## **RESEARCH ARTICLE**



MICROSCOPY RESEARCH & TECHNIQUE

WILEY

# Overview of S(T)EM electron detectors with garnet scintillators: Some potentials and limits

Revised: 14 October 2020

Petr Schauer<sup>1</sup> Ondřei Lalinský<sup>1</sup> | Miroslav Kučera<sup>2</sup>

<sup>1</sup>Institute of Scientific Instruments of the CAS, Brno, Czech Republic

<sup>2</sup>Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic

## Correspondence

Petr Schauer, Institute of Scientific Instruments of the CAS, Brno, Czech Republic. Email: petr@isibrno.cz

## Funding information

Czech Academy of Sciences, RVO:68081731; Ministry of Education, Youth and Sports of the Czech Republic, Grant/Award Number: LO1212; Ministry of Education, Youth and Sports of the Czech Republic and European Commission, Grant/Award Number: CZ.1.05/2.1.00/01.0017; Ministry of Industry and Trade of the Czech Republic, Grant/Award Number: TRIO FV30271

Review Editor: Chuanbin Mao

## Abstract

The paper is focused on a complete configuration and design of a scintillation electron detector in scanning electron and/or scanning transmission electron microscopes (S(T)EM) with garnet scintillators. All processes related to the scintillator and light guide were analyzed. In more detail, excitation electron trajectories and absorbed energy distributions, efficiencies and kinetics of scintillators, as well as the influence of their anti-charging coatings and their substrates, assigned optical properties, and light guide efficiencies of different configurations were presented and discussed. The results indicate problems with low-energy detection below 1 keV when the scandium conductive coating with a thickness of only 3 nm must be used to allow electron penetration without significant losses. It was shown that the short rise and decay time and low afterglow of LuGdGaAG:Ce liquid-phase epitaxy garnet film scintillators guarantee a strong modulation transfer function of the entire imaging system resulting in a contrast transfer ability up to 0.6 lp/pixel. Small film scintillator thicknesses were found to be an advantage due to the low signal self-absorption. The optical absorption coefficients, refractive indices, and the mirror optical reflectance of materials involved in the light transport to the photomultiplier tube photocathode were investigated. The computer-optimized design SCIUNI application was used to configure the optimized light guide system. It was shown that nonoptimized edgeguided systems possess very poor light guiding efficiency as low as 1%, while even very complex optimized ones can achieve more than 20%.

## **KEYWORDS**

cathodoluminescence, conductive coating, garnet film scintillator, light guide, SEM electron detector

#### INTRODUCTION 1

Evaluating potentials and limits of a scintillation detector of electrons in a scanning electron microscope and/or scanning transmission electron microscope (S(T)EM) requires assessing several processes associated with the transfer of signal electrons from a specimen to a current amplifier of S(T)EM detection electronics (Reimer, 1993, 1998; Schauer, Lalinsky, & Kucera, 2019). To evaluate the detector, it is ideal to study a whole complex of processes, such as the efficiency of signal electron collection at the scintillator surface, electron energy losses in

a conductive coating on the scintillator, or the efficiency and kinetics of electron to photon conversion (of cathodoluminescence [CL]) in scintillator luminescent centers. It is also necessary to study the propagation of light signal generated in the scintillator, that is, absorption, reflection, and refraction of light and its coupling to the light guide. Similar processes related to the transfer of light signal must be studied not only in the scintillator but also in the light guide, including light coupling to the photomultiplier tube (PMT) photocathode. A study of photon conversion to photoelectrons, their subsequent collection at the first dynode, and the process of multiplication in PMT are no MICROSCOPY

longer urgent because the properties of PMTs have long been known and commercially offered in manufacturers' catalogs.

The analyses in this paper do not focus on those processes that depend on an arrangement of the chamber and electron optics of a particular electron microscope. In other words, processes that cannot be considered in general and are closely linked to the relevant microscope are omitted. So the analysis of the signal electron collection (Mullerova & Konvalina, 2009) will not be presented. As already announced, due to the low urgency, the processes associated with the photoelectron processing in PMT are also omitted. The rest of the important processes mentioned above will be analyzed. However, the study of all the mentioned processes is not equally important. Either because they have a relatively smaller effect on the detector performance, or because their theoretical, technical or technological limits have almost been met. This paper aims to point out these facts.

In the scintillation detector, the essential and indispensable element is the scintillator. However, individual processes that take place in the scintillator have a different priority of importance. The key priority is to use an extremely fast scintillator with a short decay time and a low afterglow, which guarantees a strong modulation transfer function (MTF) of the entire imaging system (Bok & Schauer, 2014b; Joy, Joy, & Bunn, 1996). Therefore, a study of scintillator kinetics is crucial. All processes associated with light processing occur in the order of picoseconds, but the conversion in the scintillator takes place in time many orders of magnitude longer. Especially, in the past, the CL efficiency of the scintillator has been exaggerated. However, the CL signal-to-noise ratio-based detective quantum efficiency (DQE) has a much higher priority (Joy et al., 1996). Unfortunately, the scintillator's efficiency is often unnecessarily reduced due to poor light collection on its exit side. This problem also needs attention. Moreover, under certain circumstances, there is also a loss of scintillator efficiency due to the absorption of signal electrons outside the scintillation material, that is, in its conductive coating, or in its substrate, if used. Although the light guide has a lower priority than the scintillator, it is not advisable to ignore its efficiency.

The scintillator crucially determines the performance of any electron detector in S(T)EM. Therefore, in the scintillation detector, the choice of fast scintillator is of great importance. Much faster scintillation materials are still being sought (Dujardin et al., 2018; Lecog, Gektin, & Korzhik, 2017; Tamulaitis et al., 2017). The generation of photons during energy conversion in the scintillator is the most critical detection process in S(T)EM. Physically, this is the phenomenon of the CL, and the determining physical quantities of this process are mainly the CL efficiency and CL time response. The time response is usually expressed using CL decay characteristics, from which it is possible to read both the decay time and afterglow. The cerium-activated yttrium aluminum garnet (YAG:Ce-Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce<sup>3+</sup>) and expensive yttrium aluminum perovskite (YAP:Ce-YAIO<sub>3</sub>:Ce<sup>3+</sup>) single crystal scintillators are usually utilized in the S(T)EM scintillation electron detectors (Autrata, Schauer, Kvapil, & Kvapil, 1978; Autrata, Schauer, Kvapil, & Kvapil, 1983; Chapman, Craven, & Scott, 1989; Everhart & Thornley, 2004; Hejna, 1987; Hibino, Irie, Autrata, & Schauer, 1992; Pawley, 1974; Reimer, 2013). Much attention has been paid to the examination of their conversion efficiency, their time response, and their spectral characteristics in the last decades (Mihokova et al., 2007; Nikl et al., 2014; Schauer, 2011; Zorenko et al., 2005; Zorenko et al., 2010; Zych, Brecher, & Glodo, 2000). This paper focuses on the aluminate garnet scintillators, mainly because they are easier to prepare and, therefore, cheaper. Their easier preparation also offers more considerable variability in the preparation technologies and the incorporation of different co-dopants (Bolek et al., 2020; Buryi et al., 2019; Gorbenko, Zorenko, Paprocki, et al., 2017; Gorbenko, Zorenko, Witkiewicz, et al., 2017; Kucera et al., 2016; Kucera & Prusa, 2017; Nikl et al., 2014; Witkiewicz-Lukaszek et al., 2018; Witkiewicz-Lukaszek et al., 2020).

Unfortunately, the decay time of most of the single crystal garnet scintillators used so far is too long. Moreover, far more destructive is that these scintillators have high afterglow (Chewpraditkul et al., 2009; Nikl et al., 2007; Nikl et al., 2013; Nikl & Yoshikawa, 2015). A very promising reduction in garnet scintillator time response was reported in papers on the Ce<sup>3+</sup> activated single crystalline epitaxial garnet film scintillators (Babin, Chernenko, Kučera, Nikl, & Zazubovich, 2016; Kucera & Prusa, 2017; Prusa et al., 2015; Zorenko et al., 2012; Zorenko et al., 2015). These scintillators were first tested in SEM in 2016 (Bok et al., 2016), where the GAGG:Ce garnet films were applied. Later it was found that the significantly improved CL response time, satisfying CL efficiency, and better light collection predetermine the LuGAGG:Ce,Mg (i.e., LuGaGdAG:Ce,Mg) garnet films for an S(T)EM application (Schauer et al., 2019). The positive effect of Mg co-doping and a relative small thickness are why some of our previous results concerning the garnet films are also summarized in this paper.

In this paper, the scintillator is understood not only as the scintillation material itself but also with its conductive coating and substrate, if necessary. If the detector is equipped with a bulk single crystal scintillator having a thickness much larger than units of micrometers, and if the detected electron energies are in the tens of kiloelectron volts, the study of interaction volumes in the scintillator is of little importance. However, this paper aims to also assess the epitaxial garnet film scintillators on some substrates, which have high application potential for the electron detectors in S(T)EM (Bok et al., 2016; Kucera & Prusa, 2017; Schauer et al., 2019). There is also an effort to assess the possibilities of low-energy electron detection, where the conductive coating on the scintillator can cause significant losses due to electron absorption. Usually, no attention is paid to the interaction volumes or the influence of the conductive coating when assessing the scintillation electron detector. However, in this paper, it is analyzed.

The light guide efficiency of the scintillation detector for S(T)EM has only very rarely been published. Unfortunately, some publications offer only a 2D analysis of the light collection, simplifying the problem only to a very rough estimate (Danilatos, 2012). If a quantitative analytical approach is chosen, only very simple system geometries with a high symmetry can be calculated (Carrier & Lecomte, 1990b; Filippov, Rau, Sennov, Boyde, & Howell, 2001). A method which makes use of a Monte Carlo (MC) simulation is more generally applicable. It does

not depend on the simulated systems' symmetry, but some works are still limited to parallelepipeds (Carrier & Lecomte, 1990a; Lerche et al., 2008; Xiaoguang, 1984) and others to rotational symmetries (Schauer & Autrata, 1992). The universal 3D MC method, intended for nearly any scintillation detection system for S(T)EM, is the SCIUNI application (Schauer, 2007). As will be shown in this paper, poorly designed and unsatisfactorily coupled light guides, especially in the backscattered electron (BSE) scintillation detectors, can have very low efficiencies, even as little as 1%, sometimes less. It is also a priority for the light guide to produce it inexpensively, whether by choosing an inexpensive material or simplifying its machining and assembly. Certainly, it is also necessary to take into account the risk of damaging or destroying the light guide. Although probably the smallest priority, but not negligible, is the right choice of the photocathode and fast multiplier system. It should not be a problem with a commercially well-designed component. It is just a matter of choosing the right commercial product (Hamamatsu, 2016), and it will not be addressed in this paper.

