Appendix No.2 to the Lukasz Swiderski application for initiation of the habilitation procedure

### 1. Name and Surname.

Lukasz Swiderski

## 2. Diplomas and scientific degrees.

Master of Science:

University of Warsaw, Faculty of Physics, 2002.

Title of the thesis: "The study of fusion barrier distribution by means of a cyclotron." supervisor: dr. hab. Ernest Piasecki

Doctor of Physics:

University of Warsaw, Faculty of Physics, 2005.

Title of the dissertation: "The study of fusion barrier distribution in reactions of Ne and Sn as well as Ne and Ni nuclei." supervisor: dr. hab. Ernest Piasecki

### 3. Information about the research career.

- 2016 Head of Radiation Detectors Division, National Centre for Nuclear Research (NCBJ), Otwock-Swierk
- 2013 2014 Head of Radiation Detectors Division, National Centre for Nuclear Research (NCBJ), Otwock-Swierk
- 2012 Adjunct in National Centre for Nuclear Research (NCBJ), Otwock-Swierk
- 2006 2011 Adjunct in Soltan Institute for Nuclear Studies (IPJ), Otwock-Swierk
- 2002 2005 PhD studies at the Faculty of Physics, University of Warsaw

## 4. The achievement justifying the application for habilitation, as defined in Art. 16, Par. 2 of the Act of 14 March 2003 on Academic Degrees and Academic Title and on Degrees and Title in Arts:

## a) title of the scientific achievement,

"The development of detection techniques in order to study the relation between the nonproportionality of response, the energy resolution of scintillators and the scintillation decay time."

## b) list of publications, that constitute the basis for the habilitation procedure

[A1] Ł. Świderski, M. Moszyński, W. Czarnacki, A. Syntfeld-Każuch, M. Gierlik, Nonproportionality and energy resolution of Nal(Tl) at wide temperature range (-40°C to +23°C), IEEE Trans. Nucl. Sci. Vol. 54 No 4 (2007) 1372.

(contribution: 60%, carrying out measurements, data analysis, discussion of results, writing publication)

- [A2] Ł. Świderski, M. Moszyński, A. Nassalski, A. Syntfeld-Każuch, T. Szczęśniak, K. Kamada, K. Tsutsumi, Y. Usuki, T. Yanagida, A. Yoshikawa, Light yield non-proportionality and energy resolution of praseodymium doped LuAG scintillator, IEEE Trans. Nucl. Sci. Vol. 56 No 3 (2009) 934.
  (contribution: 50%, carrying out measurements, data analysis, discussion of results, writing publication)
- [A3] Ł. Świderski, M. Moszyński, A. Nassalski, A. Syntfeld-Każuch, T. Szczęśniak, K. Kamada, T. Tsutsumi, Y. Usuki, T. Yanagida, A. Yoshikawa, W. Chewpraditkul, M. Pomorski, Scintillation properties of praseodymium doped LuAG scintillator compared to cerium doped LuAG, LSO and LaBr<sub>3</sub>, IEEE Trans. Nucl. Sci. Vol. 56 No 4 (2009) 2499. (contribution: 40%, carrying out measurements, data analysis, discussion of results, writing publication)
- [A4] Ł. Świderski, M. Moszyński, W. Czarnacki, J. Iwanowska, A. Syntfeld-Każuch, T. Szczęśniak, G. Pausch, C. Plettner, K. Roemer, Measurement of Compton edge position in low-z scintillators, Radiat. Meas. Vol. 45 (2010) 605. (contribution: 60%, construction of experimental setup, carrying out measurements, data analysis, discussion of results, writing publication)
- [A5] Ł. Świderski, M. Moszyński, W. Czarnacki, A. Syntfeld-Każuch, T. Szczęśniak, R. Marcinkowski, G. Pausch, C. Plettner, K. Roemer, Energy resolution of Compton electrons in LaBr<sub>3</sub>:Ce scintillator, IEEE Trans. Nucl. Sci. Vol. 57 No 3 (2010) 1697. (contribution: 50%, construction and optimization of experimental setup, carrying out measurements, data analysis, discussion of results, writing publication)
- [A6] Ł. Świderski, R. Marcinkowski, M. Moszyński, W. Czarnacki, M. Szawłowski, T. Szczęśniak, G. Pausch, C. Plettner, K. Roemer, Electron response of some low-Z scintillators in wide energy range, J. Instrum. Vol. 7 (2012) P06011. (contribution: 60%, construction of experimental setup, carrying out measurements, data analysis, discussion of results, writing publication)
- [A7] Ł. Świderski, R. Marcinkowski, M. Szawłowski, M. Moszyński, W. Czarnacki, A. Syntfeld-Każuch, T. Szczęśniak, G. Pausch, C. Plettner, K. Roemer, Non-proportionality of electron response and energy resolution of Compton electrons in scintillators, IEEE Trans. Nucl. Sci. Vol. 59 No 1 (2012) 222.
  (contribution: 40%, construction and optimization of experimental setup, carrying out measurements, data analysis, discussion of results, writing publication)
- [A8] Ł. Świderski, M. Moszyński, W. Czarnacki, M. Szawłowski, T. Szczęśniak, G. Pausch,
  C. Plettner, K. Roemer, P. Schotanus, *Response of doped alkali iodides measured with*

gamma-ray absorption and Compton electrons, Nucl. Instrum. Meth. A Vol. 705 (2013) 42.

