Measurement of fundamental nuclear decay data using Metallic Magnetic Calorimeters

Publishable Summary for 17FUN02 MetroMMC Measurement of fundamental nuclear decay data using metallic magnetic calorimeters

Overview

The overall objective of this project is to improve the knowledge of electron capture (EC) decay and subsequent atomic relaxation processes. New theoretical calculation techniques and extensive experiments using specially adapted metallic magnetic calorimeters (MMCs) are being developed to determine important decay data which are relevant when studying the influence of EC decay in cancer therapy on the DNA level or the early history of the solar system as well as for primary activity standardisations in radionuclide metrology. The experimental parts are being complemented with a new approach based on microwave coupled resonators (MCRs).

Need

Determining the age of the solar system or how cancer treatments damage DNA are two research areas that both rely on very precise nuclear decay probabilities produced by EC during radioactive decay. Atomic data for EC decays have been derived from measurements and calculations performed more than 20 years ago, and these are now causing significant measurement problems. Compiled data, for example, are based on the frozen core approximation with no explicit description of multiple ionisation processes. Therefore, accurate experimental X-ray emission intensities are needed to establish consistent EC decay schemes and theoretical models of subsequent relaxation processes.

The precise knowledge of EC probabilities is pivotal when calculating the electron and photon emissions resulting from EC decay. The accurate knowledge of these emission spectra is a prerequisite for state-of-the-art liquid scintillation counting (LSC) techniques which are frequently used for primary activity determination in radionuclide metrology. The uncertainties of fractional EC probabilities define the resulting uncertainty of the activity determined by LSC, e.g. the triple-to-double coincidence ratio (TDCR) method. A sound improvement of the quality of LSC measurements therefore requires improved computation methods of emission spectra which, in turn, can only be developed based on new theoretical approaches and experimentally determined EC probabilities of the highest achievable accuracy. In addition, X-ray emission intensities are key data used to quantify the activity of a radioactive material by X-ray spectrometry.

Some of the technical developments were carried out within the EMPIR project 15SIB10 MetroBeta for pure beta-emitting isotopes with endpoint energies in the range from 70 keV to 700 keV. To study EC decays and X-ray emissions, new developments for MMC-based techniques are currently being carried out for high precision measurements with both internal and external sources.

Objectives

The specific objectives of the project are:

1. To improve experimental techniques for spectrometry using novel cryogenic detectors based on MMCs and MCRs for radionuclide metrology in the energy range of 20 eV - 100 keV.
2. To determine fractional EC probabilities of selected radionuclides by means of spectrometry based on novel cryogenic detectors with high energy resolution and very low energy threshold using sources embedded in the detector absorber.

3. To measure absolute X-ray emission intensities of selected radionuclides by using a combination of high-resolution spectrometry based on novel cryogenic detectors using external sources and accurate primary activity determination.
4. To improve theoretical models and *ab initio* calculations of the EC process and subsequent atomic relaxation and to validate them with the high-precision experimental data from this project.
5. To facilitate the take-up of the technology and measurement infrastructure developed in the project by radionuclide metrologists, nuclear physicists and other researchers.

Progress beyond the state of the art

Spectrometry by means of energy dispersive MMC detectors has been applied to radionuclide metrology comparatively recently. However, this technique has been demonstrated to be particularly suitable for measurements of radioactive decay emission spectra in the energy range from a few eV up to hundreds of keV with very high energy resolution. The measurement of fractional EC probabilities requires the further development of MMCs with significantly reduced low energy threshold, of order 20 eV.

MMC-based detectors have been successfully applied to high-resolution spectrometry of X-rays and gamma-rays with energies above 10 keV, typically emitted from radionuclides with high atomic number. The excellent energy resolution allows X-ray satellites (due to multiple-ionised states) to be distinguished; these are invisible with conventional spectrometers. The measurement of absolute X-ray emission intensities of nuclides with low atomic number requires the energy range to be extended down to 1 keV with very high detection efficiency.