## 2 | MATERIALS AND METHODS

## 2.1 | Investigated scintillators

One standard Ce<sup>3+</sup> activated yttrium aluminum garnet (YAG:Ce– $Y_3Al_5O_{12}$ :Ce<sup>3+</sup>) single crystal scintillator and five Ce<sup>3+</sup> activated epitaxial garnet film scintillators were included for investigation in this paper. The YAG:Ce scintillator, which is often used as a reference crystal in our laboratory (Schauer et al., 2019), was grown with the Czochralski method and subsequently cut and polished into a disc with a diameter of 10 mm and a thickness of 1 mm in Crytur Ltd. The electron impact side of this scintillator was perfectly polished, while the light exit side was matted.

The Ce<sup>3+</sup> activated garnet film scintillators were prepared in the Technology Laboratory of Charles University, Prague, by isothermal dipping liquid phase epitaxy (LPE) (Kucera & Prusa, 2017). The films were grown from the BaO-B<sub>2</sub>O<sub>3</sub>–BaF<sub>2</sub> flux and deposited onto different oriented Czochralski-grown substrates. Such film scintillators were subsequently shaped to discs of 10 mm diameter. The film thicknesses were determined by weighing. The composition was determined by electron probe microanalysis and the Mg content by glow discharge mass spectrometry. The electron impact sides of all film scintillators were smooth, while the light exit sides of their substrates were matted.

The following five film scintillators were included in this overview.

- The sample, hereinafter referred to as "LuGdGaAG:Ce, high Ce" is a highly Ce<sup>3+</sup> activated lutetium gadolinium gallium aluminum garnet film. This film scintillator is also presented as LuGAGG:Ce in other papers (Chewpraditkul et al., 2020; Schauer et al., 2019; Schauer, Lalinsky, Kucera, Lucenicova, & Hanus, 2017).
- 2. The sample, hereinafter referred to as "LuGdGaAG:Ce,Mg" is nearly the same film scintillator as the previous one, but  ${\rm Mg}^{2+}$

MICROSCOPY

co-doped. The chemical formula of both mentioned film scintillators is  $(Ce_{0.01}Lu_{0.27}Gd_{0.74})_{3-w}Mg_w(Ga_{2.48}Al_{2.46})O_{12}$ . Both garnet film scintillators were deposited onto (100) oriented GdGaAG  $(Gd_3Ga_3Al_5O_{12})$  substrates with the Ce content of 1% and the Mg contents w = 0 and w = 0.03, respectively. The thicknesses of both films were about 16 µm. More detailed information about both samples is in our previous paper (Schauer et al., 2019).

- 3. The sample, hereinafter referred to as "LuGdAG:Ce, low Ce&Gd" is a slightly Ce<sup>3+</sup> activated lutetium gadolinium aluminum garnet film with a very low gadolinium content. The data for this film scintillator, like for the next two, including the chemical formula and exact contents of Ce, Gd, and Ga, are in Table 1.
- 4. The sample, hereinafter referred to as "LuGdGaAG:Ce, medium Gd&Ga" is like the previous film scintillator but highly Ce<sup>3+</sup> activated and with only medium gadolinium and gallium content.
- 5. The sample, hereinafter referred to as "LuGdGaAG:Ce, high Gd&Ga" is like the previous film scintillator but with high gadolinium and gallium content.

# 2.2 | Investigated conductive coatings of scintillators

To prevent the scintillator from being electrically charged, and to conduct and measure an excitation current during electron beam characterization, each scintillator was coated with a thin conductive film. The coating also usually helps to increase optical reflectivity, which results in a light signal collection improvement. A thin aluminum (Al) film with a thickness of 50 nm was used at an electron energy of 10 keV, commonly used for electron detection in S(T)EM, and mostly used in this paper. Unfortunately, such standard Al coating absorbs too much electron energy and causes too high losses at low-energy electron detection. Therefore, in these cases, some YAG:Ce single crystal scintillators were also provided with very thin Scandium (Sc), Nickel (Ni), or indium tin oxide (ITO) coatings to investigate their CL properties for low-energy electrons in the range of 0.7-10 keV. All coatings were deposited in a radiofrequency (RF) sputtering unit with 152 mm cathodes in the RF mode using the arrangement schematically drawn in Figure 1. Reactive sputtering was performed in argon and argon-oxygen atmospheres, respectively. The argon and oxygen fluxes were regulated with high accuracy by mass-flow controllers (Tikhonravov, Trubetskov, Hrdina, & Sobota, 1996). The sputtering procedure was carried out similarly as in the paper of Skoupy, Fort, and Krzyzanek (2020). The thickness was tested and calibrated using a Talystep surface profilometer.

## 2.3 | CL characterization

The scintillators' response to the impact of accelerated electrons was studied using the equipment built in our laboratory in Bok and Schauer (2014a). The device is designed to study CL of solids. With

WILEY-

Sample	Film thickness(µm)	х	у	z	Substrate
LuGdAG:Ce, low Ce&Gd	18.0	0.007	0.12	0	LuAG
LuGdGaAG:Ce, medium Gd&Ga	23.0	0.02	2.0	2.0	YAG
LuGdGaAG:Ce, high Gd&Ga	11.1	0.02	3.0	3.3	YGaG

Abbreviation: LPE, liquid phase epitaxy.



FIGURE 1 The setup of the radio-frequency sputtering unit with 152-mm cathodes

this instrument, schematically drawn in our previous paper (Schauer et al., 2019), a collimated e-beam of adjustable energy excited the investigated sample, and a light guide collected the CL emission from the opposite side of the sample. The CL emission spectra were measured in the continual mode using a spectrometer. CL kinetics were measured in the pulsed spectrally unresolved mode using a blanked e-beam. In both modes, a PMT was used as a light detector.

All spectra were measured using an e-beam energy of 10 keV and excitation current of 30 nA, a spot diameter of 2 mm, and a wavelength range of 200-800 nm at room temperature. The Hamamatsu R943-02 PMT was used for spectra detection. All CL spectra were corrected for the spectral response of the apparatus. Unless otherwise indicated, decay kinetics were measured using an e-beam energy of 10 keV and a current of 150 nA, a spot diameter of 2 mm, an excitation time, and excitation frequency of 50 ns and 1 kHz, respectively. The ET Enterprises 9113B PMT and Tektronix DPO7254 oscilloscope were used for decay intensity detection.

### 2.4 **Optical characterization**

The optical properties of film and single crystal scintillators were obtained using the double-beam UV-VIS-NIR spectrophotometer Varian Cary 5. As with the LPE film growth, the single crystal substrates were annealed with subsequent changes in color centers. Measurements were made both in the reference sample mode and nonreference sample mode. In the case of the reference sample measurements, a suitably annealed substrate was used as the reference sample at the LPE film optical transmittance measurement. Reference samples of different thicknesses were used at the single crystal optical transmittance measurement. When measured without the reference sample, the obtained values were corrected to the reflectivity using the refractive index. The optical transmittance of light guides was measured on 50 mm long cylindrical samples. The refractive indices for all samples were obtained with the minimum deviation method (Kuwano, 1978; Kuwano, Saito, & Hase, 1988).

contents tabulated

The optical reflectivity at the internal scintillator-coating boundary was calculated using the matrix method (Knittel, 1976). The optical reflectivity was measured using the spectroscopic reflectometry method to verify these calculated results experimentally (Ohlídal & Navrátil. 1984).

### 2.5 Electron interaction in solids

The MC model for the electron interaction with the matter was used to determine the excitation energy distribution and electron trajectories, including penetration depth in both scintillators and their conductive coatings. The simulation is an extension of the single scattering model utilizing the screened Rutherford cross-section and Bethe slowing-down approximation (Joy, 1995) when secondary processes associated with the diffusion of excited electrons have also been included. The MC model simulated 3D processes associated with interactions of primary electrons in the investigated scintillators and/or conductive coatings. However, the simulation results are presented as 2D projections of 3D processes either to the surface plane of the material (transversal plane) or to any plane with the primary e-beam (longitudinal plane in the depth of the material). In this paper, simulated 3D trajectories of electrons are also expressed by a 2D projection of trajectories in the longitudinal plane along with the material depth.

The algorithms used were compiled for OS Windows as an application called "SCATTER" (Schauer & Bok, 2013). The perpendicular impact of electrons was simulated in this paper, but it is not a problem to simulate an inclined impact. Calculations were done for the different energies of the electron beam that interacted with the studied garnet scintillator and their conductive coatings.

#### 2.6 **Computer-optimized design**

The computer-optimized design (COD) method, intended for nearly any scintillation detection system for S(T)EM, was used to assess and eventually optimize light signal collection at the PMT photocathode. The method uses the MC simulation of photon transport in scintillation detection systems (Schauer, 2007). The application is called SCIUNI and is an extension of the SCINTIL code for rotationally symmetric systems (Schauer & Autrata, 1992). The method uses random photon generation from a luminescent center about 10,000 times and describes the 3D trajectory and the photon transport's efficiency to the PMT photocathode. The value of the light transfer efficiency is given by the probability that the photon will reach the PMT photocathode.

The result of the simulation is the signal transfer efficiency of the scintillation detector. The signal transfer efficiencies can be obtained either as mean values independent of the signal electron impact location or in the form of 3D surface or contour graphs depending on the impact location. The COD method enables the comparison of the signal transfer efficiencies of the different scintillation detectors with specific scintillator or light guide optical properties and optimizes the shapes and sizes of both the scintillators and light guides.