(contribution: 65%, construction and optimization of experimental setup, carrying out measurements, data analysis, discussion of results, writing publication)

- [A9] Ł. Świderski, M. Moszyński, A. Syntfeld-Każuch, M. Szawłowski, T. Szczęśniak, Measuring the scintillation decay time for different energy depositions in NaI:TI, LSO:Ce and CeBr<sub>3</sub> scintillators, Nucl. Instrum. Meth. A Vol. 749 (2014) 68. (contribution: 70%, construction and optimization of experimental setup, carrying out measurements, data analysis, discussion of results, writing publication)
  - c) the report on the scientific aim of the mentioned above publications and results along with the discussion of an eventual application.

## 4.1. Introduction

One of my main research activity is the development of experimental techniques that allow for characterization of basic properties of scintillators. Scintillators are a class of materials that can be used for detection of ionization radiation. Absorption of the energy carried by particle or photon results in excitation of atomic states in scintillator and is followed by deexcitation of states, e.g. by emission of visible, ultraviolet or infrared light. The light generated during this process can be detected by a photodetector characterized with sufficient sensitivity in the range matching the emission spectrum of the scintillator. As a result, the signal induced in the photodetector can serve for recording the scintillator response to the radiation that it is exposed to.

Scintillators may be grown in a form of various substances, that may take on different states of matter, i.e. solids, liquids or gas. Scintillators may be amorphous (plastics, ceramics, powders) or have crystalline structure. The common feature of most of the scintillators is that they can be grown in large sizes, up to several or even tens of centimeters. It is their significant advantage over semiconductor detectors, which usually have dimensions in the range between fraction of a millimeter and few centimeters. In case of large volume semiconductor detectors like germanium, cooling down to low temperatures, close to boiling point of liquid nitrogen, is required.

The possibility of growing scintillators in a form of large pieces makes them the most commonly used detectors for high energy gamma radiation. Taking into account features like detection efficiency, cost of fabrication and technical requirements for operation, scintillators are the best choice detectors for quanta of energy above 100 keV. However, this does not mean that they cannot be used for detection of charged particles, like the electrons, protons, heavy ions and, indirectly – neutrons. Wide scope of available scintillating materials makes them play an important role in various fields of science and technology, like: high energy physics (calorimeters) and nuclear reaction physics experiments (spectrometers), medicine (e.g. computed tomography, positron emission tomography, magnetic resonance imaging,

dosimetry), cargo and personal inspection (scanners at airports or trans-shipment centers) or various sectors of industry (e.g. mining).

After the defense of my PhD thesis on probing the fusion barriers distributions in nuclear reactions, I have joined the team lead by prof. Marek Moszyński in the Department of Detectors and Nuclear Electronics of the Soltan Institute for Nuclear Studies in Świerk. The team was involved in characterization of scintillator detectors for detection of gamma-rays, neutrons and charged particles. The research was also devoted to characterization of various photodetectors for scintillation readout and creating opportunity for carrying out measurements by means of different experimental techniques. Having chance to make use of my teammates experience I started to investigate the relation between scintillators light yield, decay time, scintillation efficiency as a function of absorbed energy and energy resolution.

The energy resolution is one of the most important parameters that decide on use of a given scintillator in a certain experiment. The selected set of publications presents description of experimental techniques that allow for a detailed analysis of the processes determining the scintillators energy resolution. These works are devoted to two main topics: the design of new techniques that allow to relate the non-proportional response of a scintillator with its energy resolution and to inspect the influence of slow scintillation components on the energy resolution.

### 4.2. Nonproportionality vs. energy resolution

The relation between nonproportional response of scintillator and its intrinsic energy resolution has been widely proved for several recent decades. Already in 1956 G.G. Kelly et al. published results indicating that energy resolution of thallium activated sodium iodide is worse than the value resulting from the number of photoelectrons detected by the photomultiplier [1]. However, the authors of this manuscript doubt that nonproportional response of NaI:Tl to electrons could lead to substantial deterioration of the energy resolution observed. In the same year, D. Engelkemeir was pointing out that the difference in the scintillation efficiency could be the source of nonlinear response of NaI:Tl to gamma photons [2], however when discussing his results he only pointed out the need for further inspection of the observed effect.

In years 1960-1962 A. Meyer, R.B. Murray and R. Gwin proved that there exists the relation between nonproportional response of NaI:Tl and CsI:Tl scintillators to gamma photons, electrons, charged particles and the excitation density [3],[4] induced in a scintillator due to detection of mentioned quanta and particles.

Until the end of 60s last century there were first models developed to describe the effect of nonproportionality and quantify its influence on the energy resolution for quanta and particles detected by a scintillator. P. Iredale pointed out the role of delta electrons that are generated during slowing down of particles in matter and carry energy sufficient for further ionization of the scintillation medium [5]. The delta electrons have continuous energy distribution and they are produced with statistical fluctuations. On the other hand, J.R. Prescott and G.H. Narayan put their attention to the complexity of the gamma-ray absorption process, that may be

preceded with multiple Compton scattering and of the photoabsorption that may be followed by a cascade of X-rays and Auger electrons [6]. These phenomena result in generation of large quantity of low-energy secondary electrons that are characterized with continuous energy distribution. Taking into account that the scintillation efficiency depends on the energy of particles that ionize the scintillator, secondary electrons have a direct impact on the deterioration of the energy resolution.

Despite many years of studies carried out to explain the nonproportionality effect in scintillators, the origin of this phenomenon and specifically, the reason for the differences in energy dependence of nonproportionality for different scintillators, is still not described with sufficient precision. Therefore there are big efforts made to develop models that will be able not only to define the processes that convert the energy deposited in the scintillator into light, but also will allow to predict the scintillator response to absorbed radiation depending on the chemical composition (including scintillator matrix and eventual dopants), the crystal structure and the state of matter (gas, liquid or solid as crystal or ceramics etc.) [7]-[10]. However, the development of the models requires constant development of experimental techniques that enable scintillator characterization. Thus the collected data are used for validation of the assumptions that serve as a base for introduced models and for measuring the values of parameters that are used as an input for a quantitative description of modelled phenomena.