Prior to the start of this project, methods to calculate EC probabilities were mainly established for allowed EC transitions. These calculations suffered from significant uncertainties. Improvements of the computation methods are now being realised based on more accurate experimental data. However, accurate experimental data were scarce and obtained from indirect methods. Emission spectra of Auger electrons and X-rays are highly influenced by electron correlation leading to multiple ionisation of the atom as well as shake-up and shake-off effects. Theoretical treatment of these multiple excitations has been developed for the understanding of core level X-ray excitations in the field of solid-state physics but was only very recently being transferred to the calculation of EC spectrometry.

Results

Experimental techniques for spectrometry based on novel cryogenic detectors for radionuclide metrology in the energy range of 20 eV – 100 keV

New source-absorber-detector designs are required for the measurement of fractional EC probabilities and are being developed. The MMC-systems for high-resolution radioactive decay emission spectrometry currently in operation at the three participating National Metrology Institutes CEA, PTB and KRISS are being improved to achieve a lower energy threshold in the order of 20 eV and better energy resolution.

The designs of novel MMC sensors for EC spectrometry and MMC detector arrays for X-ray spectrometry have been defined. The layout of both detector types, i.e. the detectors for EC spectrometry using internal sources and MMC detector arrays for measuring absolute X-ray intensities was determined. Two wafers containing detectors for EC spectrometry and two wafers containing MMC detectors for measuring absolute X-ray intensities have been fabricated at UHEI-KIP and delivered to PTB for wafer-scale characterization of resistances at room temperature. These measurements showed that the yield of the fabrication run is rather high. Detector properties at T = 4.2 K are as good as expected. The characterization at very low temperatures (< 20 mK) is ongoing. Preliminary results confirm the high quality of the fabricated detectors with a very high signal to noise ratio. The EC spectrometry detectors have been used in several measurements for EC spectrometry and have shown to work as anticipated. In order to perform a calibration of MMC gain stability and linearity by means of applying Joule heat pulses directly to the detector, a sufficiently fast current pulse source has been identified. Its pulse duration and amplitude linearity have been characterized and found to meet the requirements.

Setups for absorber/EC-source preparations by means of picoliter drop dispensing and subsequent diffusion welding have been successfully put into operation. Tests of laser surface ablation have continued on Au foils. Different spot patterns were produced. Subsequent micro-fluidic drop depositions of non-radioactive carrier solution showed hydrophobic character of the laser-treated Au surfaces. Crystallite sizes have been assessed by means of Scanning Electron Microscopy. It was found that sizes of the crystallites formed on the laser-treated Au surfaces remained in the range of a few micrometres.

An X-ray detector is running, and it works well. 6/8 pixels are recording. The decay constants are as expected. The sensitivity is about 15 mϕ_0/keV @ 15 mK and 60 mA (a bit lower than expected). The different pixel pulse shapes and sizes look very homogenous.

A microwave coupled resonator system has been modelled and designed, based on a 12 mm diameter single crystal sapphire resonator, coupled to a 1 mm diameter perovskite single crystal $SrTiO_3$. Both the absorption characteristics for X-rays and the thermal characteristics of the system have been modelled. Construction of the cooler system and thermal characterization system is completed, and we have demonstrated a cooler base temperature of 2.2 K. Further work on another perovskite, $CaTiO_3$ single crystal puck was carried out which showed that below 20 K operating temperature the thermal time constant is below 1 ms. The heat capacity of the puck was also shown to have a strong T^3 dependence on temperature down to below 10 K. Progress has been good and all aspects of the novel calorimeter performance have been demonstrated, including highly successful demonstration of the required rapid thermal time constant of a coupled resonator, very high *df/dT* response and promising development of low noise microwave detection system.