## 3 | RESULTS

## 3.1 | Electron interaction in scintillators

The application SCATTER was used to obtain MC simulation results presented in this section. As mentioned before, the scintillator is an object composed of the conductive coating, scintillation material, and substrate, if used. It makes no sense to study electron trajectories in the conductive coating of the scintillator. Here the electrons are transferred over a negligibly short distance, and such a study is pointless. On the contrary, electron trajectories are significant for bulk single crystals. Therefore, their simulations were performed in the YAG:Ce single crystal and will be presented. However, the electron trajectories have little predictive value for the energy distribution of absorbed electrons, and they are unusable to determine losses due to electron passages through the solid. Nevertheless, simulating the longitudinal distribution of absorbed electrons towards the depth of the material is very useful for assessing the efficiency of a scintillation detector (Schauer et al., 2019). Therefore, the longitudinal distribution simulations were performed for all three components of the scintillator, that is, for the conductive coatings, the scintillators, and their substrates, if necessary.

## 3.1.1 | Excitation electron trajectories

The MC simulation of the 3D electron trajectories projection in the YAG single crystal for the primary electron energies of 2, 5, and 10 keV is shown in Figure 2. This figure of trajectories provides useful visual information on electron interaction volumes and electron penetration depths, but it supplies only inadequate data for analysis of energy deposition. The primary e-beams and the scintillator surface boundary are indicated by the vertical and horizontal lines, respectively. In this case, the zero tilt of the e-beam is simulated, indicated by the normal line to the scintillator surface. Only 100 trajectories were chosen for each simulation in order to be able to distinguish (at least partially) the individual trajectories.

It is evident that with increasing electron energy, the electron penetration depth increases rapidly. At the lowest energy of 2 keV the penetration depth is only 42 nm, at 5 keV, it is 140 nm, and at the maximum simulated energy of 10 keV, the electron penetration depth is as large as 540 nm. The perpendicular electron impact is simulated here, for which the penetration depths are largest. With an oblique impact, the depth decreases with increasing e-beam tilt. Attention must be paid to the penetration depth when using the film scintillators. These results are significant especially when assessing the



**FIGURE 2** The electron trajectories in YAG for the impact electron energy of 2, 5, and 10 keV. One hundred trajectories were simulated for each energy. The perpendicular electron impact was simulated

material: YAG energy: 2 keV, 5 keV,10 keV tilt: 0° 100 trajectories WILEY

<sup>6</sup> WILEY-RESEAR MICROSCOPY

detection of the low-energy electrons, where the processes of electron-photon conversion take place in a thin surface layer of the scintillator and so its surface, including the conductive coating, play a much more critical role than in the detection with the standard energy of 10 keV.

### **Excitation energy distribution** 3.1.2

The same MC simulation method was used to obtain the longitudinal distribution of absorbed electrons towards the scintillator's depth. This distribution is of primary importance when assessing the appropriate thickness of the scintillator. Replacing a standard bulk single crystal scintillator with a thin scintillation film in an electron detector in S(T)EM involves the risk that the penetration depth of the electrons will be higher than the thinness of the scintillation film.

The longitudinal distribution of absorbed electron energies in the YAG:Ce single crystal and the LuGdGaAG:Ce film scintillator for the primary electron beam of 2, 5, and 10 keV are shown in Figure 3. To reduce the statistical errors of this simulation, the total number of the primary electrons simulated should be at least 10<sup>3</sup>. It is challenging to obtain the simulated value of the energy deposited near the scintillator's surface since these values are strongly affected by surface effects such as secondary electron (SE) and BSE emission. Therefore, the energies deposited at the surface were dropped from processing. Evidently, if the simulation does not include all processes following electron interaction and diffusion that are important, especially for the low e-beam energy, these results are only of limited predictive value. Concerning this, the results of the simulation should be understood as a rough estimate. However, it is sufficient for estimating the required scintillator thickness.

From the results in Figure 3, it is clear that even for the highest energy of 10 keV, which is typical for detection in S(T)EM, the maximum of energy losses in the LuGdGaAG:Ce film scintillator lies already at 125 nm, and the total interaction energy is absorbed to the depth of 500 nm. At the same time, the typical thicknesses of the studied scintillators are at least 20 times larger. Thereby, all energy is deposited in the scintillator, and the substrate is not excited. Therefore, there is no need to worry about insufficient scintillator thickness. The film scintillators may be even more than 10 times thinner than those studied.

The MC simulation of the longitudinal distribution of absorbed energy was performed not only for the scintillators but also for their conductive coatings. The aim was to determine a reduction in the scintillation detector efficiency due to energy absorption in this conductive coating. The electron energy loss distributions in the 5 nm thick Al, 3nm and 5 nm thick Sc, and 4 and 7 nm thick ITO films, all for the 0.7 and 5 keV e-beam energy, respectively, are shown in Figure 4. The coating with a thickness of only units of nanometers has to be used to allow the low-energy electron penetration through the conductive coating without significant losses.

However, using ultrathin films in the order of units of nanometers, problems can occur with scintillator charging. In such a case, it is tremendously challenging to guarantee sufficient conductivity of the scintillator surface so that it can efficiently collect the signal electrons. Ensuring sufficient conductivity is difficult even though all conductive materials studied have a high electrical conductivity. Even worse, the extremely thin films show almost no optical reflectivity. All the mentioned problems will be discussed in more detail in connection with energy conversion in the scintillator in Section 3.2.2. In any case, lowenergy electron detection requires special attention.



FIGURE 3 The Monte Carlo (MC) simulation of the longitudinal distribution of electron energy losses in the YAG:Ce single crystal and LuGdGaAG:Ce film scintillators for the different incident electron energy



FIGURE 4 Electron energy loss distributions in the 5-nm thick Al conductive coating, 3- and 5-nm thick Sc films, and 4- and 7-nm thick indium tin oxide (ITO) films, all for the 0.7-keV, and 5-keV e-beam energy, respectively

## 3.2 | Energy conversion in scintillators

CL characterization in this section was performed using the experimental equipment described in Section 2.3. Again, it should be noted that the scintillator here means not only the scintillation material itself but also the conductive coating on its surface and eventually also its substrate if used. All measurements were performed including the coating and the substrate if the film scintillators are considered. No corrections were made to the effect of the coating and substrate. However, all CL measurements are only relative (in arbitrary units of the CL intensity), so this approach is satisfactory. The results of the study of the garnet scintillator efficiency and kinetics and the effect of the coating when detecting the low-energy electrons are presented in this section.

## 3.2.1 | Overview of garnet scintillator efficiency

In the past, inadequate importance was given to the scintillator efficiencies without any relation to noise (Autrata et al., 1978; Autrata et al., 1983; Bril, Blasse, & Poorter, 1970; Hatzakis, 1970; Robbins, Cockayne, Lent, Duckworth, & Glasper, 1979). It is not necessary to overestimate the efficiency itself. However, it affects DQE, so it cannot be completely ignored. In this paper, only the most essential overview of the CL intensity results from our laboratory will be given. Other results can also be found in our previous papers (Schauer et al., 2017; Schauer et al., 2019). A comparison of the garnet CL intensities and maximum CL emission wavelengths is shown in Table 2. The values for the standard YAG:Ce single crystal and five Ce<sup>3+</sup> activated garnet film scintillators are tabulated.

The emission spectra of all mentioned scintillators with a relative CL intensity scale are shown in Figure 5. The results were measured in the wavelength range from 200 to 800 nm at room temperature. Subsequently, the results were corrected for the device spectral transmittance and detector spectral sensitivity. Significantly,  $Ce^{3+}$ -related 5d-4f emission is seen in the CL spectra of all the studied scintillators. It is also important to observe that the YAG:Ce single crystal, unlike all the film scintillators studied, exhibits broad UV CL emission at 250–400 nm. This emission is associated with a different preparation

of the LPE film scintillators, which will be explained in more detail in connection with CL kinetics in Section 3.2.3.

MICROSCOPY

It can be seen that most of the studied film scintillators show only a slightly different CL intensity related to the standard YAG:Ce single crystal. The mentioned difference will be discussed in Section 4. Complete 5d-4f emission bands of all studied scintillators lie in the wavelength range from about 440 to 800 nm with the maximum emission around 540 nm. Emission in this spectral region has the significant advantage that no demands are placed on the UV transmittance of the light guide and the UV sensitivity of the photocathode. The absence of UV emission allows using much cheaper light guide materials. It also dramatically expands the choice of a cheap PMT, although broadband yellow emission of the garnets is not ideal spectral matching to the standard photocathodes.

Figure 6 shows the measurement results of the sensitivity spectra of the most common multialkali (S20) and SbCs (S11) PMT photocathodes often used in the S(T)EM scintillation detectors. The almost ideal GaAs photocathode is shown as the reference one. The maximum sensitivity of the most standard photocathodes is in the spectral



**FIGURE 5** Cathodoluminescence emission spectra of the standard YAG:Ce single crystal and five Ce<sup>3+</sup> activated garnet film scintillators. Intensity values are corrected for the device spectral transmittance and detector spectral sensitivity

S20 PMT photocathode for the standard YAG:Ce single crystal and the five Ce <sup>3+</sup> activated garnet film scintillators					
Scintillator	Scintillator form	Relative CL intensity <sup>a</sup> (%)	$\lambda$ of max. CL emission (nm)	S20 PMT matching <sup>b</sup> (%)	
YAG:Ce	Crystal	95	532	73	
LuGdGaAG:Ce (high Ce)	Film	100	544	80	
LuGdGaAG:Ce (high Gd&Ga)	Film	100	540	81	
LuGdGaAG:Ce,Mg (high Ce)	Film	76	546	79	
LuGdGaAG:Ce (medium Gd&Ga)	Film	67	539	81	
LuGdAG:Ce (low Ce&Gd)	Film	82	521	84	

**TABLE 2** The CL intensity, the wavelength of the maximum CL emission, and the spectral matching of the scintillator to the sensitivity of the S20 PMT photocathode for the standard YAG:Ce single crystal and the five Ce<sup>3+</sup> activated garnet film scintillators

Abbreviations: CL, cathodoluminescence; PMT, photomultiplier tube.

<sup>a</sup>Plot areas in the range of 430-800 nm compared, corrected for the device spectral response.

<sup>b</sup>Compared to ideal (theoretical) photocathode.

WILEY



**FIGURE 6** Spectral response of the most common multialkali (S20) and SbCs (S11) photomultiplier tube (PMT) photocathodes often used in the scanning and/or scanning transmission electron microscope (S(T)EM) scintillation detectors. The almost ideal GaAs photocathode is shown as the reference one

region around 420 nm. This property is a disadvantage for garnet scintillators having the emission band of 440–800 nm with the maximum around 540 nm. It can be seen in Figure 6 that the photocathode S11 has a lower sensitivity in the garnet emission spectral region. Therefore, it is necessary to use S20 red extended photocathode with the garnet scintillators. Scintillator spectral matching to the S20 photocathode is also shown in Table 2. The PMT matching here is given in relation to the theoretically ideal PMT photocathode. As is shown, spectral matching of the garnet scintillators to the S20 photocathode is quite favorable, with values of 73–84% related to the ideally matched photocathode. Bad spectral matching of the scintillators to the PMT photocathode can significantly reduce the scintillation detector efficiency.