The scintillator nonproportionality is defined as an energy dependence of a deviation of the scintillation efficiency from the value measured at an arbitrary fixed energy. In other words, it is the number of recorded information carriers per energy unit normalized to the same quotient measured at the reference energy (usually at 662 keV, equal to the energy of gamma-rays emitted from  $^{137}$ Cs):

$$NP(E) = \left(\frac{L}{E(keV)}\right) / \left(\frac{L_{662}}{662 \, keV}\right)$$

Ideally proportional scintillator would then be characterized with:

$$NP(E) \equiv 1.$$

The mentioned information carriers may be, e.g.: photons, in the case we describe scintillators, photoelectrons – if we report the measurements done with a photomultiplier or electron-hole pairs – in the case of measurements performed by means of semiconductor detectors.

The energy resolution of a scintillator is defined as a quotient of the measured full width at half maximum (FWHM) of the recorded peak and the peak centroid, giving information about the amplitude of the recorded pulse. As in the case of nonproportionality, the energy resolution is also a function of energy deposited in the scintillator and may be expressed as a sum of a few independent components [11]:

$$(\Delta E/E)^2 = \delta_{int}^2 + \delta_{stat}^2 + \delta_{trans}^2 + \delta_{noise}^2,$$

where:

- $\delta_{noise}$  (noise component) quantifies the noise of a photodetector and electronics (negligible in the case of photomultipliers, possible to be measured in the case of semiconductor detectors),
- $\delta_{trans}$  (light transport component) quantifies the spread related with a transfer of light to a photodetector (negligible in the case of currently used photodetectors if the scintillator-photodetector coupling is done correctly),
- $\delta_{stat}$  (statistical component) quantifies the spread related with the number of information carriers and is defined with the equation below:
  - $\delta_{stat} = 2.355 \times \sqrt{\frac{1+\varepsilon}{N_{phe}}}$  in the case of photomultipliers,

where  $\varepsilon$  is the PMT gain variation and  $N_{phe}$  is the number of detected photoelectrons,

-  $\delta_{stat} = 2.355 \times \sqrt{\frac{F}{N_{eh}}}$  in the case of semiconductor photodetectors,

where F is the "excess noise factor" that quantifies the spread of the photodetector gain and  $N_{eh}$  is the number of measured electron-hole pairs,

•  $\delta_{int}$  (intrinsic resolution) – quantifies the component related with the quality and performance of the scintillator.

The value of intrinsic resolution measured for a scintillator may be influenced by several factors, like inhomogeneous structure of the sample, light self-absorption in the scintillator or inhomogeneity of the reflection coefficient of the light reflector, which is used for wrapping the scintillator to maximize the light that reaches the photodetector. These effects may lead to variation of the light output reaching the photodetector, depending on the location of the scintillator act inside the sample. However, the experience of the scintillators manufacturers and users allows to minimize the influence of these factors on the intrinsic resolution. In this case the nonproportionality of a scintillator response to absorbed energy becomes the main factor limiting the intrinsic resolution of a scintillator.

Moreover, in the case of some scintillators that deexcite by emission of a few decay components, the influence of long decay components on the intrinsic resolution is observed. Specifically, extending the shaping time constant in the shaping amplifier, that is equivalent to integration of a larger part of the scintillation pulse emitted by a scintillator, resulted in a noticeable improvement of a scintillator intrinsic resolution.

In 1999 prof. Moszyński and his team published results, presenting this effect for CsI:Tl coupled to an avalanche photodiode (APD) in room temperature [12]. In the following years similar effect was also observed for ZnSe:Te [13], CsI:Tl [14] and CsI:Na [15] at room temperature and for NaI:Tl at liquid nitrogen temperature [16].

On the other hand, there was also observed an opposite effect in the case of LGSO scintillators [17]. LGSO crystals are characterize with a relatively short decay time (65 ns) that is accompanied with afterglow, i.e. the light emitted in a fairly slow process of phosphorescence. In the recalled publication a correlation between the afterglow and the

intrinsic resolution of the scintillator was pointed out. In the case of LGSO crystals with higher afterglow level, their intrinsic resolution was worse.

### 4.3. The influence of long decay components - NaI:Tl at low temperatures

During my work in the Department of Detectors and Nuclear Electronics (currently named Radiation Detectors Division) I had opportunity to cooperate with dr. Czarnacki team. Dr. Czarnacki is the leader of a group that is involved in the design and manufacture of devices for radiation measurements based on semiconductor detectors, mostly silicon detectors. My first experiments in prof. Moszyński team were focused on the response of NaI:Tl in a wide range of temperatures (between  $-40^{\circ C}$  and  $+23^{\circ C}$ ). The light readout was performed using a large area avalanche photodiode (LAAPD) that was placed in a cryostat equipped with a Peltier element, all customized by dr. Czarnacki team. The setup allowed to carry out measurements in a controlled, stabilized temperature. The description of the measurement procedure and acquired results are presented in the publication [A1].

