Reviewer Report

on the thesis ***"Studies on the applications of the Triple-to-Double Coincidences Ratio method for primary activity standardization using liquid scintillation counting"*** submitted by **Mr. Chavdar Dutsov**, for awarding the educational and scientific degree Doctor (PhD) in Physics (Nuclear Physics)

by

Prof. Georgi Rainovski, DSc., member of the scientific jury

Overview of the submitted documents

The candidate has presented a thesis (in English), extended abstracts (in English and in Bulgarian) and a list of publications. The candidate also has presented all necessary documents required by the law and the internal regulations at Sofia University for awarding a PhD degree. The candidate has declared that the presented thesis is based on original scientific investigation and this statement is supported by an originality check report from Assoc. Prof. Krasimir Mitev, scientific supervisor of the candidate.

Biographical data of the candidate

The candidate obtained a bachelor's and a master's degrees in Physics (Medical physics) from Sofia University, Faculty of Physics, Department of Atomic Physics in 2016 and 2018, respectively. In 2018, the candidate was enrolled in the doctoral program "Nuclear physics" at Faculty of Physics, University of Sofia in the group of Assoc. Prof. Krasimir Mitev. The candidate successfully completed the program in 2021. In the course of the program, the candidate participated in several scientific projects funded by the Bulgarian National Science Fund, the Sofia University Science Fund, and the European Metrology Program for Innovation and Research. In the period 2019 to 2021, the candidate had two extended research visits at LNHB, CEA Saclay, France. From 2018 up to now, the candidate works as a physicist at the Metrology of Ionizing Radiation laboratory, Department of Atomic Physics at Faculty of Physics, Sofia University "St. Kliment Ohridski".

Overview and scientific achievements of the thesis

The thesis consists of three parts which encompassed twelve chapters. The first part (Chapters 1 and 2) is an extensive overview of liquid scintillation counting (LSC) and its applications. The process of forming the scintillation signal is described in detail along with its time and energy properties. Photomultipliers used in LSC systems are also briefly described. In chapter 2, the free parameter model is introduced in detail and its applications for calculating the detector efficiency are demonstrated. The triple-to-double coincidences ratio (TDCR) method is extensively described as an application of the free parameter model for absolute activity determination. Analogously, the Compton coincidences method is introduced as another application of the free parameter model. The first part of the thesis contains no original scientific results, but it is of importance since it sets the ground for the follow up studies and demonstrates that the candidate has learned the experimental techniques and acquired the necessary knowledge and understanding to carry out the foreseen research. It is also worth to

mention that the first part is built on an extensive bibliographical study containing 99 references out of total 153 used in the thesis.

The second part of thesis is devoted to the study of the time distribution of light emission. The candidate has developed a software for recording and analyzing LS events (Chapter 4). The software has three modes of operation allowing for implementation different counting algorithms. The software was validated using simulate and real experimental data. The performances of two counting algorithms were compared on simulated and real data sets obtained under different conditions. The results of this comparison ware published in "Applied Radiation and Isotopes". An extensive study of the distribution of the time intervals between signals from two photomultipliers, the so called cross-correlation distribution, is presented in Chapter 5. Analytical expression for the cross-correlation distribution of prompt scintillation events was derived. In Chapter 6, it is shown that this expression can be used to obtain the detection efficiency of a two-photomultiplier system and the mean number of detected photons. These results comprise one of the major scientific contributions of the thesis and are published in "Scientific Reports", Nature Research.

The third part of the thesis is devoted to various applications of the time distribution measurements in metrology. This part is based on the results from several original scientific studies conducted by the candidate. In the first study the candidate has re-measured the lifetimes of excited states of ^{57}Fe and ^{237}Np by LS-LS and LS-γ techniques (Chapter 6). The lifetimes are extracted by analyzing the decay curves associated with the states of interest, the so-called slope method. The obtained results are quite precise and, in overall, in agreement with the adopted values, demonstrating the applicability of the LS counting for lifetime measurements. The results from this study are published in "Nuclear Inst. and Methods in Physics Research, Section A". On this study, however, I have one criticism and one question. Both are related to the extraction of the lifetime of the 136.5 keV level in ^{57}Fe. First, the statement on page 106, namely *"The half-life of this level is too short to be studied with the gamma detector due to its timing resolution."* as a general statement is not correct. There are many studies in which sub-nanosecond lifetimes are measured with HpGe detectors in similar setups (for example see W. Andrejtscheff *et al.*, Nucl. Instrum. Methods 204, 123 (1982)). The methods for extracting such short lifetimes seem also applicable in the analysis of LS–γ data. The question is the following:

> If the lifetimes of the first and second excited states of ^{57}Fe were comparable in values, would the logic for selecting the proper LS-LS coincidences and the method for extracting the lifetime of the 136.5 keV level in ^{57}Fe be the same as the those described in the thesis? In other words, in the described situation would it be possible to extract the lifetime of the second excited state of ^{57}Fe?

