Journal of Physics: Conference Series **2462** (2023) 012025 doi:10.1088/1742-6596/2462/1/012025

Depth profiling of LE-μSR parameters with `musrfit`

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Abstract. The study of thin-film and multi-layered structures with nanometer resolution is possible with low energy μSR (LE-μSR). Modeling of the measured μSR parameters such as diamagnetic asymmetry and relaxation rate as a function of sample depth can be obtained from a series of experimental implantation energy measurements and its correlation with the simulated stopping profiles. The fitting approach assumes a sharp transition between regions with distinct properties. The fitting method, previously developed in matlab, was implemented in `musrfit`, a free μSR data analysis framework written in C++. The main goal is to make this fitting method widely available for energy dependent measurements and to increase the modeling possibilities within `musrfit`.

1. Introduction

Study of thin-films and heterostrustures is possible with low-energy muon-spin spectroscopy due to the tunability of the muon implantation energy in the range of 1 to 30 keV. However, for each implantation energy the muons' stopping depth width extends over a few nanometers and depends on the density of the studied material. `musrfit` is a software tool used for analysis of time differential μSR data [1]. Additionally, it is possible to implement user-defined functions in the `musrfit` suite[2]. Here, we describe the implementation of the depthProfiles user function, which models in `musrfit` the depth dependence of the μSR parameters measured as function of energy in the real space.

2. Fitting approach

The fitting strategy assumes that the parameters have a characteristic, constant value for each region, and between each region there is a sharp transition. The variation of the properties of the sample can be due to the existence of layers of different materials, presence of defective regions, intermixing near the interface, or, depletion regions where the charge carriers concentrations may change [3; 4]. This is represented as a step-like behavior of the LE-μSR parameter as function of sample depth.

Smooth transitions between the different sample regions have been tested as well. However, the results did not provide a better fit [5]. Typically the experimental data is not recorded for finer energy steps than 1 keV. Thus, the amount of data points is not enough to justify a more complex modeling function.

In a three-layer system, for example, the normalized μSR parameter *f(E)*, measured as a function of energy (for example, the diamagnetic fraction), is then fitted to the function:

$$f(E) = p_{0a}(E)f_{0a} + p_{ab}(E)f_{ab} + p_{b\infty}(E)f_{b\infty}, \tag{1}$$

where f_{ab} is the value of the μSR parameter in the region between a and b, and p_{ab} is the probability of the muons stopping in the same region for a given implantation energy (figure 1) and calculated as:

$$p_{ab}(E) = \int_a^b P(x,E)dx. \tag{2}$$

$P(x,E)$ is the probability of the muon beam implanted with energy E to stop at a depth x (figure 1), and is calculated using the Monte Carlo simulation TRIMSP [6]. The muon stopping distribution data (`rge` files) is read at startup of the fitting procedure.

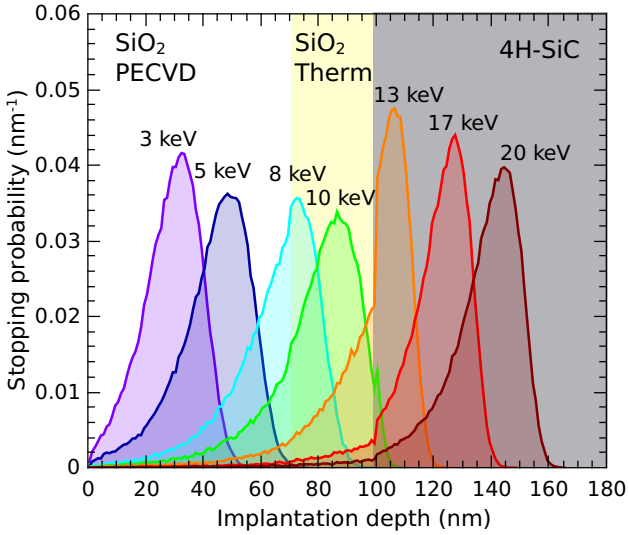

Figure 1. Stopping distribution of the low-energy μ^+ in a 70 nm PECVD-SiO_2/ 30 nm thermal-SiO_2/ 4H-SiC sample. The stopping profiles of the muons was simulated with TRIMSP for 10^5 muon events per energy value.

The depth dependence of the μSR parameter can then be modeled with a step function in the case of a three-layer system:

$$f(x) = \begin{cases} f_{0a} \text{ for } 0 \leqslant x < a, \\ f_{ab} \text{ for } a \leqslant x < b, \\ f_{b\infty} \text{ for } x > b \end{cases} \tag{3}$$

The function fits as many steps as necessary according to the number of initial parameters set by the user. The initial values for these parameters and description of the fitting model are provided in the `msr` input file to `musrfit`, which can be edited on the `musredit` interface. Here, it is given an example for a 3-step fit, where five fit parameters are used: f_{0a}, f_{ab} and $f_{b\infty}$ are the diamagnetic fractions characteristic of each layer, and a and b are the depth at which there is an abrupt change of the properties.