As a part of the measurements the change of scintillation light yield was recorded as a function of crystal temperature. The light output increased while the temperature decreased. The rise of the scintillation efficiency is related with the intensification of the long decay time component. This effect is manifested through substantial increase of recorded light if long shaping time constants are used in the spectroscopy amplifier, which is equivalent to integration of larger parts of the light pulse. An interesting observation is that using different shaping time constants results in different shapes of the nonproportionality curves. This is an indirect evidence that different decay components are characterized with different scintillation nonproportionality functions. Extending the shaping time constant causes the reduction of nonproportionality in the range of low energy gamma-rays and that, in turn, results in improvement of intrinsic resolution of the scintillator. The occurrence of this effect was also confirmed in measurements performed with CsI:Tl scintillators.

The course of the measurements presented in this publication is summarized by a set of results showing that the best intrinsic resolution, recorded at various temperatures, was achieved in the case when shaping time constant was adjusted to integrate the entire light pulse.

### 4.4. The influence of long decay components - LuAG:Pr

Due to the expertise that our group possess in the field of measurement techniques used for characterization of spectroscopic properties od scintillators, in 2007 we have started a cooperation with the team lead by prof. Akira Yoshikawa (Tohoku University at this time) and dr. Kei Kamada (employed by Furukawa K.K. at this time) from Japan. Their group is involved in crystal growth with special focus on search for new scintillating materials. At the period mentioned prof. Yoshikawa proposed doping scintillators with 3-valent praseodymium ions ( $Pr^{3+}$ ) as an alternative to doping scintillators with cerium, which in some cases resulted in discovery of new materials with interesting scintillation properties. One of the first examples was LuAG:Pr (Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr), which was characterized with relatively large brightness (~15000 ph/MeV) and fast scintillation decay time (~20 ns).

We received several samples of LuAG:Pr from dr. Kamada in order to characterize their response to gamma-rays. The results of this study was presented in publications [A2],[A3]. LuAG:Pr turned out to be a scintillator with fairly proportional response, the deviation from proportionality was only about -2% at 60 keV and about -7% at 32 keV. The shape of nonproportionality curve was resembling those of oxide scintillators (e.g., BGO, LSO, LuAP) and rare-earth halides (e.g., LaBr<sub>3</sub>, LaCl<sub>3</sub>). An interesting observation was made when measured number of photoelectrons and corresponding energy resolution was compared at different shaping times. Extending the shaping time constant from 3 µs to 12 µs resulted in increase of recorded light by about 20%. This effect was caused by the presence of long scintillation components with decay time constants of the order of several microseconds and intensity ranging 75% of the entire pulse, while the fast component with decay time constant of about few tens of nanoseconds carried barely about 25% of the light intensity. Despite this fact, the nonproportionality shape remained the same, in contrast to the effect observed for alkali iodides (e.g. NaI:Tl, CsI:Tl, ZnSe:Te). The increase of recorded light was neither leading to significant improvement of energy resolution. Extending the shaping time constant in the quoted range resulted in slightly better energy resolution (5.0% at 12 µs vs. 5.2% at  $3 \mu s$ ). The outcome was that the intrinsic resolution was better at short shaping time (2.5%) than at long shaping (3.0%). The results of these measurements indicate different characteristics of the long scintillation component present in LuAG:Pr than it was observed in, for example alkali iodides. Taking into account the information included in the long decay component by integrating the light that is carried in it does not lead to improvement of nonproportionality and energy resolution, moreover it has a negative influence on the value of intrinsic resolution of this scintillator. Similar effect was observed by the team of prof. Moszyński for LGSO:Ce crystals [17], where correlation between deterioration of intrinsic resolution and increase of the afterglow in these scintillators was pointed out. Analogously, one may assume that long decay components in LuAG:Pr crystals are similar to afterglow.

It was also indicated in the publication [A3], that it is not only the crystal structure, but also the activator introduced to identical crystal structure that can change the nonproportionality characteristics, that may also be correlated with intrinsic resolution of scintillators. As the examples, differences in nonproportionality and intrinsic resolution were presented in group of scintillators LaBr<sub>3</sub>:Ce-LuAG:Ce-LSO:Ce and LuAG:Pr-LuAG:Ce, where the less proportional response to gamma-rays of different energies, the worse the scintillator's intrinsic resolution.

## 4.5. Nonproportionality and energy resolution for Compton electrons and gamma-ray absorption

For ten and a few years the team of prof. Moszyński was cooperating with dr. Guntram Pausch from a company named Target in Solingen, Germany. In 2008 dr. Pausch proposed us to implement Compton coincidence technique (CCT) [18],[19] for characterization of scintillators. Specifically, he aimed at plastic scintillators, which due to low density and atomic number usually are not capable to absorb the entire energy of incident gamma-rays, therefore the energy spectra of plastics do not allow to record full energy peaks. For this

reason the energy calibration of plastics and evaluation of their energy resolution for gammaray sources is troublesome.

Hence I built a detection system comprising a tested scintillator and a reference germanium detector (HPGe). The detection system allowed for recording in coincidence the energy of Compton electrons in the investigated scintillator and the energy of Compton-scattered gamma-rays absorbed in the reference HPGe detector. In the first part of the work I focused my attention on the events, in which gamma-rays emitted from the source scatter backwards inside the scintillator (backscattering). This allowed for precise measurement of the signal amplitude corresponding the Compton edge position in the scintillator energy spectrum. The results were published in [A4], where it was depicted that the position of Compton edge in relation to the maximum of Compton continuum depends on the type of the scintillator and is related to its energy resolution.