# 3.2.2 | Influence of conductive coating of scintillator

The conductive coating causes losses due to the absorption of the low-energy electrons, losses due to possible scintillator charging, and losses due to extremely reduced optical reflectivity of the coating. It is difficult to distinguish the effect of all individual losses experimentally. However, this is unnecessary. The experimental determination of the influence of the conductive coating on the scintillator efficiency was performed in the beam energy range 0.7–10 keV using the experimental equipment described in Section 2.3. The comparison of the relative CL efficiency (light yield/keV) dependence of the YAG:Ce single crystal scintillator with various thin conductive coatings on the energy of incident electrons is shown in Figure 7. The investigated scintillator was coated with the 25 and 45 nm thick Al, 3 and 5 nm thick Sc, and 4 and 7 nm thick ITO film, respectively.



**FIGURE 7** The relative cathodoluminescence (CL) efficiency (light yield/keV) dependences of the YAG:Ce single crystal scintillator with various conductive coatings on the energy of incident electrons

An overview of the parameters of selected coatings is in Table 3. The atomic numbers, atomic weights, densities, thicknesses, and transmissions of 700-eV electrons are tabulated. Electron transmission was determined by the MC simulation described in Section 2.5 and used and discussed in Section 3.1.2. The MC simulation for the Al coating was performed for a larger and different set of thicknesses compared to the experimental set of Figure 7. The electrical resistivity values have a certain instability in such ultrathin film in air, which is given by surface oxidation. Therefore, the values given in the table are for guidance only. They are to be understood as the maximal rounded measured values after coating stabilization. The aim was not to compare the AI coatings in the standard electron detector with the electron energy of 10 keV but in the low-energy electron detector. Unfortunately, often, no attention is paid to low-energy electron detection. If a scintillator with a standard 45-55 nm Al is used, the low-energy detection efficiency is very low. At the same time, there are ways to fix it.

The results show that the traditional AI coating (standard thickness of 45–55 nm) is excellent for standard SE and BSE electron detection in S(T)EM with a standard electron energy of around 10 keV. However, when detecting electrons with the energy of around 2 keV, the detectors with AI already lose about 96% efficiency. So, AI is practically useless at this electron energy. At the standard electron energy of 10 keV, the ITO coating causes up to half the efficiency compared to AI. In such a case, the transparency of the ITO coating has a negative effect when the light's total reflection is applied only in limited directions. However, for the 2-keV electrons, the ITO coating is better. When detecting the electrons with this lower energy, ITO guarantees six to seven times higher efficiency than AI. However, AI and ITO coatings are entirely unusable for the detection of low-energy electrons with energies below 1 keV. For such detection, the Sc coating with a thickness of 5 nm

Film	Atomic number	Atomic weight	Density (g cm <sup>-3</sup> )	Thickness (nm)	Electrical resistivity (Ω/sq)	700 eV electron transmission (%)
Aluminum	13	26.98	2.70	3.8	<1,000	63.2
(AI)				5.0	<500	48.0
				10.0	<100	15.1
				15.0	<50	1.3
Scandium	21	44.96	2.99	3.0	<10,000	69.0
(Sc)				5.0	<10,000	45.1
ITO	24.21 <sup>a</sup>	55.11 <sup>a</sup>	7.16	4.0	<10,000	6.24
				7.0	<10,000	0.0

**TABLE 3** Some parameters of scintillator coatings for low-energy detection. The electrical resistivity must be understood as the maximal rounded measured value after coating stabilization

Abbreviations: CL, cathodoluminescence; ITO, indium tin oxide. <sup>a</sup>Weighted values.

has proven successful. It is even comparable to the Al coating for energies of 10 keV. Furthermore, as the only film examined, Sc is applicable for detecting electrons with energies below 1 keV. All studied coatings have sufficient electrical conductivity to prevent scintillator surface charging.

## 3.2.3 | CL kinetics of garnet scintillators

The results in this section will be limited to an overview of the five selected film scintillators compared to the reference YAG:Ce single crystal. The issue of scintillator kinetics was sufficiently published, unlike other important processes in the detector. It has also been studied in detail in many of our previous works (Kucera et al., 2016; Schauer, 2011; Schauer et al., 2017; Schauer et al., 2019).

The decay characteristics of the garnet scintillators described in Section 2.1 were measured at room temperature. The corrected results for the pulse width and instrument response function are shown in Figure 8. The measurement was performed at the e-beam energy of 10 keV, excitation pulse width and current of 50 ns and 150 nA, respectively, the e-beam spot diameter of 2 mm and excitation repetition frequency of 1 kHz. Fittings of the experimental decay curves were carried out, assuming the presence of tunneling recombination using the sum of exponential and power functions (Schauer et al., 2017). The presented decay characteristics are normalized to unity at the beginning of the decay. Such a graphical representation with a multiorder dynamic range allows the determination of both the scintillator decay times (i.e., the time when the intensity drops to 1/*e*) and afterglows.

The decay time and afterglow values taken from the decay characteristics in Figure 8 are given in Table 4. The dynamic range of the CL device was  $10^{-4}$ . Therefore, the afterglow of less than 0.01% could not be measured. It can be seen that the LuGdGaAG:Ce (high Gd&Ga) and LuGdGaAG:Ce,Mg (high Ce) film scintillators have extremely low decay times. Their values are about three times lower than those of YAG:Ce single crystal. Even all the other film scintillators have a lower



**FIGURE 8** The decay characteristics of the film scintillators and the reference YAG:Ce single crystal scintillator measured at room temperature for the e-beam energy of 10 keV. The measurement results were corrected for the pulse width and the instrument response function (IRF)

decay time than YAG:Ce. At the same time, most film scintillators show a much smaller afterglow at 2  $\mu$ s after excitation of the order of 0.01% and lower than the YAG:Ce single crystal with the afterglow of about 0.3%. Only the LuGdAG:Ce (low Ce&Gd) film scintillator has a higher afterglow than the YAG:Ce single crystal. As previously expressed in introductory Section 1, in the S(T)EM imaging systems, the afterglow must be low even at the microsecond time range (after the excitation), which nearly all investigated film scintillators meet. Compared to the standard YAG:Ce single crystal scintillators, which exhibit a decay time and afterglow (2  $\mu$ s after excitation) of 86 ns and 0.3%, respectively, all studied films provide a significant improvement in CL decay kinetics. These results are related to the absence of the antisite defects in the LPE film scintillators mentioned in Section 3.2.1.

Scintillator	Material form	Decay time (ns)	Afterglow at 2 μs (%)
YAG:Ce	Crystal	86	0.3
LuGdGaAG:Ce (high Ce)	Film	60	<0.01
LuGdGaAG:Ce (high Gd&Ga)	Film	22	<0.01
LuGdGaAG:Ce,Mg (high Ce)	Film	27	0.02
LuGdGaAG:Ce (medium Gd&Ga)	Film	37	<0.01
LuGdAG:Ce (low Ce&Gd)	Film	60	0.5

**TABLE 4**The values of the decaytime and afterglow of the filmscintillators, and the reference YAG:Cesingle crystal scintillator measured atroom temperature for the e-beam energyof 10 keV. A dynamic range of the CLdevice was 10<sup>-4</sup>; therefore, the afterglowof less than 0.01% could not bemeasured

Abbreviation: CL, cathodoluminescence.

**TABLE 5** Electrical resistivity and optical reflectivity of Al and ITO conductive coatings

			Calculated inter	nal optical reflectivi		
Film	Thickness (nm)	Electrical resistivity ( $\Omega$ /sq)	Tilt 0°	Tilt 30°	Tilt 60°	Measured optical reflectivity (%)
Al	50	<10	85.7	85.7	84.8	84
	35	<10	84.6	84.6	84.0	83
ITO	10	<10,000	8.4	17.8	94.2	8

Abbreviation: ITO, indium tin oxide.

# 3.3 | Utilization of light signal from scintillator luminescence centers

In many publications, when studying the performance of a scintillation electron detector in S(T)EM, much more space is devoted to the energy conversion in the scintillator than the utilization of the light signal generated (Everhart & Thornley, 2004; Frank, 2002; Healy & Mott, 2016; Nedela, Tihlarikova, Runstuk, & Hudec, 2018). The detection of the photons from the scintillator centers is indeed an extremely fast process, so it does not degrade the MTF of the detector. However, the process can be very inefficient. Therefore, in this section, attention will be paid to the relevant optical properties of the scintillators, light guides, optical cement, and other components used. Attention will also be paid to the typical light-guiding configurations and the COD of the light-guiding system.

## 3.3.1 | Optical properties of scintillators

The study of light signal transfer from the luminescent centers to the PMT photocathode is impossible without knowledge of optical absorption and the refractive index of the scintillator. Also, knowledge of the optical reflectivity of the conductive coating from the scintillator internal side is a benefit because it can cause a loss of efficiency. The internal reflectivity of the Al and ITO films was evaluated using the matrix, and experimentally verified results were measured by the spectroscopic reflectometry, both methods are specified in Section 2.4. The results are shown in Table 5. The reflectivity was calculated for the perpendicular incidence and the tilts of 30 and 60°, respectively. The measured results are only for perpendicular light beam incidence. All reflectivity results are for the monochromatic light of 550 nm.

The study of electrical resistivity and internal reflectivity was reduced to the Al and ITO coatings with thicknesses of 50, 35, and 10 nm, respectively, which are suitable for detecting electrons with the standard energy of 10 keV. The obtained electrical conductivity values in Table 5 are sufficient to remove the charge of the detected electrons. The calculated and measured values of Al reflectivity are at the maximum of their theoretical values of about 86%. The reflectivity of the transparent ITO coating is given by the Fresnel reflection and transmission coefficients, not by the specular reflection of the nontransparent metals. Therefore, the ITO reflectivity is extremely dependent on the incidence tilt, and it is advantageous only for very oblique light incidence. The given reflectivity values are the input parameters of the SCIUNI COD application.