Determine fractional EC probabilities of selected radionuclides by means of spectrometry based on novel cryogenic detectors with high energy resolution and very low energy threshold using sources embedded in the detector absorber

When the source is embedded in the absorber for measuring fractional EC probabilities, new techniques are required to prepare sources/absorbers taking the thermal coupling and hence the chemical form (e.g., crystal structure and size) into account. In particular techniques aiming at very fine dispersion of the radioactive material within the absorber, required for complete thermalisation of the particle energy, are being addressed.

Five of six radionuclide solutions have been acquired, characterized for non-radioactive properties and the activities of three radionuclide solutions have been measured traceable to primary standardisations. All relevant decay data (energies and probabilities of emitted radiations, half-lives) and chemical properties of each of the 6 selected radionuclides have been compiled. Based on these data the absorber dimensioning by Monte Carlo simulation has been completed as far as the absorber thickness is concerned. Manganese-54 sources have been prepared by means of an automated micro-fluidic drop dispensing apparatus on gold/nanoporous gold (np-Au) samples and sandwiched with gold foils to form Au/np-Au/Au absorbers enclosing the ^{54}Mn activity.

A first spectrum of the ^{65}Zn decay has been measured using an MMC designed for the predecessor project MetroBeta. The detector behaved according to the specifications, while the shape of the spectrum shape was deformed, likely caused by the source material prepared from liquid solution.

Monte Carlo (MC) simulations have been performed for an external ^{57}Co calibration source. The results show small numbers of scattering events on the absorber and some secondary particle generation in the Pb collimator, which are in good agreement with measured calibration data. Thus, the simulations can be used to improve the energy calibration of the MMC, by correcting for these slight artefacts. Two Au/np-Au/Au absorbers containing ^{54}Mn sources have been measured using an MMC performing rather well in terms of energy resolution. However, a continuous background is present in the spectrum underneath and between the EC peaks. Extraction of the fractional EC probabilities requires quantification of the Compton component of the background via detailed MC simulation. First simplified simulations have been completed; more detailed simulations will be performed shortly.

Method to determine absolute X-ray emission intensities of selected radionuclides by using a combination of high-resolution spectrometry based on novel cryogenic detectors using external sources and accurate primary activity determination

The MMC based measurement of absolute X-ray emission intensities of nuclides with low atomic number requires the energy range to be extended down to 1 keV with very high detection efficiency.

The activity limits are defined for each radionuclide to be electroplated or dropped deposited on stainless steel. The source activities are limited by the counting statistics and the count rate capability of the 8-pixel array MMC. The activities of the dropped deposited sources are determined by weighing with uncertainties less than 0.5 % traceable to primary activity standardization. Also defined are the dimensions of the source support and the active area to be compatible with the electroplating set-ups. The new CEA detector module for the 8-pixel array was designed to integrate the sampler exchanger, the magnetic deflector and the MMCs. After production the complete module was assembled with the motor, the sample exchanger, the SQUDs and the 8-pixels MMC array. The module was cooled down without problem at 11.8 mK. The X-ray emission intensities of ^{109}Cd were measured using the MMC SMX3 with an energy resolution of 22 eV.

The principle of operation of this new Microwave Coupled Resonator calorimeter system was demonstrated by the first detection of single particle decays from a very low activity ^{241}Am radioactive source, installed in the Microwave Coupled Resonator detector. The source activity is only 200 Bq so the count rate (allowing for the solid angle of the detector at the source) is only ~ 1 detection every 4 seconds. This first detection is an important proof of principal, but much further experimental and data analysis work is needed to reduce the noise and improve sensitivity.

Theoretical models and ab initio calculations of the EC process and subsequent atomic relaxation and validation with the high-precision experimental data

One of the aims of this project is to improve the theoretical predictions of EC decay and subsequent atomic relaxation processes at the percent precision level.

