Measurement of fundamental nuclear decay data using Metallic Magnetic Calorimeters

Publishable Summary for 17FUN02 MetroMMC
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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, 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 has been defined. Setups for absorber/EC-source preparations by means of picoliter drop dispension and subsequent diffusion welding have been successfully put into operation.

A novel cryogenic calorimetry technique based on a weakly-coupled microwave resonator detector is being developed within the project. This concept has been demonstrated to exhibit a very high temperature resolution. The adaptation of the resonator system to enable particle absorption and energy dispersive detection are being conducted and its performance investigated.

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. Both the absorption characteristics for X-rays and the thermal characteristics of the system have been modelled. Construction of the cooler system and thermal characterisation system is underway, and we have demonstrated a cooler base temperature of 2.2 K.

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.

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. The source activities are limited by the counting statistics and the count rate capability of the 8-pixel array MMC. Also defined are the dimensions of the source support and the active area to be compatible with the electroplating set-ups. Furthermore, the reconfiguration of the CEA setup for the multi-pixel-X-ray spectrometry has started. In this context, layout options for the MMC X-ray sensor chips are under consideration. The MCR system will require dried drop samples of activity on low microwave-loss substrates with dimensions compatible with perovskite absorber.

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 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.

Impact

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.

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

There are no publications at this early stage of the project.

Project start date and duration:	June 2018, 36 months	
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