High optical self-absorption and/or the high refraction index of the scintillator can significantly reduce the efficiency of the entire scintillation detection system in S(T)EM. The results of the optical absorption coefficient measurements of the LuGdGaAG:Ce (high Gd&Ga), LuGdGaAG:Ce,Mg (high Ce), LuGdGaAG:Ce,Mg, and LuGdGaAG:Ce (high Ce) film scintillator, of the GdGaAG single crystal substrate, as well as of the YAG:Ce single crystal scintillator, are shown in Figure 9. The results shown were obtained using the double-beam spectrophotometer, as described in Section 2.4. The absorption coefficients of the scintillators are shown in the spectral range of 350–800 nm.

Figure 9 shows that all studied scintillators exhibit significant optical absorption only in the spectral region of 400–500 nm. The absorption peak of all the film scintillators is about four times higher than that of the standard YAG:Ce single crystal scintillator. However, to assess self-absorption, it is more important that the absorption of the studied garnet scintillators is very low in the rest of the visible spectrum, where the emission band of the garnet scintillators lies. The optical absorption of the GdGaAG, LuAG, YGG, and YAG substrates is very low (shown on a

different scale, magnified 100 times) in the entire measured spectral region, and it is almost without the peaks. It is imperative and advantageous that the optical absorption of all substrates is extremely low.

The high refractive index of the scintillator may critically deteriorate the photon escape from its exit surface due to the low critical angle for the total reflection. Therefore, the refractive indices of the film scintillators, YAG:Ce single crystal scintillator and the YAG and GdGaAG single crystal substrates were measured. The results were obtained with the minimum-deviation method, as described in Section 2.4, and they are shown for the spectral range of 350–800 nm in Figure 10. The refractive indices of the LuAG and YGG substrates, and the LuGdGaAG and YAG ones are almost the same, respectively. Therefore, the LuAG and YGG substrates are not plotted. Fortunately, also the refractive indices are important only in the visible spectrum around the wavelength of about 540 nm, where the emission band of



**FIGURE 9** The spectral dependence of the optical absorption coefficients for the film scintillators and the reference YAG:Ce single crystal scintillator



**FIGURE 10** The spectral dependence of the refractive indices for the film scintillators and the reference YAG:Ce single crystal scintillator

the garnet scintillators lies. In this part of the spectrum, the refractive indices of the studied scintillators are relatively low and not very different. Therefore, all the scintillators studied have nearly the same application potential in this respect.

## 3.3.2 | Optical properties of light guides

The light guides are the essential parts of the scintillation detector regarding the efficient transport of photons from the luminescent centers to the PMT photocathode. Attention must also be paid to optical cement used for coupling the light guide to the scintillator and the PMT. As with the scintillator, the light guide must possess high optical transmittance in the spectral region of the scintillator emission band, that is, around 540 nm in the case of the garnet scintillator.

The most commonly used light guide materials for the scintillation electron detectors in S(T)EM are transparent thermoplastics and various silicate glasses based on silicon dioxide, or quartz. During the examination of the scintillation detectors in our laboratory, the optical polymethyl methacrylate (PMMA), commercial PERSPEX, commercial Merci silicate glass, and quartz light guides were studied and tested. The optical transmittance of the mentioned 50-mm long cylindrical samples was measured using the method described in Section 2.4, and the results are shown in Figure 11. The measured values were corrected to reflectivity using the refractive indexes.

Evidently, the Merci silicate and quartz glasses have much higher UV transmittance. The two more expensive materials have a transmittance higher than 97% in the whole studied spectral region. However, in the region above 500 nm, all tested materials have almost the same optical transmittance of about 98%. At the wavelength of 420 nm, where garnet emission is already beginning, PMMA and PERSPEX have a lower transmittance of only about 2%, which is no major problem for light signal collecting from the garnet scintillators using relatively inexpensive thermoplastic light guides. Thus, the PMMA light



**FIGURE 11** The spectral dependence of the optical transmittance of the quartz, Merci silicate glass, polymethyl methacrylate (PMMA), and PERSPEX light-guide material

WILEY

guides will always be simulated in the following sections dealing with the geometric configuration of light guides.

The refractive indices of all studied light guides are lower than that of the scintillators or their substrates. Such a relationship is a suitable feature for good optical coupling to scintillators. The refractive index of the silicate glasses is about 1.54, while that of PMMAs is only slightly lower with a value of 1.49. This is the main reason why the study of the light guide refractive index was abandoned. The study of optical properties of possible optical couplers in the scintillator-light guide-PMT system was also abandoned. Various types of optical cement and other immersion couplers are commercially available, and the only essential condition in their selection is that the refractive index of the coupler is within the refractive indices of the coupled components.

## 3.3.3 | COD of light-guiding system

It is practically impossible to determine the optimal choice of material, shape, and size of both scintillators and light guides for particular

detector without a computer application. Certainly, it is much easier to collect the light signal using the base-guided signal (BGS) detection systems. In addition, such systems show the highest degree of symmetry and place the lowest demands on the machinability of both the scintillator and the light guide. However, the BGS systems are suitable only for SE detection. The edge-guided signal (EGS) detection systems must be used when detecting BSEs in S(T)EM. When designing the system, it is also necessary to distinguish the machining of the scintillator exit surface and the existence of a hole in the scintillator.

The results in this section were obtained using the SCIUNI COD application described in Section 2.6 that guarantees high-quality statistical results. For each configuration, 10,000 randomly generated directions were simulated at regularly spaced locations on the scintillator surface. Using SCIUNI, the efficiencies depending on the electron impact location on the scintillator surface were expressed in the form of the 3D surface graphs. At first, simulations of three BGS detection systems were performed. The results for the (a) disc-shaped scintillator, (b) hemisphere-shaped scintillator, and (c) cone-shaped scintillator are shown in Figure 12. The light guide material of all



**FIGURE 12** The light-guiding efficiency of the BGS rotationally symmetric detection systems with the YAG:Ce scintillator in relation to the coordinates of the electron impact. (a) Disc scintillator with the matte exit surface and with other surfaces polished, no optical cement is used, (b) hemisphere, and (c) cone scintillator with all surfaces polished and optical cement used for the coupling of the scintillators. Note the different efficiency scaling compared to the next figure

rotationally symmetric ET detection systems was PMMA. All three YAG:Ce scintillators were coated with Al. Evidently, the cone-shaped scintillator in the BGS system is advantageous only if an electron collection system can collect close to the detector axis. Otherwise, the disc-shaped scintillator is better. However, it is necessary to apply a matte finish to the disc scintillator exit surface. The BGS systems with hemisphere-shaped scintillators have the lowest efficiency.

It is much more challenging to design detectors for BSEs based on the EGS systems. These systems cannot be designed with rotational symmetry. All simple (nonoptimized) EGS systems are extremely inefficient. Using EGS, it is necessary to transport the light signal to the light guide from a relatively narrow edge of the scintillator. MC simulations of the nonoptimized EGS systems and the perfectly optimized one are shown in Figure 13. The simulations (a) and (b) show very simple nonoptimized BSE detection systems with a cylindrical hole for an electron beam in the scintillator. The configuration utilizes the (a) inefficient strip light guide, while the more efficient configuration (b) same scintillator but with the cylindrical light guide having widening planes for adapting the light guide to the scintillator. The simulations (c) and (d) show the more complex BSE detection systems for the Hitachi S-4000 FE-SEM with a conical hole for an electron beam in the scintillator. The configuration (c) represents the rough (nonoptimized) design, while the configuration (d) represents the optimal final geometry. In (a) and (b), the YAG:Ce single crystal scintillators of  $\phi$  20 mm  $\times$  2.0 mm, and in (c) and (d) of  $\phi$  15 mm  $\times$  2.5 mm with the ITO coating, were simulated. In (a) and (b), the cylindrical hole in the scintillator of  $\phi$  3 mm, and in (c) and (d), the configurations, the



**FIGURE 13** The light-guiding efficiency of the edge-guided signal (EGS) detection systems in relation to the coordinates of the electron impact on the surface of the YAG:Ce single crystal disc scintillator cemented to a polymethyl methacrylate (PMMA) light guide. The simulations (a) and (b) show very simple nonoptimized backscattered electron (BSE) detection systems with a cylindrical hole for an electron beam in the scintillator. The configuration (a) utilizes an inefficient strip light guide, while the more efficient configuration (b) utilizes the same scintillator but with a cylindrical light guide having widening planes for adapting the light guide to the scintillator. The simulations (c) and (d) show the more complex EGS detection system for the Hitachi S-4000 FE-SEM with a conical hole for an electron beam in the scintillator. The configuration (c) represents the nonoptimized design, while the configuration (d) represents the final design. Note the different scaling of the efficiency axes compared to the previous figure

WILEY

14 WILEY-RESEAR MICROSCOPY

length of the detection system was 80 mm, and in (c) and (d), 150 mm. The light guide material was PMMA. The scintillator was cemented to the light guide. It is seen that all EGS systems are less efficient than the BGS ones. Although the efficiency of the BGS system is nearly independent of the cylindrical light guide size, the efficiency of the EGS detection systems is strongly dependent on the size and mainly on the shape of the light guide, even if the material, size, and shape of the scintillator are the same. It happens because a slit light-guiding profile has to expand to a wide circular one. The consequence can be seen in the enormous efficiency difference between the very simple (a) and (b) EGS configurations in Figure 13.

The presented simple strip systems do not achieve efficiencies higher than about 4% at no point in the scintillator impact surface. The efficiencies from the impact points attached to the light guide are up to 15 times lower compared to that from the system beginning. The simple widening nonoptimized systems are better because they can straighten the photons' trajectories towards the photocathode, but not from all points of the scintillator surface. With these systems. light collection from the scintillator locations attached to the connected light guide is much more efficient. In the presented widening simple EGS system, the point efficiency reaches up to 20%. However, the efficiency towards the beginning of the system gradually decreases to a value, which is six times lower. Nevertheless, such inefficient nonoptimized systems, which degrade the efficiency of the entire BSE detector, are often used in S(T)EM.

