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Conversion electron Mössbauer spectrometer with a YAG:Ce scintillator

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Abstract
In this work we report the development of a new scintillation detector of conversion electrons for ^{57}Fe conversion electron Mössbauer spectroscopy. The detector, a photomultiplier tube with a 5 μm YAG:Ce scintillator, is sensitive to both X-rays and (conversion) electrons. Electrons and X-rays have different average depths from which they come out of the sample. The detected electrons are much more likely to come from close to the surface, whereas X-rays can come from greater depths in the sample. By applying an electric field over the sample and detector, electrons were accelerated towards the detector making more electrons being detected and giving them higher energies. On the other hand, X-rays were not influenced by the electric field making the electrons distinguishable from X-rays and the background.

Keywords YAG:Ce · Mössbauer spectrometer · Conversion electron · CEMS

1 Introduction

Conversion electron Mössbauer spectroscopy (CEMS) is one of the methods by which one can investigate Mössbauer radiation in backscattering geometry. We can detect conversion electrons contributing to the resonant peak as a result of the direct internal nucleus-electron interaction conversion processes in a part of nuclear transitions from the excited to the ground state. There are a wide range of principles, techniques and devices to observe Mössbauer resonant absorption in the backscattering geometry due to the internal conversion process with the conversion electrons registration. Gas

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detectors, scintillation detectors, electron multipliers, electron energy analysers, surface barrier silicon semiconductor detectors, orange type magnetic spectrometers and beta ray spectrometers are among these devices. A review of various kinds of the electron detectors used in CEMS was presented in [1].

The features and use of the YAP:Ce and YAG:Ce scintillators as detector of gamma- and X-rays in relation to ^{57}Fe Mössbauer spectroscopy are presented and discussed in [2]. However, those scintillators are used to detect the low energy electrons in electron microscopy [3]. Consequently, the application of YAP:Ce or YAG:Ce in CEMS is possible. The detector, a photomultiplier tube with a YAG:Ce scintillator, in CEMS, is presented and discussed in this paper.

2 YAG:Ce scintillator

Physical and scintillation characteristics of YAG:Ce and other fast light pulse inorganic crystals in relation to the NaI:Tl crystal are listed in Table 1. Luminescence and absorption spectra YAG:Ce crystal manufactured by the Chochralsky method in CRYTUR (Turnov, Czech Republic) in comparison with YAP:Ce are given Figs. 1. The luminescence bands of the YAG:Ce crystal are near the maximum sensitivity spectral range of the bialkali and multialkali photocathodes of commonly used photomultipliers.

Table 1 Physical and scintillation characteristics of the fast light pulse inorganic scintillators in comparison with NaI:Tl

	YAP:Ce	NaI:Tl	YAG:Ce
Density [g/cm^3]	5.37	3.67	4.57
Hardness [Mho]	8.6	2	8.5
Index of refraction	1.95	1.85	1.82
Crystal structure	Rhombic	Cubic	Cubic
Melting point [°C]	1875	651	1970
Hygroscopic	No	Yes	No
[a]Therm. Exp. [10^{-5}/K]	4–11	4.75	8–9
Cleavage	No	Yes	No
Chemical formula	$YAlO_3$	NaI	$Y_3Al_5O_{12}$
Light output [%NaI:Tl]	45	100	20
Max. of emission [nm]	350–360	415	550
Decay constant [ns]	28	230	70
[b]Absorption length [cm]	2.22	–	2.61
Light yield [ph/MeV]	18,000	38,000	9000
[c]Relative efficiency [%]	150	–	125

[a] Linear coefficient of thermal expansion

[b] Absorption length to the 1/e value for 511 keV of gamma ray

[c] Relative luminescence efficiency for beta rays compared with a plastic scintillator