In Chapter 8 the time distribution measurements are used to develop an experimental and theoretical methods for evaluating and correcting for accidental coincidences in TDCR systems. The methods were validated by using simulated and real experimental data. It is shown that the accidental coincidences corrections will improve the non-linearity of TDCR systems used for studies of short half-life medical nuclides like ^{11}C, ^{15}O or ^{18}F and are of particular interest in measurements of sources with high activities. These results represent the second major scientific contributions of the thesis and are published in "Nuclear Inst. and Methods in Physics Research, Section A".

In Chapter 9 the influence of the coincidence window on the counting rate in the TDCR measurements is studied. More precisely, the investigated problem is related to the choice of optimal coincidence window which should allow for recording most of the emitted light and for minimizing the rate of accidental coincidences and adverse effects due to delayed light emission. Practical approaches for optimizing the coincidence window are suggested. These results are published in “Nuclear Inst. and Methods in Physics Research”.

Chapter 10 is devoted to the characterization and the first usages of a new Compton TDCR detector. The candidate has specific contributions in the software development for the data acquisition system of the detector and in setting up and preforming the first measurements with the detector. The technical specifications of the system are given in detail and the results from several measurements are presented.

Chapter 11 presents results from the validation of the TDCR system developed and operated at Sofia University. This is done through a participation in an international comparison of ^{3}Hin water activity measurements organized by the BIPM and in two international comparisons of activity measurements of ^{222}Rn in water. The results are in excellent agreement with the ones form other laboratories and confirm that the TDCR system at Sofia University allows for accurate measurements which are recognized at international level. Some of the results obtained in this chapter are published in “Metrologia”.

Chapter 12 summarizes the research and the main results obtained in the thesis. The scientific contributions are clearly defined and divided into two groups, namely contributions to new knowledge in the field of activity measurements using LS counting and contributions to methodical aspects in the field of LS counting. In my opinion, most of these contributions are indeed novel and scientifically significant.

The thesis is well structured and written. It contains a lot of technical details, and all measurements are strictly documented. The thesis reads fairly easy and the main idea, namely that the timing aspects of the LS counting offer new opportunities in the field of metrological activity measurements, can be followed form the beginning to the end. The extended abstracts correctly and sufficiently reflect the content of the thesis.

Scientific publications on the topic of the thesis

The thesis is based on nine publications in international peer-reviewed journals. This includes three publications in *Nuclear Instruments and Methods in Physics Research, Section A: accelerators, Spectrometers, Detectors and Associated Equipment* (IF 1.455, SJR 0.75, Q1), two in *Applied Radiation and Isotopes* (IF 1.513, SJR 0.48, Q2), one in *Metrologia* (IF 3.157, SJR 0.64, Q1), one in *Journal of Luminescence* (IF 3.599, SJR 0.65, Q2), one in *International Journal of Environmental Research and Public Health* (IF 3.390, SJR 0.75, Q2), and one in *Scientific Reports, Nature Research* (IF 4.379, SJR 1.24, Q1). In five of these publications the candidate is either first author or a corresponding author, including the publication in Scientific Reports, Nature Research. This leaves no doubt that the candidate has leading or significant personal contribution in all publications. The number of publications, the quality of the journals in which they are published, and the apparent personal contributions of the candidate meet and significantly exceed the requirements in the internal regulation of Sofia University for awarding PhD degree in Physics.

Conclusions

After reviewing the submitted thesis, the extended abstracts, and the scientific papers on which the thesis is built, I confirm that the thesis complies with all requirements of the Law on the Development of Academic Staff in the Republic of Bulgaria and all additional criteria in the internal regulations of Sofia University for the awarding of scientific and educational degree PhD in Physics. My personal impressions from Mr. Dutsov as a promising young scientist were fully confirmed in the course of reviewing his thesis. The number and the quality of the publications, he has presented, are impressive and well exceeds all requirements. Therefore, I stand fully convinced in the scientific merits of the candidate and his thesis and strongly recommend the scientific jury to award Mr. Chavdar Dutsov a PhD degree in Physics.

Sofia

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