Muonic atoms for fundamental and applied nuclear science

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On behalf of the MIXE, muX, and MONUMENT collaborations

Abstract

Muons are elementary particles that can orbit the atom as electrons do, but with a mass 207 times higher so that they have a Bohr radius 207 times smaller and x rays with energy 207 times higher. This allows to accurately measure those x rays, which can be applied for element identification (e.g., in heritage research) or to study nuclear-induced perturbations to extract absolute charge radii and spectroscopic quadrupole moments. Those observables may then be used for nuclear structure research or to support searches for physics beyond the standard model.

Moreover, as the muon approaches the nucleus it may be captured and induce inverse beta decay – the so-called ordinary muon capture process. The framework to determine the matrix elements involved in this process is the same as for double beta decay and can thus be used to constrain the different models applied in double beta decay investigations.

The Paul Scherrer Institute in Villigen (Switzerland) provides the highest intensity continuous muon source worldwide. It thus leads this research activity and gathers substantial interest from many collaborations. An upgrade of the muon source (IMPACT) for even higher intensity has recently been submitted for funding, which would ensure the leadership position and greatly enhance the capabilities in Europe.

Other critical aspects of this research include the germanium detection array, the production of targets (stable isotope enrichment, radioactive targets), and the theoretical framework at the frontier between atomic and nuclear physics.

Scientific context

Muons are elementary particles that can be simply described as heavy electrons, with ≈207 times the mass of their lighter cousin. The free muon has a half-life of 2.2 µs decaying back into an electron and two neutrinos. If captured by an atom, it displays quantized levels that are independent of the electronic levels, with a Bohr radius reduced respectively by the same factor 207 and consequently energies in the range of keV to MeV. Depending on energy, muons can penetrate deeply into matter and, once captured, emit x rays that are a fingerprint of the elements contained in that material. This technique can be used in a broad range of applications, from nuclear and particle physics to solid state physics and heritage science [Bis22].

This proximity of the muon orbital with the nucleus gives the muonic levels increased sensitivity to the perturbations induced by the nucleus on the atomic levels, such as the finite size correction arising from the charge distribution of the nucleus, or the nuclear electric quadrupole moment. This gives access to absolute observables such as the mean square charge radius $<r^2>$ or the spectroscopic quadrupole moment Q_S [Kne20, Ant20]. This technique has been broadly applied to study stable isotopes but the investigation of radioactive nuclei with this technique remains a challenge [Fri04].

The bound muon may decay while occupying an atomic orbital, or it may be captured by the nucleus, inducing inverse beta decay, transforming a bound proton into a neutron, producing radioactive isotopes. This ordinary muon capture process results in a reduction of the muon lifetime by opening a new channel for its disappearance. Furthermore, the matrix elements involved in this process can be determined from the same formalism as for the study of double beta decay. It generates a new set of experimental data to challenge the determination of those matrix elements, which is an important area of research within the nuclear physics community (e.g., $0\nu\beta\beta$ searches).

The study of muonic atoms is not new. However, many recent developments have led to renewed interest in their investigation. Fundamental symmetries of matter have given rise to investigations of light muonic atoms [Ant]. Moreover, the use of large-scale HPGe arrays instead of single detectors has substantially increased the sensitivity of the technique to detect the high-energy x rays, so that ever smaller samples can be investigated. Moreover, the application of the Ramsauer-Townsend effect has allowed a substantial increase in the capture efficiency and transfer of muons to ultra-small samples, down to 5 µg, opening the possibility to work with rare isotopes, whether naturally occurring but hard to enrich in large quantities, or even radioactive nuclei [Ada18].

Objectives

The study of medium to heavy muonic atoms spans many areas of nuclear physics research:

- Provide accurate charge radii to support the study of the fundamental forces and the search for physics beyond the standard model in the nuclear regime;
- Measure absolute shape observables ($<r^2>$, Q_S) of rare isotopes to complement the investigations along long isotopic chains performed by laser spectroscopy;
- Measure rates and cross sections that can be related to matrix elements of relevance in the investigation of (neutrino less) double beta decay;
- Support applications of nuclear techniques in applied research with muonic induced x-ray emission (MIXE).