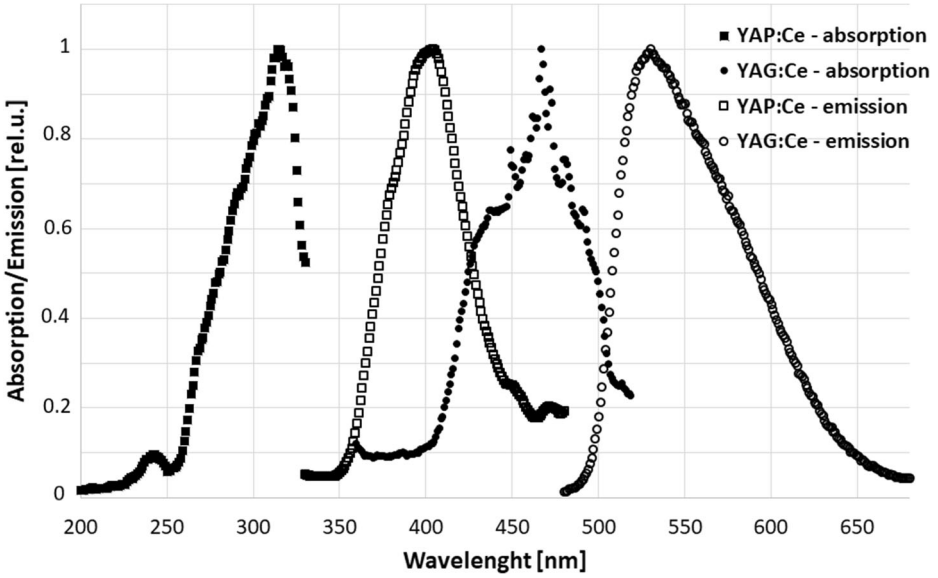

Fig. 1 Absorption and emission spectra of YAG:Ce and YAP:Ce crystals

3 Spectrometer design

3.1 Scintillation detection unit

A scintillation detector converts the energy of detected particles in light impulses, which in turn are captured by a photodetector and converted in electrical impulses. These electrical impulses are registered outside the scintillation detection unit. The scintillation detection unit was produced by CRYTUR (Turnov, Czech Republic) for the detecting ionisation radiation

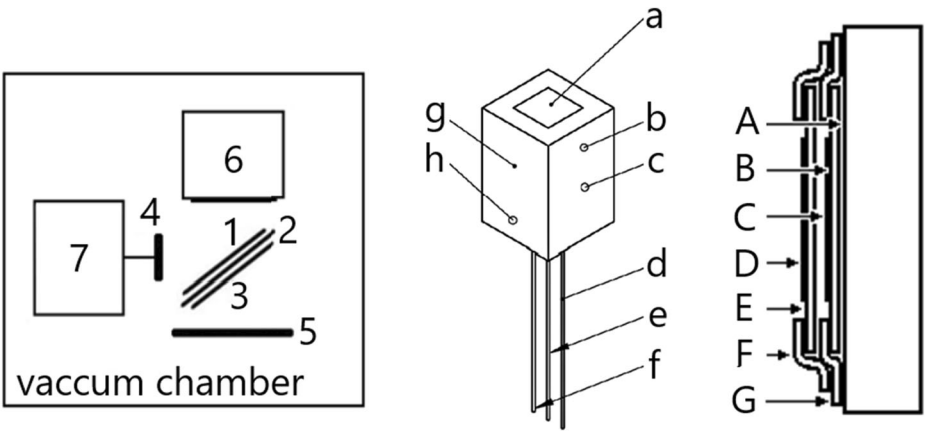

Fig. 2 On the left side: the CEMS experimental setup: testing sample (1 – α-^{57}Fe, 2 – Mylar, 3 – α-^{57}Fe2O3), 4 – ^{57}Co(Rh) source, 5 – metallic electrode, 6 – scintillation detector and 7 –transducer), in the middle: the scintillation detector: (a – YAG:C e crystal, b – photomultiplier, c – magneticshielding, d – high voltage cable, e – signal cable, f – ground, g – aluminum cover and h – voltage divider)and on the left: the testing sample (A – mounting block, B – sheet of paper, C – α-^{57}Fe2O3, D – α-^{57}Fe, E –Mylar, F – pressure-sensitive tape and G – pressure-sensitive tape