However, COD applications can significantly increase the efficiency of the BSE detectors utilizing the EGS system. The designing of the BSE scintillation detector for the Hitachi S-4000 FE-SEM was chosen to demonstrate the SCIUNI application's capability. In Figure 13, the configuration (c) represents the initial rough (nonoptimized) design. while the configuration (d) represents the final optimized result. Based on the size and shape of the pole pieces, the specimen holder, and the size and position of the PMT of the microscope, the initial design was determined by the fixed size, shape, and material of the scintillator and light guide. For the initial configuration, the simulated mean efficiency was only about 4%. After shifting the widening plane of the light guide, optimizing their angles (equal reduction) and integrating a conical lightguiding ring, the efficiency increased 400 times. Additionally, the efficiency uniformity across the scintillator impact surface is much higher in the final configuration. In general, a gradual widening of the EGS light guide is more advantageous, but this is not relevant close to the scintillator coupling.

### DISCUSSION 4

Unlike many other publications on the garnet scintillators, which focus on physical processes in scintillation material, this paper aims to determine the performance of the S(T)EM scintillation detector. The scintillator in S(T)EM forms not only scintillation material but also the unavoidable anti-charging conductive coating on its surface and substrate, if necessary. The light signal in the S(T)EM scintillation detector, which arises during energy conversion in the scintillator, is determined not only by the CL intensity in the luminescence centers but also by the efficiency of the light signal guiding to the PMT photocathode.

From the results, it is evident that it is not possible to ignore the above facts. If the scintillators are not provided with the conductive coating, they will not be able to conduct the obtained charge and will be charged. Moreover, they will not reflect the light signal to the scintillator exit surface. There is no problem with the coatings with a thickness of tens of nanometers for the electron energy in the tens of kiloelectron volts, but for low-energy detection around 0.7 keV, the coatings must only be as thin as units of nanometers to allow electron penetration without significant losses. Unlike thicker coatings, in such a thin coating, the electrical conductivity and the optical reflectivity are very low. Although the effects of the conductive coatings on the scintillator efficiency (electron absorption in Figure 4 and optical reflectivity in Table 5) have been studied separately, this is not necessary. The unresolved efficiencies in Figure 7 and Table 3 are important because they prove that AI coating is not applicable for the electron energy of 0.7 keV. Only Sc will ensure that the detector efficiency is not completely destroyed.

The efficiency of the S(T)EM scintillation detector is not, unlike its response, determined only by the processes in luminescence centers. In addition to the processes in the luminescence centers and the conductive coating already mentioned, the efficiency is also greatly influenced by the light-guiding system. As for the scintillators, the results in this paper prove that all studied garnet materials' efficiencies do not differ much. In our older paper, it was calculated that the theoretical limit of the YAG:Ce conversion efficiency is about 23 photons per 1 kV electron (p/keV) (Schauer et al., 2019). Unfortunately, there is nearly no opportunity to improve the conversion efficiency if the value of 19 p/keV was obtained. The reason is that the theoretical limit can never be achieved because it is calculated for the ideal crystal, but it is practically impossible to produce the garnet scintillator without impurities and defects. Fortunately, it is not necessary to exaggerate the CL efficiency of the scintillator. Crucially, all garnet scintillators have a sufficient CL signal-to-noise ratio, as can be seen in Figure 8. Based on this, the detector will have satisfactory DQE, which is a priority.

The big difference in assessing the performance of the LPE garnet film scintillators is the fact that their thicknesses are only in the range of 10-18 µm, and they cannot exist without the light-guiding substrate. The efficiencies of the film scintillators could be lost if the signal electrons penetrate the substrate. Signal penetration into an inactive material is a drawback for many scintillation radiation detectors where a high penetration depth exists. However, in the S(T)EM scintillation electron detector, the penetration depth even for the higher signal electrons of 10 keV is less than 1  $\mu$ m, as demonstrated in Figure 3. As will be discussed later, the great advantage is the low self-absorption of the light signal in such a thin scintillator.

The key priority in the S(T)EM scintillation detector is to use an extremely fast scintillator. The response of the whole detection system in S(T)EM is determined only by the processes in the luminescence centers. The response is not affected by the conductive coating or the processes related to photon collection, not even by the PMT properties if it is selected correctly. Therefore, the decay characteristics in Figure 8 are fundamental. The short rise and decay time and low afterglow guarantee a strong MTF of the entire imaging system. The MTF can be calculated utilizing the method of scintillation rise and decay characteristics (Bok & Schauer, 2014b) if the scintillators are excited by a 50-ns electron pulse using the spectrally unresolved experimental arrangement. The calculation represents the contrast transfer during the scanning with a dwell time of 50 ns. MTFs are not presented in this paper, but attention is paid to this in our previous paper (Schauer et al., 2019). From the decay characteristics in Figure 8, it can be proven that the detector with the fastest garnet film scintillators studied allows the imaging system in S(T)EM to have a much better resolution than that with the YAG:Ce scintillator. As previously calculated, the studied film scintillators lose the contrasttransfer ability at small details above 0.6 lp/pixel (line pairs per pixel), while the YAG:Ce single crystal scintillator does so already above 0.1 lp/pixel.

The presented results of the electron-photon conversion in the garnet film scintillators in Figures 5 and 8 show that a strategy to get significantly faster CL decays of the scintillators can be based on the creation of an additional fast radiative recombination pathway, as was discussed previously (Kucera et al., 2016; Nikl et al., 2014; Schauer et al., 2017). Unlike the Czochralski-grown single crystal garnets, the LPE garnet films possess no antisite defects that are responsible for the broad UV CL emission at 250–400 nm in Figure 5. It is caused by lower preparation temperatures during the LPE films' growth, which contributes to faster recombination in these scintillators. The antisite defects are responsible for the slow CL decay components of the single crystals and, above all, for their high afterglow in the microsecond time range in Figure 8. Developers of the scintillation electron detectors for S(T)EM have been faced with this problem for many years (Schauer, 2011).

Unfortunately, comparing the results of the CL intensity in Figure 5 and CL decay in Figure 8 shows that there is a relationship between the efficiency and response rate of the garnet scintillators. A faster scintillator response generally cannot be achieved without some loss of the CL intensity. This rule does not follow the Ce highly activated and Mg co-doped LuGdGaAG:Ce,Mg film scintillator. This scintillator shows a significant improvement in the response rate without loss of efficiency. The positive effect of Mg co-doping is evident in this scintillator because its decay time of 28 ns, the extremely low afterglow of 0.02% at 2  $\mu$ s, and its unreduced efficiency are by far the best properties ever reported for garnets. Nonetheless, in general, if the response is preferred, which is the priority in the S(T)EM scintillation detector, the reduced scintillator efficiency must be tolerated. However, this fact is acceptable, as the DQE of the detector will not be deteriorated much, while the MTF of the detector will be significantly improved. In contrast to the garnet scintillators' efficiency, which is almost close to the theoretical limit, there are still some untapped potentials in the garnet scintillators' response rate. Possibilities to improve the response time are evidenced by many recent results (Nikl & Yoshikawa, 2015).

MICROSCOPY

The processes of collecting and directing the light signal to the PMT photocathode do not affect the response rate of the scintillation detector but influence its efficiency considerably. Unfortunately, this issue is often overlooked, and only a few publications deal with the processing of the light signal in the S(T)EM detector (Danilatos, 2012; Salomoni, Pots, Auffray, & Lecoq, 2018; Schauer, 2007; Schauer & Autrata, 1979, 1992; Yamamoto, Tanji, Hibino, Schauer, & Autrata, 2000). Each scintillator is also a light guide for the light emitted from the luminescence centers guided to its exit surface. Thus, the optical properties of the scintillator and its conductive coating are of great importance. As for the coatings, contrary to low-energy detection, the high optical reflectivity of coatings is preferred at the standard electron energy of 10 keV. For 10 keV, the Al and ITO coatings with a thickness of 35-50 nm, and 10 nm, respectively, are suitable. In such a case, directionally independent Al reflectivity is at its theoretical maximum. Therefore, Al is more versatile and advantageous under these conditions. The ITO coating is better only for the low-energy electrons and for the light directed along the system axis. In the case of the film scintillator, the substrate's optical properties are also of great importance. Knowledge of optical parameters is necessary not only for assessing the quality of the light signal collection but especially for quantitative calculations. In particular, the values of the optical absorption coefficients, the refractive indices, and the mirror optical reflectance of all materials involved in the light transport to the PMT photocathode, must be known. Otherwise, it would be impossible to simulate light transport processes. Therefore, the results given in Sections 3.3.1 and 3.3.2 are crucial for the assessment of signal utilization. Even without MC simulations of light transport, these results are relevant because they immediately reveal unsuitable material for the successful light collection.

In order to assess scintillator self-absorption, absorption of the studied garnet scintillators must be very low in the region of their CL emission. As for self-absorption of the film scintillators, they have a larger Stokes shift (i.e., the difference between the absorption and emission peak positions) than the standard YAG:Ce single crystal. It is also imperative and advantageous that optical absorptions of all the substrates are the lowest. Furthermore, photon paths in the film of maximally 20  $\mu$ m are much shorter than those in the bulk scintillator. All these facts are very advantageous in terms of garnet film scintillator self-absorption. Also, the refraction indices of the presented scintillators of about 1.84 at the CL emission of 540 nm guarantee nearly the same application potential.

The light guides must not be omitted due to the efficient photon transport from the luminescent centers to the PMT photocathode. At the wavelength of 420 nm, where garnet emission is already beginning, the transmittance of PMMA and PERSPEX light guides is only about 2% lower than costly UV light guides with almost the same optical transmittance of about 98% in the spectral region above 500 nm. Therefore, in terms of optical transmission, there is no reason to use much more expensive UV light guides for the garnet scintillators. The refractive indices of all presented light guides are slightly lower than the refractive indices of the scintillators or their substrates, which is satisfactory. For these reasons, it is more advantageous to use easy machinable PMMA light guides.

-WILEY-

MICROSCOPY

However, when configuring a light guide, there is a big problem when designing its size and shape. The only reliable way is to use a suitable COD application that quickly evaluates the light-guiding system and can optimize its performance. Compared to experimental design, COD saves a lot of time and money and results in an incomparably better detector. Our SCIUNI COD application proves that nonoptimized systems have very poor light-guiding efficiency. The results show that even straightforward systems tend to have a mean efficiency as low as only 5% or less. On the other hand, even the very complex but optimized systems can achieve efficiencies of more than 20%.