Charge radii for physics beyond the standard model

The isotope ^{226}Ra has been identified as a candidate for atomic parity violation experiments [Nuñ13]. However, the interpretation of the data requires a measurement of the absolute charge radius $<r^2>$ to a precision of 0.2%. However, since radium is a radioactive element with no stable isotope, it has yet to be investigated by either muonic x-ray spectroscopy or electron scattering to determine its charge radius. With a sensitivity down to 5 µg of material, sufficiently large samples can be produced while maintaining the activity levels below the radioprotection tolerance within experimental areas of large-scale facilities. This enables the measurement of muonic x rays of ^{226}Ra to determine its absolute charge radius.

The charge radius is an observable involved in other searches of physics beyond the standard model. For example, V_{ud}remains the most determining matrix element in the verification of the unitarity of the Cabibbo-Kobayashi-Maskawa matrix. Its most accurate determination is established within the study of super-allowed beta decays, one of which is from the isomeric state ^{26m}Al [Ben19]. The different contributions to the error budget identify the precision on the charge radius as being a substantial contribution. The source of this uncertainty is mostly due to the propagation of multiple effects: the only absolute charge radius known for aluminium is that of ^{27}Al. The change in charge radius is then measured by laser spectroscopy between ^{26m}Al and ^{27}Al; however, the extraction of $\delta < r^2 >$ from the optical isotope shifts $\delta\nu$ requires knowledge of the atomic response to the nuclear size, for which the precision is limited. In that mass region, the reduced mass effects dominate, leaving a large relative uncertainty on the charge radius component. However, if measuring the charge radius $< r^2 >$ of ^{26m}Al, much higher precision may be reached.

Ground-state properties for nuclear structure

The study of ground-state properties such as nuclear charge radii and nuclear electric quadrupole moments has allowed to push our understanding of nuclear structure substantially in the last few years, across the entire nuclear landscape [Gar16, Mar18, deG20, Kos21, Ver22]. Those investigations rely on the precise determination of the nucleus-induced perturbations of atomic electronic levels. However, the extraction of the nuclear observables requires further input.

In particular, the measurement of the nuclear electric quadrupole moment Q_S comes from the study of the quadrupole component of the hyperfine structure, $B_{hfs} = Q_S V_{ZZ}$, where V_{ZZ} represents the electric field gradient generated by the electron at the site of the nucleus. That parameter may be determined to limited precision by large-scale atomic calculations. Alternatively, the measurement of several isotopes allows for a relative measurement, that then only depends upon the determination of the absolute moment from a reference isotope, e.g., by muonic x-ray spectroscopy [Ant20]: $Q_S = \frac{B_{hfs}}{B_{hfs,ref}} \cdot Q_{S,ref}$.

Furthermore, the determination of the changes in the charge radius $\delta < r^2 >$ from the optical isotope shift $\delta\nu$ arises from the relationship: $\delta\nu = \frac{A'-A}{AA'} K + F\delta < r^2 >$, where A and A' are the masses of the two isotopes being compared, and K and F are atomic parameters specific to the optical transition under investigation. The first part of this equation corresponds to the change in reduced mass from one isotope to the next, including the impact on the electron of interest as well as on the other electrons and their interactions. The second part is the response to the changes in the charge radius, and is only substantial for electrons which orbital have a non-vanishing overlap with the nucleus. While there are fluctuations between transitions, the global trend is that the reduced mass component scales with ${}^{1}/_{A^2}$ while the charge component scales with Z. The two components are roughly comparable in the tin ($Z = 50$) region, meaning that the reduced mass component dominates for lighter nuclei and that the size component is most important in the heavier systems.

The electronic parameters K and F can be determined to limited precision by large-scale atomic calculations. However, when multiple isotopes may be investigated by complementary techniques such as muonic x-ray spectroscopy or electron scattering, it is possible to benchmark the optical isotope shift using the King plot technique: $\frac{AA'}{A'-A}\delta\nu = K + F \cdot \frac{AA'}{A'-A}\delta < r^2 >$, where the plot of the modified isotope shift against the modified changes in charge radii should appear as a straight line with slope F and y-intercept K [Che12]. Fitting a line requires at least 2 points, while the changes in charge radius require a reference and a measurement. In total, three absolute radii are required to perform such an analysis. However, there are no odd-Z element with three stable isotopes. Moreover, the elements beyond bismuth ($Z = 83$) are all radioactive and cannot be studied with techniques for stable isotopes, besides thorium and uranium. Altogether, more than half of the elements do not have sufficient information to perform a King plot analysis.