and specifically low-energy electrons. It consists of a 5 μm thick YAG:Ce scintillation crystal with a 20 nm ITO layer on the detection side and a photomultiplier tube with magnetic shielding. Even though the light output of a YAP:Ce crystal is greater (see Table 1) YAG:Ce crystal was used because it is possible to produce this crystal 5 μm thin. The rhombic crystal structure of the YAP:Ce makes a thickness less than a 100 μm unrealistic to produce. Thin scintillation crystals are less suitable for the registration of X-rays if the radiation length for X-rays is greater than thickness of the crystal. However, they are suitable for the registration of electrons if the radiation length is less than the crystal thickness. Using the thin scintillation crystal, we can decrease the contribution of X-rays to the output signal of detector.

3.2 Experimental arrangement

The experimental set-up (Fig. 2) is a normal CEMS arrangement with the scintillation detection unit as the detector and a transducer with a ^{57}Co source. A high voltage power supply has been added to the set-up to create a voltage difference between the photocathode of the photomultiplier (a negative voltage of −650 V) and a metallic electrode (a negative voltage up to −11 kV) located behind the sample. The resulting electric field ε has an intensity that ranges between 0 and 5 kV/cm. The whole set-up is built into a Pfeiffer vacuum chamber with a Pfeiffer Duo 3 M rotary vane pump and a Hipace 80 turbopump, enabling the vacuum chamber to be pumped down to pressures below 10^{-5} hPa.

3.3 Software adaptation

The software used to register the spectra is a modified version of the Nuclear DSP System – Mössbauer" spectrometer presented in [4]. The original software is designed to measure in an energy window, which has to be chosen before any measurement by multichannel analyser. During the measurement itself, all data within this energy window are summed creating a 1D-array, whereas all the data outside the window are ignored. In the modified version, it is no

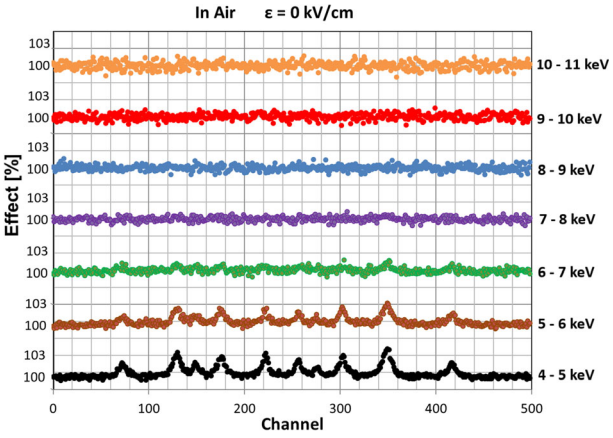

Fig. 3 The Mössbauer spectra of testing sample in air for different energy windows

longer necessary to define the precise measuring window wanted. All the data are saved in a 2D-array and a specific measuring window can be looked at during or after the measurement providing greater flexibility during the measurement. Therefore, we can have Mössbauer spectra for different energy windows (see Fig. 5) from one process of data accumulation.

4 Results and discussion

To be able to distinguish between electrons and X-rays a set of measurements have been performed with a testing sample containing two ^{57}Fe chemical compounds. The first component was the 1 μm thin layer of α-Iron applied on a 70 μm thick sheet of Mylar. The second one was a 100 μm thick layer of pressed α-Fe_2O_3 powder. The two components were arranged as a layered sample, with the α-Iron in front of the α-Fe_2O_3 layer as seen from the scintillation detector (Fig. 2). The interface layer of Mylar absorbs conversion electron originating in the α-Fe_2O_3 layer.