The new version of the BetaShape code has been used for several studies. In addition, realistic nuclear structure was included in EC decay modelling. Nuclear structure calculations have been performed with the NushellX code, which provides one-body transition densities. The construction of the code is now finalized. Comparison of relevant atomic parameters computed using this program and the MCDF data is underway. The partners succeeded to couple their codes in practice, but additional tests still need to be performed for complete validation. For this purpose, CNRS code produced accurate atomic wave functions that have been made compatible with the formalism of EC decay employed by another partner. The BetaShape code was deeply modified to make it compatible with the use of external atomic wave functions that can be calculated according to different atomic configurations. Preliminary capture probabilities have been calculated for ^{7}Be, ^{41}Ca, ^{54}Mn, ^{55}Fe, ^{109}Cd, ^{125}I and ^{138}La. Results are being analyzed.

The electron configurations of the radionuclides of interest have been analysed and the input files needed for the calculation of wave functions and atomic parameters have been defined. Calculations of the EC spectrum for several EC isotopes have been performed including multiplet splitting of the different main resonances and Auger transitions to different bound electron configurations. Energy dependent life-time calculations of these states due to Auger and Fluorescence decay has been added. For cases where experimental data is available satisfactory agreement between theory and experiment is reached over the entire energy range or the EC spectrum. These calculations are implemented in Quanty, a freely available script language (www.quanty.org) and can easily be modified to test the influence of for example different nuclear models. Documentation on the web page has been updated to include the newly generated functions used in these scripts. Implementations for the fluoresence yield line widths have been implemented and are tested against different know experimental measurements.

Impact

By now the project's results were disseminated in different ways. Stakeholders were identified and invited to join the first Stakeholder workshop in Saclay in October 2019. A second workshop was organised in Heidelberg in October 2020 for all those interested in EC spectra calculations using the Quanty program. On eight occasions the project and its results were presented to European and International regulatory bodies. A broader scientific community was the target audience of nine articles in peer-reviewed journals, 13 presentations and poster presentations held at national and international conferences and 14 additional presentations held at other external events. News and events as well as links to publications can be found on the project website http://empir.npl.co.uk/metrommc/.

Impact on industrial and other user communities

The project will help to reduce uncertainties of nuclear decay data and to obtain activity standards with improved accuracy which are required for industrial applications. In some cases (e.g., ^{41}Ca), the reduced uncertainties of EC probabilities will lead to a considerable reduction of the uncertainty of the corresponding half-life which is relevant for radioactive waste management. Improved knowledge of the emission probabilities of Auger electrons and X-rays at each energy level is very important for EC nuclides used in nuclear medicine since the estimation of the administered dose greatly depends on these data.

Impact on the metrology and scientific communities

Experimentally determined EC probabilities and X-ray emission intensities will lead to improvements of theoretical calculation methods. The measured data and improved calculation methods will be an invaluable contribution to the realisation of the SI unit becquerel in radionuclide metrology. Radionuclide metrologists will be enabled to reduce uncertainties, which is important for several other fields where precise radioactivity measurements matter. This comprises geo- and cosmochronology, nuclear medicine as well as industrial applications, but also research in other fields. The improved calculation techniques of the EC process, and its subsequent atomic relaxation are essential for a sound research of radiation effects in human tissue on the DNA level. Improved calculation methods have already been presented to the "nuclear decay data evaluation community". Computed fractional EC probabilities of ^{55}Fe obtained from the BetaShape program were recommended by the Key Comparison Working Group (KCWG) of the CCRI(II) to be used by all participants of a key comparison on ^{55}Fe, which has high influence on the determined activity. As a result, the project is already having a significant impact on the radionuclide metrology community.

The developments will also contribute to new basic research experiments which require measurements of ionizing radiation with high energy resolution. As an example, short baseline neutrino oscillation experiments at nuclear reactors would benefit from accurate EC probability measurements for the indispensable evaluation of background sources.

Beyond the direct impact from the measurements on EC decaying nuclides, the advances in MMC and related readout techniques triggered by this project will be highly beneficial in numerous fields of applied and fundamental research in which MMCs play an increasingly important role. Some of the developments will also be applicable to other types of cryogenic detectors, thus reaching even more fields of research and further extending the outreach of the project.