As a next step we decided to use the described experimental technique for investigating the relation between nonproportionality of scintillation efficiency and the energy resolution of scintillators. In order to verify the appropriateness of the method and to quantify the precision of the measurements we recorded LaBr<sub>3</sub> response to Compton electrons in the process of backscattering. The experimental procedure and the results were published in [A5]. The choice of the scintillator was not random as LaBr<sub>3</sub> is characterized with the best energy resolution amongst scintillators. Thus it is the most sensitive object for validation the mentioned experimental method. In the course of the experiment we measured the energy resolution of Compton electrons for 6 radiation sources with energies from 122 keV to 1116 keV. Moreover, 8 sources with energies between 32 keV and 1116 keV were used in order to measure the same quantity via detection of full energy absorption peaks in LaBr<sub>3</sub> scintillator. The intrinsic resolution of the scintillator turned out to be independent of the measurement method, and the values measured by means of direct  $\gamma$ -ray absorption and using CCT were lying along common curve.

After validation of the performance of our detection setup we decided to expand the scope of carried out scintillator studies. The HPGe detector is characterized with a very good energy resolution of about 1-2 keV in the range of gamma quanta recorded during Compton scattering measurements. Due to this fact it is possible to determine precisely the energy transferred to an electron in a scintillator, at a level significantly better than its energy resolution. In the case when a tested scintillator and a reference detector are sufficiently large and the distance between them is relatively small (not larger than a few times size of the detectors), it is possible to register Compton scatterings in a large range of scattering angles during one measurement session. High energy resolution of an HPGe detector allows for accurate selection of events related with scatterings at different angles, therefore it is not necessary to use slits or to place the detectors at large separation in order to reduce the solid angle, at which the reference detector is monitoring the investigated scintillator, as it was done by the inventors of this technique [18]. It is worth to mention that in recent years a group of scientists from Lawrence Berkeley National Laboratory (LBNL) in the USA designed and built the detection setup for a quick characterization of scintillators by means of the CCT. This setup is called SLYNCI [20] and it consists of 5 HPGe detectors that are monitoring the tested scintillator at various angles simultaneously with respect to the direction determined by the gamma-rays emitted by an intense radioactive source (about 1 mCi) towards the tested sample. The cost of this experimental setup amounts to about several hundreds of thousands of USD, while our detection system was built using only one HPGe detector. By using tight measurement geometry and various radioactive sources of small activity (about few  $\mu$ Ci) we were able to achieve the results of equal precision in an approximate acquisition time (about several tens of hours of measurements).

The CCT was extended in a similar way by the group of dr. Ugorowski from the Kansas State University (KSU) [21]. However, beside presenting two test cases, the KSU team did not continue scintillator studies by means of this method, in particular the were not looking for correlation between nonproportionality of light yield and energy resolution of scintillators. Our group made use of the detection setup built in our laboratory for nonproportionality and energy resolution measurements of several scintillators, being representatives of materials characterized with different response characteristics. The results of these measurements were published in [A6] and [A7].

In the publication [A6] we described the application of the CCT for measuring the nonproportionality and intrinsic resolution of scintillators with low atomic number. This technique is particularly useful for energy calibration of organic scintillators, as their low density and atomic number does not allow to register full energy peaks. As the examples we presented a plastic scintillator BC408 and a liquid scintillator EJ301. Low energy threshold limiting the range of both nonproportionality characteristics was about 10 keV that corresponded to about 20 photoelectrons emitted from the PMT photocathode during every event that carried this energy.

It was also shown in the course of this work, that nonproportionality measurement for scintillators with poor energy resolution is possible when the reference HPGe detector is replaced with a scintillator. Employing a scintillator with an appropriate energy resolution (e.g. LaBr<sub>3</sub>) as reference detector in the CCT method allowed for using high energy gamma-rays (4.44 MeV from  $\alpha$ +<sup>9</sup>Be reaction in a <sup>238</sup>Pu-Be source). The Pu-Be source is a fast neutron emitter, that may degrade the HPGe detectors performance due to radiation damage induced in the germanium crystal structure. By using the Pu-Be source the nonproportionality and intrinsic resolution could be measured in the energy range up to nearly 4 MeV. This measurement confirmed that organic scintillators show specific response to electrons, as their nonproportionality curves do not saturate above energy of about 200-500 keV, as is observed typically in the case of most scintillators.

The publication mentioned above contain also the results of the measurements recorded with a light inorganic scintillator  $CaF_2$ :Eu. Due to much higher scintillation efficiency of  $CaF_2$ :Eu it was possible to record the nonproportionality curve and intrinsic resolution down to energy as low as 3 keV, that corresponds to only 10 photoelectrons emitted from the PMT photocathode. Despite large deviation from proportional response,  $CaF_2$ :Eu is still more proportional material than organic scintillators.

In the publication [A7], besides detailed description of the method used for data analysis, the results obtained for the LaBr<sub>3</sub>, LYSO and CsI:Tl scintillators were presented. LaBr<sub>3</sub> represents scintillators from the group of the rare-earth halides and is characterized with

almost proportional response to the energy of electrons absorbed in the scintillator. LYSO is from the family of oxides, that are usually characterized by a substantial decrease of the scintillation efficiency at low energies deposited in the scintillator, whereas CsI:Tl belongs to the group of alkali iodides that show increase of the scintillation efficiency at low energies absorbed by the scintillator (about 10-100 keV). In the course of performed experiments it was shown that in the case of LaBr<sub>3</sub> and LYSO the scintillators intrinsic resolution is the same, irrespectively whether it was measured in the process of gamma ray absorption or it was determined by measuring the response to Compton electrons using CCT. A surprising result was observed in the case of intrinsic resolution of CsI:Tl crystal. The values were the same in the energy range between 200 keV and 1 MeV, whereas in the energy range from 50 keV up to 200 keV a substantial difference between values measured using both methods. The intrinsic energy resolution determined using gamma-ray full energy absorption was significantly better than the value obtained with Compton electrons. This difference was particularly pronounced when the shaping time constant of the amplifier was set short  $(2 \mu s)$ , what corresponds to neglecting the information carried by the long scintillation component (about several microseconds).

