Monte-Carlo simulation of transitions between different muonium states

T. Prokscha

Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland

Abstract

A Monte-Carlo method for the generation of muon decay histograms in the presence of charge-exchange processes between different muonium states is presented. The simulation is based on the ROOT toolkit developed at CERN. Histograms are saved in root file format and can be directly analysed using the musrfit framework recently developed at PSI [1]. The simulation is tested by comparing with the measured temperature dependence of the diamagnetic asymmetry in Si in the Mu0^BC ionization regime.

Keywords: Monte-Carlo simulation, muonium charge-exchange

In semiconductors transitions between various muonium states are possible, depending on temperature and the concentration of free charge carriers. In the elemental group IV semiconductors, for example, bond-centred muonium (MuBC) exists as Mu0^BC or Mu^+BC^, and MuT at the tetrahedral site as Mu0^T or Mu^−T. Thermally activated transitions between these states are well known [2, 3, 4]. Recently, we started at the low-energy μSR setup [5, 6, 7] of the Swiss Muon Source SμS a development to use illumination to change charge-carrier concentrations in a near-surface region of ~200 nm [8]. One purpose of this development will be also the investigation of photo-induced transitions between different Mu states. Even a photo-induced change of observable asymmetries could be possible, as has been recently reported in a bulk muon chemistry experiment on liquid water [9]. In order to study in more detail the effects on muon dynamics caused by changes of the electron capture/ionization and hole capture rates due to photo-generated charge carriers we developed a Monte-Carlo simulation following the approach of the time-ordered stochastic method of Senba [10, 11, 12, 13], which was introduced to investigate muon spin dynamics in muonium in the presence of charge-exchange and spin-flip/non-flip collisions. The Monte-Carlo procedure is simple and allows additionally to examine these effects at variable statistics in μSR histograms.

In a basic study we focus on muon/muonium dynamics in Si at temperatures below 250 K where it is sufficient to consider only transitions between Mu0^BC and Mu^+BC^, since MuT ionization and site changes become relevant only at higher temperatures [3, 14]. For simplicity spin-exchange collisions are ignored at the moment but they can be easily added to the existing code. In this simplified model we consider two processes:

1. Mu0^BC^ formation by electron capture, MuBC^+ + e^− → MuBC0 with the electron capture rate \( \nu^e \)

\[ \nu^e = \sigma^e_n \nu_n n, \]  

(1)

where \( \sigma^e_n \) is the cross section for electron capture, \( \nu_n \) is the temperature dependent electron velocity, and \( n \) is the electron concentration.

Email address: thomas.prokscha@psi.ch (T. Prokscha)
2. Mu$_{BC}^0$ ionization by
   (a) thermal activation, Mu$_{BC}^0 \rightarrow$ Mu$_{BC}^+ + e^-$
   (b) hole capture, Mu$_{BC}^0 + h^+ \rightarrow$ Mu$_{BC}^+$
with the ionization rate $\nu^j$

$$\nu^j = v_0 \exp (-E_A/k_B T) + \sigma^j_p v_p p,$$
(2)

where the prefactor $v_0$ is of the order of $10^7$ MHz, $E_A$ is the thermal activation energy of Mu$_{BC}^0$. $\sigma^j_p$ is the cross section for hole capture, $v_p$ the temperature dependent hole velocity, and $p$ is the hole concentration.

Note, that $\sigma^e_p \sim 100 \cdot \sigma^j_p$ because electron capture of a positive center usually occurs into excited states with large radius. Since $\sigma^e_p$ is of the order of an “atomic” cross section of about $10^{-15}$ cm$^2$, $\sigma^j_p$ can reach enormously large values of $10^{-12} - 10^{-13}$ cm$^2$ [15]. Assuming equal electron and hole densities in an illumination experiment the electron capture rate is about two orders of magnitude larger than the hole capture rate.