With the advent of muonic x-ray spectroscopy of microscopic samples, it becomes possible to extend the study of absolute charge radii to rare isotopes near the valley of stability, through which a sufficient number of radii become available for a King plot analysis.

Studying matrix elements of relevance for double beta decay studies

Nuclear models aimed at the description of the nuclear matrix elements of $0\nu\beta\beta$ decays have traditionally been tested in connection with two-neutrino $\beta\beta$ ($2\nu\beta\beta$) decays and β decays. However, some time ago it was proposed that the ordinary muon capture (OMC) could also be used for this purpose [Kor02]. The $2\nu\beta\beta$ and β decays are low-momentum-exchange processes ($q \sim$ a few MeV), whereas both $0\nu\beta\beta$ and OMC are high-momentum-exchange processes ($q \sim 100$ MeV). In this way the $0\nu\beta\beta$ and OMC are similar processes and possess similar features: they are able to excite high-lying nuclear states with multipolarities J^π higher than $J^\pi = 1^+$. The $0\nu\beta\beta$ decay proceeds between the 0^+ ground states of parent and daughter even-even nuclei through virtual states of the intermediate odd-odd nucleus. These same virtual states can be accessed by the OMC from either the daughter nucleus (electron-emitting $\beta\beta$ decays) or the parent nucleus (positron-emitting/electron-capture (EC) $\beta\beta$ decays) [Zin19].

Applied research with muon-induced x-ray emission (MIXE)

Compared to protons, e.g., muons can penetrate quite deeply into a sample. Moreover, the depth at which they stop and interact with atoms is dependent upon the muon energy, so that it is possible to perform depth analysis of material composition using muon-induced x-ray emission [Bis22].

This approach is reminiscent of proton-induced x-ray emission (PIXE), which also allows for depth analysis. However, muonic x rays have much higher energy than electronic x rays, so that they are less suppressed by absorption within the sample itself, as well as better separated from one element to the next. This allows a much more accurate investigation of the sample composition.

Finally, and most important for heritage investigation, MIXE is a completely non-destructive technique that fully preserves the material.

Methodology

Muon source

Negative muons are produced at accelerator facilities from impacting high-energy protons on a graphite target, from which pions are emitted and decay in flight to muons. While most of the outcoming particles are π^+ that decay to μ^+, there is also a fraction of π^- that decays then to μ^-. Those possess a wide range of energies, from which the muon beamline selects a certain momentum range.

There are but a handful of muon sources in the world, the most intense being located at the Paul Scherrer Institute in Villigen (Switzerland). The protons are provided by the ring cyclotron accelerator (590 MeV, 2400 μA) to several facilities, one of which is used for the research presented here.

Muon capture

Macroscopic samples can be produced with sufficient thickness to stop and capture the muons. The particles may first be slowed down through a degrader and then stopped in a sample. Fine tuning of the capture depth may be achieved by selecting the momentum of the muons through the optics of the muon beamline.

Microscopic samples are more challenging to investigate as they are not thick enough to efficiently capture muons. Instead, a high-pressure gas filled with 100 bar hydrogen gas is used to efficiently stop and capture the muons. Those are transferred to deuteron (0.25% admixture), which are then further slowed and may travel unimpeded through the hydrogen thanks to the Ramsauer-Townsend effect. Upon reaching the back of the cell, the muon is then transferred to the microscopic sample deposited on a substrate. Samples as low as 5 µg have been successfully investigated [Ada18]. Combined with the radioprotection limit for the experimental area, this suggests that isotopes with half-lives above 20 years are within reach of this technique [Ada22].

Targets

Heritage investigations require samples (targets) that are of a size compatible with the facility. So far, samples such as coins and small items have been investigated. Larger samples would require arranging the detection array to match with the peculiarity of the sample.

Other, purely elemental investigations require nowadays enriched material of very high grade (99% and above) as they often relate to specific isotopes that are under investigation, and the muonic x-ray transitions of interest, such as the 2p-1s transition, feature substantial isotope shift. Muon capture studies are also concerned about working with specific isotopes, in particular to estimate the possible emissions of ternary particles in ordinary muon capture (e.g., if states above the particle emission threshold are populated). Those enriched targets are typically built on a light-element backing, like advanced forms of carbon (glassy carbon, pyrolithic graphite) to minimize the background in the muonic x-ray spectrum.