The differences in the complexity of the processes leading to scintillation emission via photoabsorption as compared to single Compton scattering take their toll on the nonproportionality characteristics measured by means of both mentioned methods. Photoabsorption is often preceded by a series of a few Compton scatterings that produce electrons with energy smaller than the absorbed gamma-ray. Moreover, as a result of photoabsorption, there is a cascade of low energy electrons emitted, originating from the excited atom that absorbed the gamma quantum. There are also low energy X-rays emitted that subsequently lead to the emission of further low energy electrons, as well as Auger electrons are observed with energies close to the electron binding energy, i.e. between fraction of keV and about 100 keV. The scintillation efficiency of these electrons is usually significantly different than the scintillation efficiency of a single electron born during a Compton scattering despite the fact, that the energy absorbed by the scintillator in both scenarios is the same. This fact leads to a separation of the nonproportionality curves measured by mans of the both methods.. In the case of rare-earth halides and oxides (like LaBr<sub>3</sub> and LYSO) we observe that nonproportionality curve measured using photoabsorption is placed below the curve recorded for Compton electrons. This is a result of lower scintillation efficiency of the electrons created during photoabsorption process as compared to a single Compton electron of higher energy. An opposite effect is present in the case of alkali iodides (like CsI:Tl), where scintillation efficiency of the electrons with energies between about 1 keV and 100 keV is higher than for electrons of higher energy. Therefore the response of a scintillator measured using photoabsorption is placed below the nonproportionality curve determined by means of single Compton electrons.

The influence of the nonproportionality on the intrinsic resolution for three crystals from the family of alkali iodides (NaI:Tl, CsI:Na, CsI:Tl) was presented in [A8]. The differences in the shapes of the intrinsic resolution curves measured with gamma-ray absorption and Compton electrons are related with the fact that photoabsorption is followed by a cascade of low energy electrons. It is worth noting that characteristic minima in the intrinsic resolution curves

determined using photoabsorption occur only in the case of alkali iodides, where scintillation efficiency is increasing while the energy of electrons is going down to about 10 keV, and then is declining at lower energies [22]. This effect is absent for other kinds of scintillators that are characterized with monotonically decreasing scintillation efficiency as the electron energies are getting lower. Hence, the absorbed gamma-rays with energies between 20 keV and 200 keV populate significant amount of the electrons at the energy range between about 1 keV and 100 keV.

Moreover, the assertion expressed above is confirmed by the correlation between the degree of nonproportionality of the tested scintillators and their intrinsic resolution. The response of the NaI:Tl scintillator deviates the least from proportionality and it is characterized with the best intrinsic resolution amongst tested crystals, whereas CsI:Tl that has the most nonproportional response shows the worst intrinsic resolution.

## 4.6. Separation of nonproportionality curves for scintillators with multiexponential scintillation decay

Despite the fact that alkali iodides are some of the oldest known scintillators, the nonproportionality of response is still poorly described for scintillators emitting light in several scintillation decay components with different decay times. The presence of more than one scintillation decay component points to the existence of various processes leading to the light emission in a scintillator. If these differences are sufficiently large the response of a scintillator to radiation can be probed by integrating smaller or larger fraction of the entire pulse. In the case when analogue electronics is used for light pulse readout, such measurements can be realized through a choice of an appropriate shaping time constant in a spectroscopy amplifier. The fact that the nonproportionality and the intrinsic resolution depend on the shaping time constant of an amplifier was proved experimentally for several types of scintillators (e.g. ZnSe:Te [13], CsI:Tl [14], CsI:Na [15]).

Hence, an interesting question was posed: is there a dependence between the scintillation decay time and the energy absorbed in a scintillator? This issue is particularly interesting in the case of crystals with multi-exponential scintillation decay.

In order to answer the question raised above, I built an experimental setup for registering scintillation pulses as a function of the energy recorded within the detector. The description of the experimental setup along with the data analysis procedure was published in [A9]. The direct motivation to begin this research was a paper published by a group from LBNL, that made an effort to measure the scintillation decay time of a NaI:Tl crystal using a PMT as a photodetector and a signal preamplifier [23]. However, this method required a deconvolution of pulses formed in the slow charge sensitive preamplifier, introducing large uncertainty to the measurement. Thus I used the signal directly from the anode output in order to record scintillation decay by means of an oscilloscope that was triggered using the PMT dynode signal allowed for a selection of events corresponding to the absorption of specific energy in the scintillator. In this way it was possible to record pulses that carried the energy

from about 10 keV (due to Compton scattering) up to full energy of gamma-rays emitted by a source used in a certain measurement.

The observations made during measurements done with single decay mode scintillators point to a very weak dependence on the energy absorbed by the crystal, showing a small (about 10-20%) extension of the decay time, or even no dependence on the energy. However, in the case of NaI:Tl that is characterized with two scintillation decay modes of 225 ns and 1.0  $\mu$ s a shortening of the fast decay mode by about 11% was observed. It is worth noting that thank to a registration of double decay mode pulses it was possible to separate nonproportionality curves for the fast and slow components, that is a unique result on a global scale. Registration of the decay time constants and amplitudes of the pulses allows for determination of the intensities carried by the following modes, while the determined intensities are proportional to the light carried in given modes.