More generally, for n steps: f_{0a}, ..., $f_{n\infty}$ and x_1, ..., x_{n-1}. Where f corresponds to the characteristic value of the μSR parameter in each layer and x is the transition depth between regions.

3. Example

Here the step-fit analysis in `musrfit` is exemplified for a three-layer sample consisting of two layers of SiO_2 (70 nm deposited and 30 nm resulting from thermal oxidation of silicon carbide) on SiC. The two growth methods of the oxide result in SiO_2 layers with distinct properties: plasma enhanced vapor deposition (PECVD) leads to an oxide layer with lower density and higher structural disorder, whereas thermal oxidation is known to produce a better quality SiO2. The obtained values for the thickness of PECVD SiO_2 are lower than the projected thickness of 70 nm. Although the thickness of the PECVD grown SiO_2 can be very well controlled, there can be an uncertainty of ± 3 nm. In this case the fit result shown in figure 2 is acceptable. On the other hand, the oxidation process creates a transition layer at the SiO2/SiC interface. In this region the oxide contains SiO_xC_y and not only SiO_2 [7], due to the carbon atoms stopping mainly near the interface during the early stages of oxidation[8]. The width of this layer is not exactly known due to the lack of characterization methods with appropriate depth resolution. By performing the depth dependence analysis (figure 2) we could extract the width of the substoichiometric SiO_xC_y region to be 5 nm. During the fit of the data an aditional layer was considered for the step-fit procedure, however the added defective layer thickness had a large uncertainty. Furthermore, the density of the defective region is necessary to obtain the stopping profiles of the muons and this value is not known. Thus in this analysis, this defective region is considered to be the extension of the thermal SiO_2 layer in the SiC, with enhanced diamagnetic fraction. Here, the point defects contribute to trapping of charge carrier and suppression of neutral muonium formation. In this example the diamagnetic fraction variation is used as a sensor for the presence of defects [9], allowing to distinguish between oxides with different quality, and to measure the extension of the defective region.

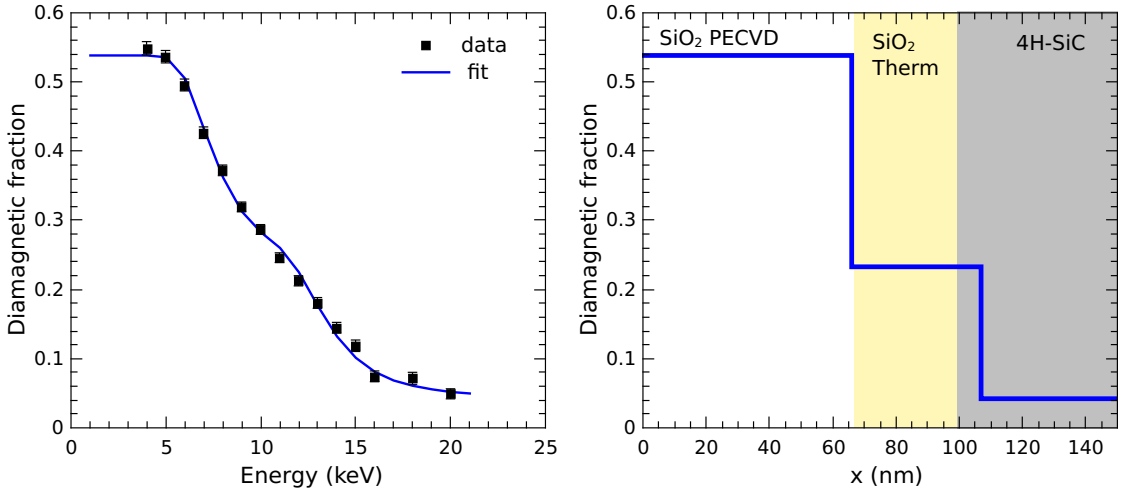

Figure 2. Fit result for the three-layer sample 70 nm+30 nm SiO_2 on 4H-SiC **Left:** Fit to the experimental data measured as a function of muon implantation energy. **Right:** Model of the depth dependence of the diamagnetic fraction.

The `musrfit` fit is in good agreement with the results obtained with the existing fitting function implementation in matlab (see table 1). However, the implementation of this fitting possibility in `musrfit` makes it easier to use and to fit directly the μSR parameters resulting from the analysis of the asymmetry spectra. In order to overcome some limitations in the cases where the density of the investigated material is not well known, an improved fitting method was been proposed in [10], and can in the future be incorporated with the depthProfiles user function.

	f_{0a}	f_{ab}	$f_{b\infty}$	a (nm)	b (nm)
`musrfit`	0.538(5)	0.232(1)	0.042(2)	66(1)	107(3)
matlab	0.543(7)	0.229(10)	0.046(7)	62(1)	104(1)

Table 1. Comparison of the fit parameters obtained with the fitting routine implemented in `musrfit` and matlab.

4. Conclusion

We have shown the implementation of a user-defined function in `musrfit` very useful for the fitting of LE-μSR data and modeling of the μSR parameters, such as diamagnetic fraction or asymmetry, in real space. Using the depthProfiles user function it is possible to accurately determine the width of relevant regions in thin-films.

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