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Joint negative muon data analysis and Monte Carlo simulation methods for the characterization of thin layers

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Summary. — Muonic atom X-ray Emission Spectroscopy (μXES) is a novel technique in the broad field of non-destructive methods for cultural heritage analysis. It relies on the interaction of a probe of negative muons with matter and following emission of X-ray radiation. Since the muon mass is about 207 times bigger than the electron, these emitted X-rays are highly energetic and are characteristic of the emitting atom, making it possible to cover a wide part of the periodic table (from lithium to uranium). The multi-elemental range, a negligible self-absorption of the X-rays and very low residual activity left in the sample after irradiation make μXES a very powerful probe for material characterization. In addition, by coupling the data analysis with Monte Carlo simulation methods, it is possible to asses the depth of the layers that are present in a given sample. In this work, preliminary results of the analysis on two gilded surfaces are reported.

1. – Introduction

After being neglected for some years, the use of negative muons in archaeometry has seen a resurgence in the last decade, with new works devoted to the characterization of cultural heritage artefacts [1-3]. Muons are fundamental particles defined by heavy mass, 207 times the one of the electron and a negative charge, which is why they are often defined as "heavy electrons". When negative muons are implanted into matter, they are captured in the outer shell of the atoms to form the so-called "muonic atom". After capture, the muon starts to cascade down to lower muonic orbitals, a process that generates the emission of high-energy X-rays that are characteristic of the emitting atom. The Muonic Atom X-Ray Emission Spectroscopy technique (μXES) uses this process to characterize materials (for a complete survey see [4]). The technique represents a new and very powerful approach to the study of artistic and historic artefacts, as material composition can be determined as a function of depths and no surface preparation is required. The muon beam energy, indeed, can be tuned so that muons probe at different depths of the sample in a non-destructive way. Furthermore, the emitted high energy X-rays (from a few keV up to ∼8 MeV), that cover a wide range of the periodic table, can overcome problems of self-absorption found in common techniques and can give information from deep inside a sample. In this work, we propose a methodology for the interpretation of negative muon data. What we explore is the possibility of using Monte Carlo (MC) simulations software to provide complementary information to the negative muon data analysis. With MC software, one can model the sample and the experimental set-up to replicate the same conditions as the real measurements and simulate muon interaction with materials. By adjusting simulation parameters, one can replicate the same conditions of the real experiment and compare the results. In this way, it is possible to assess the thickness of a layer present in the sample. Two different types of MC software were used: SRIM-TRIM and Geant4/ARBY [5,6]. The latter is a Geant4 based software developed at the university of Milano Bicocca that provides all tool of Geant4 in a more user friendly way [7]. In ARBY, it is possible to model both the sample but also all the experimental set-up (detectors, beam exit, etc.). Here, the results of a real experiment with the ones obtained by the simulations are compared.

2. – Materials and methods

The samples used for this comparison are two small laboratory made pieces of brass (SM3) and bronze (EM2) covered with a thin layer of gold (rectangular shape, $45 \times 25 \times 5$ mm). Both samples have a two-layer structure, made of a thin gold layer on top of a copper based matrix. Negative muon experiment was performed at RIKEN-RAL facility (port4) of the ISIS Neutron and Muon source [8]. The experimental setup consisted of 4 HPGe detectors placed at 15 cm and with a 30° angle from the sample position, that is 10 cm in front of the beam exit. For the experiment, the beam was not collimated. A momentum scan was performed from 15.5 MeV/c up to 24 MeV/c for an average measuring time of 4 hours. For each run, an X-ray energy spectra was recorded. The spectra were analysed by means of peak identification and peak fitting. Gold is identified from many peaks at different energies: here, the 130 KeV peak was used, since it resides within the energy range of best efficiency for the upstream detectors. From that, a profile of the variation of the peak area with the momentum (hence, the variation of the penetration depth of the beam) was determined. To asses the size of the gold layer, the samples were modelled in the simulation software. Separately, a SEM scan was performed on a sample from the same batch and a thickness of $10 \pm 1\,\mu$m was assessed. This value was taken as a starting reference for sample modelling. In SRIM-TRIM, modelling consisted in the definition of all the layers involved in the measurement: a Mylar window, the air gap between beam exit and sample surface and the gold and copper based layer. In Geant4/Arby, instead, the samples are modelled geometrically, as well as the experimental area, in a configuration file that stores all the information about the material and the setup. In both software, the modelling tries to replicate the real samples in the best possible way. For the measured sample, this was done easily, since the geometry is quite simple, with just two different layers of copper and gold. Finally, the muon interaction was simulated and, as a result, from both software, the number of muons stopped in each layer is given. Since all the muons are captured by

