position $(\overline{x}, \overline{y})$ at the sample plate as a function of the electric field in the spin rotator $E_{SR}^x$. The $E_{SR}^x$ value in table 2 is chosen such that the beam is centered on the original sample plate position (16 mm downstream of the nose sample plate). (b) Measured $A_{ud}$ versus $\mu^+$ implantation energy with and without Ar-N$_2$ layer at the moderator. “Unmoderated” muons at the plate position have a larger beam size.

Table 3. Measured $\mu^+$ beam spot $x_{RMS}$ and $y_{RMS}$ for the “unmoderated” $\mu^+$ and “moderated” $\mu^+$ using the MCP2. It should be noted that due to the finite size of the active region of the MCP2 (42 mm in diameter) the actual $x_{RMS}$ and $y_{RMS}$ could be larger, especially for the “unmoderated” $\mu^+$.

| Energy (keV) | Spin rotator | Moderator | $x_{RMS}$ (mm) | $y_{RMS}$ (mm) | $\mu^+$ type |
|-------------|--------------|-----------|----------------|----------------|--------------|
| 15          | No           | No        | 6.1            | 6.0            | unmoderated  |
| 15          | No           | Yes       | 5.3            | 4.9            | moderated    |
| 15          | Yes          | No        | 7.2            | 7.2            | unmoderated  |
| 15          | Yes          | Yes       | 6.2            | 6.0            | moderated    |

Summarizing, the new musrSim with Landau distributed losses give asymmetries closer to the measured one. However, the agreement is not yet satisfactory. Some parameters could be slightly tuned around the design value to decrease the deviation between measured and simulated $A_{ud}$ asymmetries. The newly inserted spin rotator optimal settings do not correspond to the design value manifesting that some uncontrolled beam distortion and beam transmission may occur at this beam line element. Contribution of “unmoderated” $\mu^+$ is shown in figure 14(b). As they have a larger beam spot ($x_{RMS}$ and $y_{RMS}$) as shown in table 3 and hence a lower value of $A_{ud}$. The larger beam spot could be caused by chromatic aberration in the beam transport optics due to the wide energy distribution of the “unmoderated” $\mu^+$. But this is not enough to explain the observed discrepancy between experimental and simulated $A_{ud}$ asymmetries as they accounted for only 10-15% of the measured time spectra. However, as shown in figure 12, by tweaking different parameters one could get agreement. We refrain at this stage to perform a multi-parameter tuning because of the complexity and the correlation between the various parameter.

The reason why there was a good agreement between the simulated and measured $A_{ud}$ prior to the LEM beam line upgrade, is related with the smaller beam size at the sample plate which imply a reduced defocussing effect at the “nose sample plate”. The insertion of the spin rotator has
caused a degradation of the beam quality and an unexpected increase of the beam size at the sample position. This increase could be even more substantial for the “unmoderated” muon component. The larger RMS values ($\approx 1.2$ mm) for “unmoderated” muons could be sufficient to explain the observed smaller asymmetries in figure 12.

5 Conclusions

Simulations of the complete LEM beam line after the 2012 upgrade have been presented. TOF measurements have been used to calibrate the energy losses in the start detector. Excellent agreement between the measured TOF spectra and simulations has been reached only by using Landau distributed energy straggling, accounting for muonium production, and accounting for a contribution of “unmoderated” muons with slightly larger kinetic energy which are parasitically transported by the beam line. The measured stopping power in the C-foil compares well with previous determination. This good agreement between measured and simulated TOF spectra implies also the correctness of the assumed muonium yield in the C-foil which has been implemented using velocity scaling of proton data.

Detailed analysis of the $\mu$SR time spectra require information of the muon arrival times, muon implantation energy and depolarization effects related with energy losses and Mu production in the C-foil. These information can be determined now for all beam line settings and muon implantation energies using the new Geant4 simulations.

The beam spot size at the sample position is also a very important parameter when analyzing the $\mu$SR data. Normalizations and total measurable decay asymmetries depend on this parameter. This is even more important for longitudinal $\mu$SR which is now possible due to the insertion of the spin rotator. Therefore the beam spot size at the sample position has been investigated by means of the $A_{\text{ud}}$ asymmetry using a dedicated sample plate (“nose sample plate”) with increased sensitivity to beam changes. These studies have revealed a problem with the beam transport in the LEM beam line related with the newly inserted spin rotator and the parasitic transport of “unmoderated” muons which requires detailed studies and hardware improvements.

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