Impact on relevant standards

The project will lead to improved nuclear decay data by direct measurements and by improving the theoretical calculation techniques. Hence, the outcome of this project will be a valuable contribution for nuclear decay data evaluations.

The standard of the SI unit becquerel must be established for each radionuclide individually and generally consists in the standard method used for the activity standardisation of the respective nuclide. For several pure EC nuclides, the TDCR-LSC method is the standard method. A better knowledge of the electron and photon emission spectra will have immediate impact on the activity standards for EC decaying nuclides.

Longer-term economic, social and environmental impacts

This project will accelerate innovation and competitiveness in the field of the ground-breaking technology using MMCs and more generally cryogenic detectors. On a long-term perspective, MMC-based detectors may become a tool for enhanced nuclear spectrometry with an energy resolution which is much higher than with any semi-conductor detector. In particular, spectrometry at very low energy, where the detection efficiency of conventional techniques drastically drops off, benefits from the outstanding low energy threshold of MMCs, enabling substantial reduction of systematic effects. All in all, MMC detectors enable research and applications far beyond current limits which are, at present, defined by existing spectrometers based on semi-conductors. Due to the high potential of MMCs, it is anticipated that the technology will be more and more used in various disciplines. The nuclear decay data which will be determined with better precision within this project and beyond will be important in many fields such as nuclear medicine, industry or geo- and cosmochronology.

List of publications

1. Mougeot, X.: Towards high-precision calculation of electron capture decays. In: Applied Radiation and Isotopes 154 (2019), 108884, https://doi.org/10.1016/j.apradiso.2019.108884
2. Martins, L. et al.: Multiconfiguration Dirac-Fock calculations of Zn K-shell radiative and non radiative transitions. In: X-Ray Spectrometry 49, Issue 1 (2019), 192 – 199, https://doi.org/10.1002/xrs.3089

3. Martins, L. et al.: Overview and calculation of X-ray K-shell transition yields for comprehensive data libraries. In: X-Ray Spectrometry (2020), https://doi.org/10.1002/xrs.3123
4. Paulsen, M. et al.: Development of a Beta spectrometry setup using metallic magnetic calorimeters. In: Journal of Instrumentation 14 (2019), P08012, https://doi.org/10.1088/1748-0221/14/08/P08012
5. Bockhorn, L. et al.: Improved Source/ Absorber Preparation for Radionuclide Spectrometry Based on Low-Temperature Calorimetric Detectors. In: Journal of Low Temperature Physics (2019), https://doi.org/10.1007/s10909-019-02274-8
6. Fretwell, S. et al.: Direct Measurement of the ^{7}Be L/K Ratio in Ta-Based Superconducting Tunnel Junctions. In: Physical Review Letters 125, 032701 (2020), https://doi.org/10.1103/PhysRevLett.125.032701 and https://arxiv.org/abs/2003.04921v2
7. Ranitzsch, P. C.-O.: MetroMMC: Electron-Capture Spectrometry with Cryogenic Calorimeters for Science and Technology. In: Journal of Low Temperature Physics (2019), https://doi.org/10.1007/s10909-019-02278-4
8. Braß, M., Haverkort, M.W.: Ab initio calculation of the electron capture spectrum of Ho-163: Auger–Meitner decay into continuum states. In: New Journal of Physics 22, 093018 (2020), https://doi.org/10.1088/1367-2630/abac72

This list is also available here: https://www.euramet.org/repository/research-publications-repository-link/

<table>
<tr><td colspan="2">Project start date and duration:</td><td>June 2018, 36 months + 6 months</td></tr>
<tr><td colspan="3">Coordinator:
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Project website address: http://empir.npl.co.uk/metrommc/</td></tr>
<tr><td>Internal Funded Partners:
1 PTB, Germany
2 CEA, France
3 NPL, United Kingdom</td><td>External Funded Partners:
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