The SCIUNI simulation can also find specific rules, especially for designing the efficient EGS systems for BSE detection. It was proven that the widening EGS systems are more efficient than the strip ones. The presented simple strip system does not achieve mean efficiencies higher than about 1%. The reason is that such light guides cannot transport photons whose trajectories do not meet the condition of total reflection. In other words, the strip light guides cannot change the trajectory of photons in the direction along the system axis. A gradual widening of the EGS system, shifting this widening as close to the scintillator as possible, optimizing its angel to the system axis, and integrating a conical light-guiding ring close to the scintillator contributes to the arrangement of the photon trajectories longitudinally with the system axis. This arrangement helps increase light-guiding efficiency to as high as 20%.

#### CONCLUSION 5

Except for those processes that are dependent on the electron microscope arrangement and except for photoelectron processing, all other processes in the scintillation electron detector in S(T)EM, related to the scintillator and the light guide, were analyzed in this paper. In other words, the excitation electron trajectories, including absorbed energy distributions, CL kinetics, and efficiencies of scintillators, properties of their substrates and anti-charging conductive coatings, assigned optical properties, and light guide configurations were presented and discussed in this paper.

Unlike many other publications on garnet scintillators, which focus on physical processes in scintillation material, this paper is focused on the complete configuration and design of the S(T)TEM detector with the garnet scintillators. From the results, it is evident that the mentioned detector parts cannot be ignored. As was analyzed, the scintillators with bad conductive coating will be charged and will not reflect the light signal to the scintillator exit surface. The results indicate that problems occur with low-energy detection around 0.7 keV when the Sc coating must be as thin as only units of nanometers to allow electron penetration without significant losses. The key priority in the S(T)EM scintillation detector is to use an extremely fast scintillator. The short rise and decay time and low afterglow of the highly doped LuGdGaAG:Ce (high Gd&Ga) and LuGdGaAG:Ce,Mg (high Ce) LPE garnets guarantee strong MTF of the entire imaging system. It was proven that the detector with these fastest garnet film scintillators allows the imaging system in S(T) EM to have a much better resolution than that with the YAG:Ce scintillator. The mentioned fast film scintillators lose the contrasttransfer ability at small details above 0.6 lp/pixel (line pairs per pixel), while the YAG:Ce single crystal scintillator does so already above 0.1 lp/pixel. Unfortunately, the faster scintillator response generally cannot be achieved without some loss of the CL intensity. However, this does not reduce the DQE too much. Compared to the single crystals, the big difference is the thicknesses of only about 10-18 µm when applying the LPE garnet film scintillators. Therefore, they cannot exist without some light-guiding substrates. In fact, this is an advantage due to lowlight-signal self-absorption in the thin scintillator and reduced absorption in the substrate

The optical absorption coefficients, refractive indices, and the mirror optical reflectance of all materials involved in the light transport to the PMT photocathode were investigated and used as input parameters in the MC SCIUNI simulation of the light signal transport in this paper. When configuring a light-guide system, there is a big problem when designing its size and shape. Therefore, COD application (SCIUNI) was used to solve this problem. It was shown that the nonoptimized EGS possess a very poor light-guiding efficiency of less than 1%. On the other hand, even very complex but optimized EGS can achieve efficiencies of more than 20%. In general, it was proven that a gradual widening of the EGS system, shifting this widening as close to the scintillator as possible, optimizing its angel to the system axis, and integrating a conical light-guiding ring close to the scintillator increase light-guiding efficiency.

In summary, the LPE LuGdAG:Ce garnet film scintillators are an excellent basis for the fast and efficient scintillation electron detectors in S(T)EM. They possess strong MTF and all presumptions for integration into both the BGS and EGS scintillation detection systems. In combination with the MC-optimized light guides, they can form a highly powerful detector, not only for S(T)EM but also for other e-beam devices.

## ACKNOWLEDGMENTS

The authors thank Jaroslav Sobota and Tomas Fort from the Institute of Scientific Instruments of the CAS, Brno, Czech Republic, for sputtering the conductive coatings. They also thank the company Crytur, Turnov, Czech Republic, for the supply of the YAG:Ce reference single crystal scintillator. This research was supported by the Ministry of Industry and Trade of the Czech Republic (project TRIO FV30271), and the Ministry of Education, Youth and Sports of the Czech Republic (project LO1212). The research infrastructure was funded by the Ministry of Education, Youth and Sports of the Czech Republic and European Commission (project CZ.1.05/2.1.00/01.0017), and the Czech Academy of Sciences (project RVO:68081731).

## ORCID

Petr Schauer D https://orcid.org/0000-0002-7930-0983

## REFERENCES

Autrata, R., Schauer, P., Kvapil, J., & Kvapil, J. (1978). A single crystal of YAG-new fast scintillator in SEM. Journal of Physics E: Scientific Instruments, 11(7), 707.

- Autrata, R., Schauer, P., Kvapil, J., & Kvapil, J. (1983). A single crystal of YAIO3:Ce3+ as a fast scintillator in SEM. *Scanning*, *5*(2), 91–96.
- Babin, V., Chernenko, K., Kučera, M., Nikl, M., & Zazubovich, S. (2016).
  Photostimulated luminescence and defects creation processes in Ce 3
  +-doped epitaxial films of multicomponent Lu 3– x GdxGayAl 5– y O
  12 garnets. *Journal of Luminescence*, 179, 487–495.
- Bok, J., Lalinský, O., Hanuš, M., Onderišinová, Z., Kelar, J., & Kučera, M. (2016). GAGG: ce single crystalline films: New perspective scintillators for electron detection in SEM. *Ultramicroscopy*, 163, 1–5.
- Bok, J., & Schauer, P. (2014a). Apparatus for temperature-dependent cathodoluminescence characterization of materials. *Measurement Science* and Technology, 25(7), 075601.
- Bok, J., & Schauer, P. (2014b). Performance of SEM scintillation detector evaluated by modulation transfer function and detective quantum efficiency function. *Scanning*, 36(4), 384–393.
- Bolek, P., Zeler, J., Gorbenko, V., Zorenko, T., Popielarski, P., Bartosiewicz, K., ... Zorenko, Y. (2020). Luminescent properties of nanopowder and single-crystalline films of TbAG:Ce garnet. *Physica Status Solidi B-Basic Solid State Physics.*257(8):1900495.
- Bril, A., Blasse, G., & Poorter, J. A. D. (1970). Fast-decay phosphors. Journal of the Electrochemical Society, 117(3), 346–348.
- Buryi, M., Laguta, V., Nikl, M., Gorbenko, V., Zorenko, T., & Zorenko, Y. (2019). LPE growth and study of the Ce3+ incorporation in LuAIO3:Ce single crystalline film scintillators. *Crystengcomm*, 21(21), 3313–3321.
- Carrier, C., & Lecomte, R. (1990a). Effect of geometrical modifications and crystal defects on light collection in ideal rectangular parallelepipedic BGO scintillators. *Nuclear Instruments and Methods in Physics Research Section A*, 294(1–2), 355–364.
- Carrier, C., & Lecomte, R. (1990b). Theoretical modeling of light transport in rectangular parallelepipedic scintillators. *Nuclear Instruments & Methods in Physics Research Section A*, 292(3), 685–692.
- Chapman, J. N., Craven, A. J., & Scott, C. P. (1989). Electron detection in the analytical electron-microscope. Ultramicroscopy, 28(1–4), 108–117.
- Chewpraditkul, W., Pattanaboonmee, N., Sakthong, O., Chewpraditkul, W., Yoshino, M., Horiai, T., ... Kurosawa, S. (2020). Luminescence and scintillation properties of Mg2+-codoped Lu0. 6Gd2.4Al2Ga3O12:Ce single crystal. *IEEE Transactions on Nuclear Science*, 67(6), 904–909.
- Chewpraditkul, W., Swiderski, L., Moszynski, M., Szczesniak, T., Syntfeld-Kazuch, A., Wanarak, C., & Limsuwan, P. (2009). Scintillation properties of LuAG:Ce, YAG:Ce and LYSO:Ce crystals for gamma-ray detection. *IEEE Transactions on Nuclear Science*, 56(6), 3800–3805.
- Danilatos, G. D. (2012). Backscattered electron detection in environmental SEM. Journal of Microscopy, 245(2), 171–185.
- Dujardin, C., Auffray, E., Bourret-Courchesne, E., Dorenbos, P., Lecoq, P., Nikl, M., ... Zhu, R. Y. (2018). Needs, trends, and advances in inorganic scintillators. *IEEE Transactions on Nuclear Science*, 65(8), 1977–1997.
- Everhart, T. E., & Thornley, R. F. M. (2004). Wide-band detector for micromicroampere low-energy electron currents. Advances in Imaging and Electron Physics, 133, 147–152.
- Filippov, M. N., Rau, E. I., Sennov, R. A., Boyde, A., & Howell, P. G. T. H. (2001). Light collection efficiency and light transport in backscattered electron scintillator detectors in scanning electron microscopy. *Scanning*, 23(5), 305–312.
- Frank, L. (2002). Advances in scanning electron microscopy. Advances in Imaging and Electron Physics, 123, 327–373.
- Gorbenko, V., Zorenko, T., Paprocki, K., Iskatiyeva, A., Fedorov, A., Schroppel, F., ... Zorenko, Y. (2017). Epitaxial growth of single crystalline film phosphors based on the Ce3+-doped Ca2YMgScSi3O12 garnet. *Crystengcomm*, 19(26), 3689–3697.
- Gorbenko, V., Zorenko, T., Witkiewicz, S., Paprocki, K., Sidletskiy, O., Fedorov, A., ... Zorenko, Y. (2017). LPE growth of single crystalline film scintillators based on Ce3+ doped Tb3-xGdxAl5-yGayO12 mixed garnets. *Crystals*, 7(9), 262.
- Hamamatsu. (2016). Photomultiplier tubes and related products. Japan: Hamamatsu Photonics K.K.