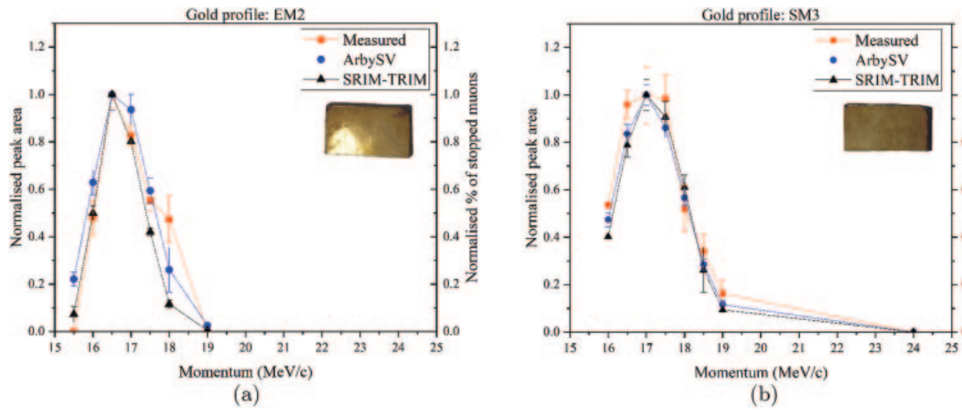

Fig. 1. – Gold depth profile from measured and simulated data analysis; (a) EM2 sample; (b) SM3 sample. Even if the sample looks similar, muon data suggest a different size of the gold layer.

the material they are implanted in, this means that the number of muons implanted in each layer is proportional to the intensities of the emitted X-rays. The simulated data, indeed, can be compared to the measurement. Moreover, since the emitted X-ray are of high energy and the investigated layer, self-absorption is not a problem. To asses the deviation from measured and simulated values, both the results from the experiment and the simulations were normalised to 1, and a chi-squared test was performed. Finally, it has to be mentioned that Geant4/Arby could reproduce the simulated X-ray spectrum, but some more work is required to compare the simulated spectra to real spectra.

3. – Results and discussion

Figure 1 shows the different results obtained from the data analysis of the real and simulated experiment. For EM2, the best fit was reached with a gold thickness of 5 μm and a density of the material decreased by 25% from the nominal value of gold (19.32 g/cm^3). Here, a reduced χ^2 of 2.60 for both SRIM-TRIM and ARBY was obtained. For sample SM3, instead, the best fit was reached with a thickness of 11 μm and standard gold density. A reduced χ^2 of 1.23 for SRIM-TRIM and 2.40 for ARBY was obtained. Especially in this sample, the simulated results are in agreement with the measured one, but also in agreement with each other. Furthermore, the value obtained for SM3 is close with the value seen in the SEM scan (10 μm). In both cases, simulation was performed by starting with a fixed gold thickness and then adjusting it depending on the results. For EM2, the process was more difficult, since the simulated gold profile with thickness around 10 μm and standard gold density produced an output with big deviation from the real values. So, assuming that here the gold layer was thinner than in the other sample, size was decreased as well as density. This was done to try to replicate the presence of air bubbles in the layer, that cannot be modelled in the simulation (for SRIM-TRIM especially). The manufacturing process used to make the sample, indeed, could have left bubbles of air in the layer that could be responsible for the shape of the profile. Modelling an uneven layer is more complicated, and that is why the results for the EM2 sample are not as good as the SM3 sample. Regarding the comparison between the two software, the results testify a good agreement. This is important especially for the GEANT4/ARBY tool, which has never been used for this type of analysis. Still, there are some discrepancies, especially in the case of the EM2 sample. Here, however, it is necessary to take into account that this sample has a more complicated structure and it is more difficult to have a good result. In ARBY, one can also modify the shape of the sample so that the thickness of the surface varies, but not in SRIM-TRIM, so for the sake of comparison, the same approach was used for the two software. For sure, the SM3 samples represent an ideal sample, since it has a quite sharp transition between layers that is easier to simulate.

4. – Conclusion

This work aims to provide a first approach to the use of two types of Monte Carlo simulation software for interpretation of negative muon data. In both cases, simulation has provided good results in agreement with the experimental results, and it was possible to asses the thickness of the gold layer. Here, laboratory made samples were used, and some difficulties arose from the presence of uneven surface. However, Geant4/ARBY software provide more tools for the modelling of samples that can help overcome geometry issues. This means that the approach can be used in other situations, with objects of historical or artistic interest that do not present a well-defined layered structure as in this case. Finally, the results provide a first validation of the Geant4/ARBY, that can be further used as a tool for negative muon data interpretation.

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