In the simulation we assume that the capture and ionization processes are Poissonian. This means that the probability density $dP$ for the occurrence of the corresponding process at time $t$ is given by

$$dP(t) = \nu^j \exp (-\nu^j t) dt.$$
(3)

Integrating Eq. 3 gives the time $t^{j-1}$ of the next capture or ionization event as a function of a random number $\eta$:

$$t^{j-1} = -1/\nu^j \log (\eta), \quad \eta \in [0, 1].$$
(4)

In the same way the muon decay time $t_d$ is “thrown”: $t_d = -\tau_\mu \log (\eta)$ with the muon life time $\tau_\mu$.

The simulation begins with the determination of the initial muon state (either Mu$_{BC}^+$ or one of the Mu$_{BC}^0$ states) according to the initial Mu$_{BC}^0$ fraction. Assume that this is Mu$_{BC}^-$(for Mu$_{BC}^0$, it works correspondingly): it calculates as a next step the muon spin phase $\phi_\mu(t^1_c) = \omega_\mu t^1_c$ at the time $t^1_c$ of the first capture event, where $\omega_\mu = 2\pi v_\mu$ is the free muon precession frequency in a transverse magnetic field. The evolution of the muon phase in the following sequence of charge-changing processes is then given by

$$\phi_\mu(t^i_c + t^i_c) = \omega_\mu t^i_c + \omega_{Mu}^j t^i_c \quad i = 1, 2, \ldots$$

where $t^i_c$ is the time between the 1st electron capture and 1st ionization event, $t^i_c$ is the time between $t^i_c$ and the 2nd capture process and so on. This sequence is terminated when the time $(t^i_c + t^i_c + \ldots)$ exceeds the decay time $t_d$. A muon decay histogram is generated where at the time bin corresponding to $t_d$ the histogram counts are incremented by $(1 + A \cdot \cos \phi_\mu)$, with $A$ the decay asymmetry. In the sequence above the Mu$_{BC}^0$ frequencies $\omega_{Mu}^j$ can be different because of various Mu$_{BC}^0$ states: the simulation “throws” – according to the amplitudes of the Mu$_{BC}^0$ states – the Mu$_{BC}^0$ frequency to be used until the next ionization event. Note, that $\omega_{Mu}$ can be zero because of the possible non-precessing fraction in anisotropic Mu$_{BC}^0$ depending on the size and orientation of the external magnetic field with respect to the symmetry axis of Mu$_{BC}^0$.

Input parameters for the simulation are the externally applied transverse field $B_{ext}$, the Mu$_{BC}^0$ fracton, the Mu$_{BC}^0$ precession frequencies and corresponding amplitudes for a given geometry ($B_{ext} || (100)$, for example), the rates $\nu^j$ and $\nu^e$, and the total number of muons. The simulation is based upon the R00T toolkit developed at CERN. Histograms are generated and saved to files, which can be directly analyzed with the musrfit framework [1]. The simulation is implemented as a R00T class (PSimulateTransition) and is contained in the tests section of the musrfit software package, which is distributed under GNU GPL. A R00T macro runMuSimulation.C is available to run the simulation interactively in a R00T session. On a TwoCore 2.8 GHz PC one run with $10^7$ muon decay events takes between a few seconds and a few minutes, depending on the capture and ionization rates; the higher these rates the more charge-exchange steps have to be computed until the muon decays.

As an example we show in Fig. 1 the effect of increasing ionization rate ($\nu^e = 0$ fixed) on the Mu$_{BC}^0$ signal. The Mu$_{BC}^0$ lines first become broader as expected, and since $\nu^e = 0$ the Mu$_{BC}^0$ signal with small relaxation begins to appear
Figure 1: Simulated Si data, decay asymmetry 0.27, $10^7$ events, $B_{ext} = 1 \text{ kG} \parallel (100)$, assuming 100% $\mu^0_{BC}$ fraction. The theoretical amplitudes of the lines are 0.27 for $\nu_{12}$, 0.47 for $\nu_{34}$, and 0.26 non-precessing. Red lines: fit to the data (black). a) and b) $\nu' = 0$, time and Fourier spectra, respectively, $\mu^0_{BC}$ non-relaxing; c) $\nu' = 1 \text{ MHz}$, d) $\nu' = 10 \text{ MHz}$ (corresponding to $T \sim 160 \text{ K}$), muon line $\nu_\mu$ starts to become observable.