- Hatzakis, M. (1970). A new method of forming scintillators for electron collectors. *Review of Scientific Instruments*, 41(1), 128–128.
- Healy, O. E., & Mott, R. B. (2016). Real-world electron detector performance in scanning electron microscopes. *Microscopy and Microanalysis*, 22(S3), 596–597.
- Hejna, J. (1987). A ring scintillation detector for detection of backscattered electrons in the scanning electron-microscope. *Scanning Microscopy*, 1 (3), 983–987.
- Hibino, M., Irie, K., Autrata, R., & Schauer, P. (1992). Characteristics of YAG single-crystals for electron scintillators of stem. *Journal of Electron Microscopy*, 41(6), 453–457.
- Joy, D. C. (1995). Monte Carlo modeling for electron microscopy and microanalysis. New York: Oxford University Press.
- Joy, D. C., Joy, C. S., & Bunn, R. D. (1996). Measuring the performance of scanning electron microscope detectors. *Scanning*, 18(8), 533–538.
- Knittel, Z. (1976). Optics of thin films (An optical multilayer theory). New York, NY): John Wiley & Sons.
- Kucera, M., Onderisinova, Z., Bok, J., Hanus, M., Schauer, P., & Nikl, M. (2016). Scintillation response of Ce 3+ doped GdGa-LuAG multicomponent garnet films under e-beam excitation. *Journal of Luminescence*, 169, 674–677.
- Kucera, M., & Prusa, P. (2017). LPE-grown thin-film scintillators. In M. Nikl (Ed.), Nanocomposite, ceramic and thin film scintillators (pp. 155–226). Singapore: Pan Stanford Publishing.
- Kuwano, Y. (1978). Refractive-indexes of Yaio3-Nd. Journal of Applied Physics, 49(7), 4223–4224.
- Kuwano, Y., Saito, S., & Hase, U. (1988). Crystal-growth and opticalproperties of Nd-Ggag. *Journal of Crystal Growth*, 92(1–2), 17–22.
- Lecoq, P., Gektin, A., & Korzhik, M. (2017). Addressing the increased demand for fast timing. In *Inorganic scintillators for detector systems physical principles and crystal engineering* (2nd ed., pp. 103–123). Switzerland: Springer International Publishing.
- Lerche, C. W., Ros, A., Herrero, V., Esteve, R., Monzo, J. M., Sebastia, A., ... Benlloch, J. M. (2008). Dependency of energy-, position- and depth of interaction resolution on scintillation crystal coating and geometry. *IEEE Transactions on Nuclear Science*, 55(3), 1344–1351.
- Mihokova, E., Nikl, M., Mares, J. A., Beitlerova, A., Vedda, A., Nejezchleb, K., ... D'Ambrosio, C. (2007). Luminescence and scintillation properties of YAG:Ce single crystal and optical ceramics. *Journal* of Luminescence, 126(1), 77–80.
- Mullerova, I., & Konvalina, I. (2009). Collection of secondary electrons in scanning electron microscopes. *Journal of Microscopy*, 236(3), 203–210.
- Nedela, V., Tihlarikova, E., Runstuk, J., & Hudec, J. (2018). High-efficiency detector of secondary and backscattered electrons for low-dose imaging in the ESEM. *Ultramicroscopy*, 184, 1–11.
- Nikl, M., Kamada, K., Babin, V., Pejchal, J., Pilarova, K., Mihokova, E., ... Yoshikawa, A. (2014). Defect engineering in Ce-doped aluminum garnet single crystal scintillators. *Crystal Growth & Design*, 14(9), 4827–4833.
- Nikl, M., Vedda, A., Fasoli, M., Fontana, I., Laguta, V., Mihokova, E., ... Nejezchleb, K. (2007). Shallow traps and radiative recombination processes in Lu 3 Al 5 O 12:Ce single crystal scintillator. *Physical Review B*, 76(19), 195121.
- Nikl, M., & Yoshikawa, A. (2015). Recent R&D trends in inorganic singlecrystal scintillator materials for radiation detection. Advanced Optical Materials, 3(4), 463–481.
- Nikl, M., Yoshikawa, A., Kamada, K., Nejezchleb, K., Stanek, C., Mares, J., & Blazek, K. (2013). Development of LuAG-based scintillator crystals—A review. Progress in Crystal Growth and Characterization of Materials, 59 (2), 47–72.
- Ohlídal, I., & Navrátil, K. (1984). Spectroscopic methods for optical analysis of thin films. Folia Facultatis Scientiarum Naturalium Universitatis Purkynianae Brunensis: Physica, 25(37), 5–83.

WILEY-

- Prusa, P., Kucera, M., Mares, J. A., Onderisinova, Z., Hanus, M., Babin, V., ... Nikl, M. (2015). Composition tailoring in Ce-doped multicomponent garnet epitaxial film scintillators. *Crystal Growth & Design*, 15(8), 3715–3723.
- Reimer, L. (1993). Detector systems. In *Image formation in low-voltage scanning electron microscopy* (pp. 29–40). Bellingham, Washington: SPIE Press.
- Reimer, L. (1998). Electron detectors and spectrometers. In P. W. Hawkes (Ed.), Scanning electron microscopy: Physics of image formation and microanalysis. Berlin Heidelberg New York: Springer-Verlag.
- Reimer, L. (2013). Scanning electron microscopy: Physics of image formation and microanalysis. Springer Series in Optical Sciences, Heidelberg, Germany: Springer-Verlag.
- Robbins, D. J., Cockayne, B., Lent, B., Duckworth, C. N., & Glasper, J. L. (1979). Investigation of competitive recombination processes in rareearth activated garnet phosphors. *Physical Review B*, 19(2), 1254–1269.
- Salomoni, M., Pots, R., Auffray, E., & Lecoq, P. (2018). Enhancing light extraction of inorganic scintillators using photonic crystals. *Crystals*, 8 (2), 78.
- Schauer, P. (2007). Extended algorithm for simulation of light transport in single crystal scintillation detectors for S(T)EM. *Scanning*, 29(6), 249–253.
- Schauer, P. (2011). Optimization of decay kinetics of YAG:Ce single crystal scintillators for S (T) EM electron detectors. *Nuclear Instruments and Methods in Physics Research Section B*, 269(21), 2572–2577.
- Schauer, P., & Autrata, R. (1979). Electro-optical properties of a scintillation detector in SEM. Journal de Microscopie et de Spectroscopie Electroniques, 4(6), 633–650.
- Schauer, P., & Autrata, R. (1992). Light transport in single-crystal scintillation detectors in SEM. Scanning, 14(6), 325–333.
- Schauer, P., & Bok, J. (2013). Study of spatial resolution of YAG:Ce cathodoluminescent imaging screens. Nuclear Instruments and Methods in Physics Research Section B, 308, 68–73.
- Schauer, P., Lalinsky, O., & Kucera, M. (2019). Prospective scintillation electron detectors for S(T)EM based on garnet film scintillators. *Microscopy Research and Technique*, 82(3), 272–282.
- Schauer, P., Lalinsky, O., Kucera, M., Lucenicova, Z., & Hanus, M. (2017). Effect of Mg co-doping on cathodoluminescence properties of LuGAGG:Ce single crystalline garnet films. *Optical Materials*, 72, 359–366.
- Skoupy, R., Fort, T., & Krzyzanek, V. (2020). Nanoscale estimation of coating thickness on substrates via standardless BSE detector calibration. *Nanomaterials*, 10(2), 332.
- Tamulaitis, G., Vaitkevicius, A., Nargelas, S., Augulis, R., Gulbinas, V., Bohacek, P., ... Auffray, E. (2017). Subpicosecond luminescence rise

time in magnesium codoped GAGG:Ce scintillator. Nuclear Instruments & Methods in Physics Research Section A, 870, 25–29.

- Tikhonravov, A. V., Trubetskov, M. K., Hrdina, J., & Sobota, J. (1996). Characterization of quasi-rugate filters using ellipsometric measurements. *Thin Solid Films*, 277(1–2), 83–89.
- Witkiewicz-Lukaszek, S., Gorbenko, V., Zorenko, T., Paprocki, K., Sidletskiy, O., Fedorov, A., ... Zorenko, Y. (2018). Epitaxial growth of composite scintillators based on Tb3Al5O12:Ce single crystalline films and Gd3Al2.5Ga2.5O12:Ce crystal substrates. *Crystengcomm*, 20(28), 3994–4002.
- Witkiewicz-Lukaszek, S., Mrozik, A., Gorbenko, V., Zorenko, T., Bilski, P., Fedorov, A., & Zorenko, Y. (2020). LPE growth of composite thermoluminescent detectors based on the Lu3-xGdxAl5O12:Ce single crystalline films and YAG:Ce crystals. *Crystals*, 10(3), 189.
- Xiaoguang, Y. (1984). A study of light collection efficiency in scintillation detectors. Nuclear Instruments and Methods in Physics Research Section A, 228(1), 101–104.
- Yamamoto, K., Tanji, T., Hibino, M., Schauer, P., & Autrata, R. (2000). Improvement of light collection efficiency of lens-coupled YAG screen TV system for a high-voltage electron microscope. *Microscopy Research and Technique*, 49(6), 596–604.
- Zorenko, Y., Gorbenko, V., Savchyn, V., Voznyak, T., Sidletskiy, O., Grinyov, B., ... Douissard, P. A. (2012). Single crystalline film scintillators based on the orthosilicate, perovskite and garnet compounds. *IEEE Transactions on Nuclear Science*, 59(5), 2260–2268.
- Zorenko, Y., Gorbenko, V., Zorenko, T., Sidletskiy, O., Fedorov, A., Bilski, P., & Twardak, A. (2015). High-performance Ce-doped multicomponent garnet single crystalline film scintillators. *Physica Status Solidi* (*RRL*)-*Rapid Research Letters*, 9(8), 489–493.
- Zorenko, Y., Mares, J. A., Prusa, P., Nikl, M., Gorbenko, V., Savchyn, V., ... Nejezchleb, K. (2010). Luminescence and scintillation characteristics of YAG:Ce single crystalline films and single crystals. *Radiation Measurements*, 45(3–6), 389–391.
- Zorenko, Y. V., Gorbenko, V. I., Stryganyuk, G. B., Kolobanov, V. N., Spasskii, D. A., Blazek, K., & Nikl, M. (2005). Luminescence of excitons and antisite defects in Lu3Al5O12:Ce single crystals and single-crystal films. *Optics and Spectroscopy*, *99*(6), 923–931.
- Zych, E., Brecher, C., & Glodo, J. (2000). Kinetics of cerium emission in a YAG:Ce single crystal: The role of traps. *Journal of Physics-Condensed Matter*, 12(8), 1947–1958.

How to cite this article: Schauer P, Lalinský O, Kučera M. Overview of S(T)EM electron detectors with garnet scintillators: Some potentials and limits. *Microsc Res Tech*. 2020;1–18. https://doi.org/10.1002/jemt.23634