The enrichment and target production process employ the state-of-the-art techniques for target production, such as for actinide target production, or featuring combinations of radiochemistry and mass separation, such as featured for novel medical radioisotopes [PRISMAP]. As such, it has also been recently demonstrated that samples implanted at energies up to 100 keV into the substrate may still be investigated, hereby opening up a new range of possible isotope separation-implantation for muX.

Detection setups

The energy of the muonic x rays ranges from tens of keV to MeV, a range very similar to that of gamma-ray detection in nuclear physics. The detection arrays are thus very similar, consisting typically of HPGe detectors surrounding the sample. For example, in 2019, during the CERN Long Shutdown 2, PSI hosted MiniBall for a dedicated campaign on muonic x rays. Otherwise, the MIXE, MONUMENT, and muX collaborations at PSI operate an array composed of different kinds of germanium detectors, including LEGe, BEGe, and REGe.

In order to improve the signal to background ratio, triggers are also provided by plastic scintillators measuring the entering muons (coincidence) and by an array surrounding the sample for muons outside of the sample coverage or to identify Michel electrons arising from the decay of muons (anti-coincidence). Neutron scintillator detectors are also available to identify ternary particle emission.

The lightest elements feature x-ray emission below 20 keV. Such energies are out of range of HPGe detectors and require alternatives. Silicon drift detectors have been successfully tested at muX and the possibility to use microcalorimeters in the future is being investigated [Ant22].

Muonic cascades

The analysis of the data requires a thorough understanding of the experimental elements (synchronization of the plastic scintillators and x-ray detectors, detector response). However, once the muonic x-ray spectra can be produced and the lineshape is understood, the analysis requires a deep understanding of the atomic cascade that follows the capture of the muon.

The atomic transitions are dependent upon many factors, including the nuclear effects that are typically sought after. A framework with sufficient accuracy is thus required and is being developed [Ant20]. However, due to the comparable energy scale of the muonic atomic levels and nuclear levels, the problem cannot be tackled from a perturbation theory perspective and requires a direct interaction between atomic and nuclear theory. Such developments are being made at different centres across Europe to support the ongoing experimental work.

Readiness

In the last 5 years, the muX collaboration has demonstrated its ability to study muonic x rays at PSI. From a simple 2-detector setup in 2016 with macroscopic samples [Ant20], it has grown to a setup that exploits a large HPGe detector array for multiple collaborations covering research interests from physics beyond the standard model to nuclear structure and heritage science [Ada22, Bis22].

As such, it has shown that it can investigate the depth profile of unknown material for heritage science. The technique offers unique, non-destructive opportunities that are an added value to application outside of nuclear science [Bis22].

Nuclear ground-state properties ($<r^2>$, Q_S) and cross sections (OMC) can be determined accurately and with minimal to no dependence on nuclear models, providing crucial input for the extraction of observables from laser spectroscopy, as well as fundamental inputs for searches of BSM physics.

Expected challenges & recommendations

Like many experiments at large-scale facilities, the main challenges are **access and sensitivity**. The access to beam time at PSI is competitive and based on scientific excellence. Nuclear structure research is however not a core activity at PSI, which is more versed in particle physics, material science, and applications of nuclear research. It would thus be important to **recognize the benefit of muonic atoms for nuclear research**.

Concerning the sensitivity of the technique, this can be optimized from 3 perspectives:

- High muon beam intensities. This can be achieved thanks to the IMPACT upgrade that has recently been proposed at PSI. It is therefore essential that **the realization of IMPACT at PSI be endorsed by NuPECC**.
- The sensitivity can be enhanced by increasing the efficiency of the detector array. **Dedicated campaigns with advanced germanium arrays for muonic atoms** should be encouraged, as was performed in 2019 with MiniBall.
- Target production is a major technical challenge for muonic x-ray spectroscopy of radioactive samples: the production and separation of radioisotopes, and the making of radioactive targets by different techniques are key to the success of this programme. **Radiochemistry should thus be better integrated in the nuclear infrastructures to facilitate such activities**.

Finally, the analysis and interpretation of the data from muonic atoms requires a strong theoretical framework at the frontier between atomic and nuclear physics. **More collaborations between nuclear theory and atomic theory communities should be encouraged** to support this effort, through common events (workshops, trainings, …) and support for mobility of researchers.

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