Both components respond with a sextet, the difference between them is the different magnetic splitting. The α-Iron (B = 33 T) has a narrower sextet than α-Fe_2O_3 (B = 51 T) which makes it easy to discriminate between the two. The first Mössbauer spectra were accumulated at atmospheric pressure (in air) without an electric field applied for different energy windows at the same time (Fig. 3). The Mössbauer effect was only observed for the energy windows 4-5 keV and 25–35. Since the spectra were registered in air only X-ray photons were detected because at atmospheric pressure electrons are absorbed by the molecules in the air and cannot reach the detector.

Next Mössbauer spectra were accumulated in a vacuum with ε = 0 kV/cm (Fig. 4). The contribution of the α-^{57}Fe subspectra has increased for both energy windows 4-5 keV and 5-6 keV. Moreover, the α-^{57}Fe Mössbauer spectra can be observed for the energy windows 6-7 keV and 7-8 keV due to the detection of the conversion electrons. However, the spectra for the energy windows 4-5 keV and 5-6 keV contain contributions

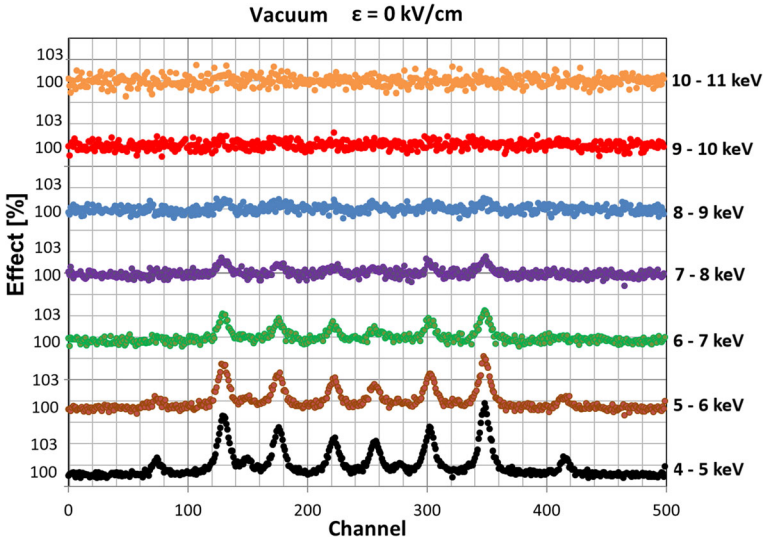

Fig. 4 The Mössbauer spectra of the testing sample in a vacuum for different energy windows

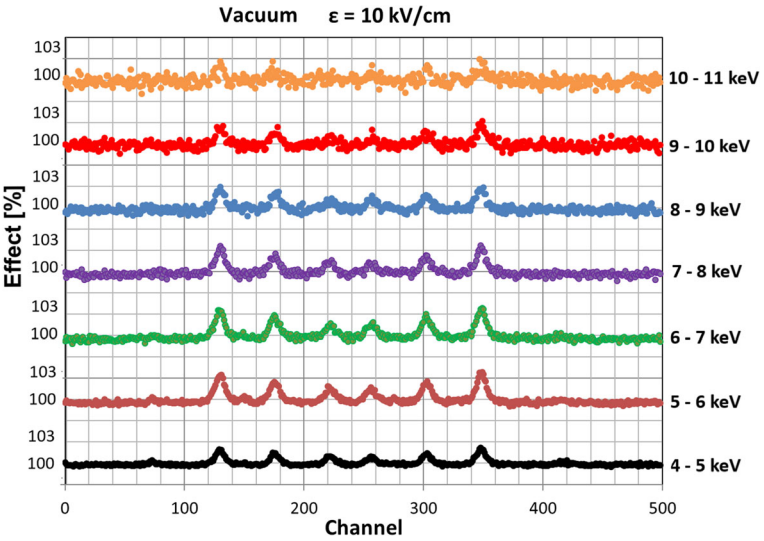

Fig. 5 The Mössbauer spectra of the testing sample in a vacuum with a 5 kV/cm supplemental electric field for different energy windows

of electrons and X-ray photons in the α-^{57}Fe subspectrum. The energy of the conversion electrons can be increased by applying a supplemental electric field between the detector and the sample for accelerating the electrons. The influence of such an electric field is demonstrated in the Mössbauer spectra (Fig. 5). The application of a 5 kV/cm electric field has two effects on the resulting spectra. The first one is the contribution of the α-iron subspectra increases for all energy windows; secondly the Mössbauer subspectra for both α-^{57}Fe and α-$^{57}Fe_2O_3$ are drowned out for the energy windows 4-5 keV and 5-6 keV by increased noise levels.