The conclusion of the performed measurements was that the fast component that carries more than 90% of the light emitted by the NaI:Tl crystal is characterized with the same type of nonproportionality as observed for other doped alkali iodides. In contrast, slow scintillation mode shows opposite trend as a function of the energy deposited in the crystal, typical for the scintillators from the group of oxides, as well as rare-earth halides. The shape of both relations explains the fact of reducing the nonproportionality response when the shaping time constant in the amplifier is increased. Integrating a larger part of a pulse increases the contribution of the slow component in relation to the fast decay mode. Once both components show opposite trends of scintillation efficiency as a function of energy, processing them simultaneously reduces the nonproportionality of the scintillator response.

Due to measurements of fast and slow scintillation mode intensities it was also possible to determine the ratio between both components as a function of the energy absorbed by the scintillator. Above the energy of 100 keV the ratio is fixed, about 90% of light is emitted in the fast decay mode, while 10% falls within the slow component. Below the energy of 100 keV the contribution of the slow mode is getting lower, reaching the level of just 3% at the energy of about 10 keV.

A similar effect was observed in the course of studies on CsI:Tl crystals [24]. CsI:Tl is responding much slower (main decay mode about 1  $\mu$ s) than NaI:Tl (main decay mode about 0.22  $\mu$ s), nevertheless in the case of CsI:Tl the contribution of the fast decay mode is increasing, mostly at the cost of the longest component. Interestingly, the results presented in both publications were obtained using different experimental techniques. In the case of CsI:Tl crystals, discussed in the publication [24], the light pulses were recorded using the Thomas-Bollinger method (by means of a so-called "single photon method"), whereas the results for NaI:Tl [A9] were obtained by recording the pulses using a digital oscilloscope gated to choose a specific energy deposited in the crystal.

### 4.7. Summary

Described in the previous chapters experimental techniques, that have been developed by myself within the last few years, allow for significant extension of the experimental data that may be used for more precise characterization of light emission in scintillators.

The study of nonproportionality response modification through a choice of appropriate signal shaping time constant sheds light on the influence of long decay modes on the energy resolution of scintillators. Taking into account long decay modes in the signal processing can influence in various ways the spectroscopic properties of scintillators. This fact denotes different origins of processes that are responsible for generating scintillation with different decay times.

Comparing scintillators nonproportionality responses with their intrinsic resolution measured by means of Compton electrons and gamma-ray absorption was developed by myself on a unique scale in the scientific community dealing with the scintillators characterization. A particularly interesting issue is the observation of the differences in the intrinsic resolution measured with the two mentioned methods for the scintillators from the group of alkali iodides.

The measurement of nonproportionality for separated scintillation components is a brand new experimental technique, that gives an insight to the scintillation properties that has never been investigated before. The fact that the two scintillation modes have different, and even reversed nonproportionality profiles explains the observed correlations between applied shaping time constant and nonproportionality response for the scintillators from the group of alkali iodides.

The observations of mentioned phenomena are of special importance for so-called "scintillator engineering", because they can be helpful for describing the mechanisms that are responsible for differences in the nonproportionality characteristics of various trends. The data obtained by means of the innovative methods presented above can be used for validation of theoretical models, that are developed in order to create a complete description of the processes leading to scintillation. Having a complete description of the influence of a scintillator composition on the light emission process, including the dopants and the crystal structure, it will be possible to modify the intensity ratios of various scintillation components. In this way the response of a scintillator to absorbed radiation could be made close to proportionality. As a result, it would be possible to grow scintillators with optimized energy resolution.

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# 5. Description of other scientific achievements.5.1 Nuclear reaction physics

In the course of my PhD studies I was involved in a series of experiments devoted to the measurements of fusion barrier distributions. These experiments were carried out on the cyclotron at the Warsaw University Heavy Ion Laboratory (HIL) by the group led by dr. hab. Ernest Piasecki. My input was to set up the detectors and readout electronics, to prepare the experimental chamber, to analyze the data and to compare the results with model calculations. The description of the experiments together with obtained results comprised the material that was used in my master's dissertation and, subsequently in my PhD thesis. Moreover, the results were published in the series of publications [B1]-[B5].

Just after my doctoral defense and the beginning of my work at Soltan Institute for Nuclear Studies, I participated to a similar extent in the next experiments on the same subject. One of those experiments was carried out on the tandem accelerator at Jyvaskyla University (Finland). The results of these experiments were published in [B16], [B38], [B57].

Already during my doctoral studies I was involved in an experiment devoted to dynamical fission in the reactions Sn+Ni and Au+Au. The measurements were done by means of a detection setup called CHIMERA, which was placed at the cyclotron in Laboratori Nazionale del Sud INFN in Catania (Italy). My contribution to the research was limited to minor participation in the measurement sessions and the data collected in this experiment was published before my doctoral defense [B8]-[B11], as well as in the following years [B19]-[B24], [B46]-[B47].

In the period of preparation of the experiments at the HIL cyclotron I was also involved in the tests of monolithic semiconductor " $E-\Delta E$ " telescopes. These detectors were investigated on the usefulness for energy and atomic number measurements of heavy ions being a product of inelastic scattering. The person responsible for the detectors construction was dr. hab. Andrzej Kordyasz and the results of the tests were published in [B6]-[B7]. My role was to help in preparation of the experimental setup (the target for beam scattering, the detector for beam energy monitoring via measurement of elastically scattered ions) and to take part in the measurement sessions on the beam provided by the cyclotron.