Figure 2: Asymmetry $A_D$ of the diamagnetic signal $\mu^+_{BC}$ in nominally undoped Si (100) as a function of temperature at 100 G and 1 kG. The temperature $T$ is related to the ionization rate $\nu'$ by Eq. 5, with $E_A = 215 \text{ meV}$ and $v_0 = 3 \cdot 10^7 \text{ MHz}$. a) simulation data with and without non-precessing $\mu^0_{BC}$ component, assuming 100% $\mu^0_{BC}$ fraction. b) Solid symbols: experimental data, measured at the GPS instrument at PSI. Open symbols and lines: simulation with non-precessing fraction of 26%. The asymmetry in the simulation is scaled to match the experimental asymmetry at 260 K.
at $\nu' \sim \nu_\mu$ (Fig. 1d). The simulation has been tested as follows: the observable $\mu_0^{BC}$ asymmetry should depend on the applied field in the $\mu_0^{BC}$ ionization region (140 - 250 K), if the non-precessing $\mu_0^{BC}$ component is nonzero, see Fig. 2a. Figure 2b compares measured data with predictions from the simulation. Here, only the $\mu_0^{BC}$ thermal ionization process is taken into account, which means $n = p = 0$ in Eqs. 1 and 2. The temperature $T$ is then related to $\nu'$ – which is the input parameter for the simulation – by

$$T = \frac{E_A}{k_B(\log(v_0) - \log(\nu'))},$$

with $v_0 = 3 \cdot 10^7$ MHz and the activation energy $E_A = 215$ meV [14]. The qualitative agreement is excellent: a splitting at 140 K and merging at 220 K of 100 G / 1 kG data. This additionally confirms the presence of the non-precessing $\mu_0^{BC}$ component. In the experiment the non-precessing $\mu_0^{BC}$ fraction is directly observable as a shift of the $\mu$SR $\alpha$ parameter at $T \lesssim 140$ K, below the $\mu_0^{BC}$ ionization region. The simulation shows that the non-precessing $\mu_0^{BC}$ component is the origin of the field dependence of the $\mu_0^{BC}$ asymmetry in the ionization region. This can be explained the following way: if $\nu' \gtrsim \nu_\mu$ the transition from non-precessing $\mu_0^{BC}$ to $\mu_+^{BC}$ is fast enough to obtain a coherent precession signal at $\nu_\mu$. At 1 kG $\nu_\mu$ is $\sim 12$ MHz larger than at 100 G which means that an at least 12 MHz larger ionization rate (i.e. a higher temperature) is needed at 1 kG to obtain an increase in the diamagnetic asymmetry.

Compared to the simulation the deviation between 100 G and 1 kG data between 160 K and 200 K is smaller in the experiment. This arises from the simplification in the simulation where the non-precessing muon spins are pointing towards the positron detector. This causes a larger decay asymmetry than in the experiment, where the direction of the non-precessing muon spins depends on the symmetry axis of the $\mu_0^{BC}$ state and the applied magnetic field, so that only a projection of spin polarization is pointing towards the positron detector.

In summary we have shown that the described simple and fast Monte Carlo approach is well suited to study transitions between different Mu states. It has been successfully tested by comparing the predictions of the simulation with temperature dependent changes of $\mu_0^{BC}$ in Si. Simulations with $\nu'$, $\nu' \neq 0$ are in progress to study effects of illumination on the $\mu_0^{BC}$ signals. The simulation can be easily extended to account for additional processes, such as spin-flip collisions or charge-changing cycles involving $\mu_i^+$. I would like to acknowledge the support of Hubertus Luetkens and Robert Scheuermann in the Si measurements on the GPS instrument of SpS. I am grateful to Kim Chow whose computer code I used to calculate the $\mu_0^{BC}$ frequencies and corresponding amplitudes.

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