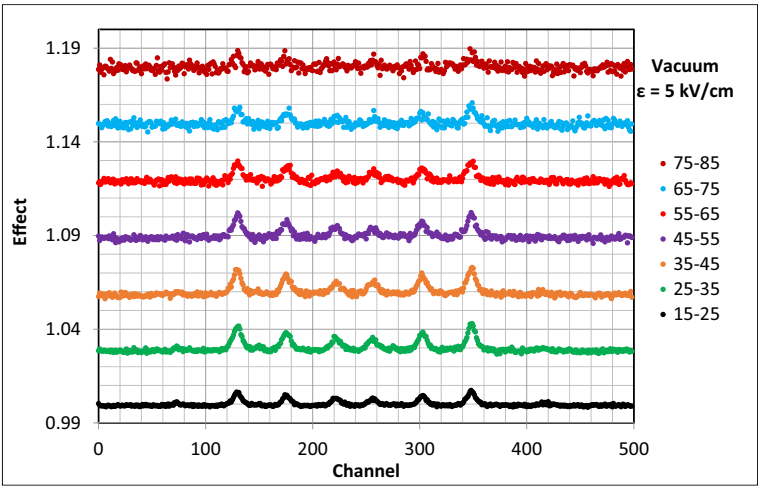

Fig. 6 The Mössbauer spectra of the testing sample in a vacuum with a 5 kV/cm supplemental electric field for different energy windows

5 Conclusions

The investigation of the scintillation detector with a 5 μm thick YAG:Ce scintillation crystal demonstrate the possibility to use this detector for registration low-energy electrons in CEMS in spite of partially detecting secondary X-rays which can be considered noise for CEMS.

The application of a supplemental electric field accelerates conversion electrons and consequently increases the contribution of conversion electrons to the Mössbauer spectrum. Electrons could be distinguished from X-ray photons by choosing a suitable energy window.

Based on the ability of the software to measure in different energy windows at the same time and the likelihood of higher energy electrons coming from smaller depths of a sample one can expect the described CEMS to be able to function as DCEMS. However, the testing sample used to prove the ability of the spectrometer to detect electrons is not suitable to test the DCEMS capabilities of the spectrometer. In order to test the DCEMS capabilities of the spectrometer a new testing sample having a special thin (less than 1 μm) sandwich with alternating layers of ^{57}Fe different chemical compositions is proposed.

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References

1. Frydrych, J., Mashlan, M., Pechousek, J., Jancik, D.: Conversion Electron Detectors for ^{57}Fe Mössbauer Measurements. In Mössbauer Spectroscopy in Materials Science (Proceedings of the International Conference – MSMS´08) AIP Conf. Proc. **1070**, 170–184 (2008)
2. Mashlan, M., Jancik, D., Kholmetskii, A.L.: YAG:Ce and YAP:Ce – suitable fast detectors for transmission Mössbauer spectroscopy. Hyperfine Interactions. **139**(1–4), 673–678 (2002)
3. Autrata, R., Schauer, P.: Cathodoluminescent properties of single crystal materials for electron microscopy. Luminescence: Proceedings of the 13th Prefferkorn Conference. 1–12 (1995)
4. Pechousek, J., Mashlan, M., Frydrych, J., Jancik, D., Prochazka, R.: Improving detector signal processing with pulse height analysis in Mössbauer spectrometers. Hyperfine Interactions. **175**(1–3), 1–8 (2007)