### 5.2 Neutron detection

After I was employed in the Department of Nuclear Electronics in the Soltan Institute for Nuclear Studies, I started to study the properties of liquid scintillators for neutron detection. In the light of emerging crisis of helium-3 supply manifested by sudden increase of its price, liquid scintillators have attracted again the attention of the scientific community as a cheap and effective alternative for helium-3 counters. A specific position among tested detectors was filled by the scintillators loaded with boron-10. Liquid scintillators are sensitive to fast neutrons (through elastic scattering) due to large content of hydrogen. When loaded with B-10 isotope their detection efficiency is raised due to the detection of slow neutrons. However, the problem is to discriminate the signals induced by neutrons and gamma-rays. The issues related with optimizing the performance of liquid scintillators on the neutron/gamma discrimination were discussed in several publications [B25], [B42]-[B44], [B52], [B62].

The fact that our group has been very active in the field of scientific research resulted in establishing the collaborations with facilities engaged in production of new scintillators proposed for neutron detection as an alternative for counters based on He-3. Thanks to cooperation with prof. Nikolai Galunov from the Institute for Single Crystals of National Academy of Sciences of Ukraine we had an opportunity to study the neutron response of composite scintillators. These detectors are formed as small grains of stilbene or p-terphenyl bound in a matrix made of transparent resin [B53], [C2]. The breakthrough in the scintillators field came when the plastics with neutron/gamma discrimination capability were introduced. We received the first samples of such plastic scintillators thanks to a collaboration with the group of prof. Natalia Zaitseva from the Lawrence Livermore National Laboratory (LLNL, USA) and with the group of prof. Alex Gektin from the Institute for Scintillation Materials (ISMA) in Kharkov (Ukraine) [C3]. The compound developed by the team from the LLNL was patented and is currently used for production of commercially available scintillators. We have published the results of the tests on the first samples due to a cooperation with the company Scionix Holland B.V., which offers these detectors known as EJ299-33 and EJ299-34 [B61].

In the same period of time we were invited to cooperate with a Japanese company Tokuyama, which was developing a method for production of LiCaAlF6 (LiCAF) crystals doped with either cerium or europium. In the case of these detectors neutron detection is realized via capture reactions on Li-6 nuclei, that comprise the crystal structure of the mentioned

scintillator. The samples that we received from our partners were characterized on their response to gamma radiation and thermal neutrons [B54].

The results presented in the publications [B53]-[B54], [B61], [C2] were the basis of the doctoral dissertation by Dr. Joanna Iwanowska-Hanke. The PhD thesis was defended in April 2016. In the case of this PhD thesis I was the secondary doctoral supervisor.

### 5.3 Scintillators characterization

Since about year 2000 the main activity of prof. Moszynski group was related with studies of the main scintillator properties. As a member of this group I took part in numerous experiments, data analysis and discussions of the results concerning the recorded light yield [B17], the energy resolution [B30], [B67] or the scintillation pulse shapes [B31]. In cooperation with my colleagues I was also involved in the study of the relations between the nonproportionality and the energy resolution in scintillators such as: CsI:Tl [B18], [B60], CsI and CsI-CsBr [B28], BGO [B29], NaI [B35], LSO:Ce:Ca [B40], CsI:Na [B45], LFS-3 [B55], CdWO<sub>4</sub> and ZnWO<sub>4</sub> in LN<sub>2</sub> temperature [B58], GAGG:Ce [B59], [B64] and high pressure Xe gaseous scintillator [B63].

In 2006-2010 we hosted in our Division for several times prof. Weerapong Chewpraditkul from the King Mongkut's University of Technology Thonburi (KMUTT) in Bangkok (Thailand). In the course of collaboration with KMUTT we carried out tests of gamma-ray spectrometry performance of the following scintillators: CsI:Na and CsI:CO<sub>3</sub> [B27], LuAG:Ce, YAG:Ce and LYSO:Ce [B34], [B39], as well as LaCl<sub>3</sub>:Ce [C1].

A large part of the research were also the studies of photodetectors used for the scintillation light readout in gamma-ray spectrometry. Among the detectors tested by myself, there were photomultipliers [B26], [B51], silicon PIN-diodes and avalanche photodiodes (APD) [B26], [B32]-[B33], silicon drift detectors (SDD) [B33], [B41] and multi pixel photon counters (MPPC) [B50]. Separate task was the problem of the detectors performance optimization paying special attention to achieve possibly best coincidence timing resolution [B36]-[B37], [B48]-[B49].

### 5.4 Neutron activation analysis

In the recent few years I was also involved in the research guided by dr. Michal Gierlik team. This group is dealing with neutron activation analysis (NAA) for the detection of hazardous materials, for example explosives. The published results were focused on the choice of optimal scintillator with high detection efficiency for high energy gamma-rays (about several MeV) and good energy resolution [B56]. Subsequently, an active anti-Compton shield was proposed in order to reduce unwanted gamma-ray background [B65]. Application of the anti-Compton resulted in an increase of the detection setup sensitivity. The collected data was then used for the optimization of the algorithms implemented for identification of potentially hazardous materials [B66].

#### 5.5 Plasma diagnostics in thermofusion reactions

In the last three years I was involved in the preparations and partly in the realization of a project devoted to construction of detectors for plasma diagnostics in devices like TOKAMAK. The activity included the choice of scintillators and photodetectors for high energy gamma-ray monitor, so called "Gamma Camera", the choice of a scintillator and a photomultiplier for building the gamma-ray spectrometer and the design of detectors for monitoring of lost alpha particles that escape from the plasma. A part of these works was presented at the 1st EPS conference on Plasma Diagnostics (ECPD) in Frascati (Italy) in April 2015 [K20], and subsequently published in Proceedings of Science [C4].