Sensors and Materials, Vol. 35, No. 2 (2023) 423–428
MYU Tokyo

S & M 3178

Gamma-ray Energy Dependence of Scintillation Decay Time

Takayuki Yanagida,[1*] Kenichi Watanabe,[2] Takumi Kato,[1]
Daisuke Nakauchi,[1] and Noriaki Kawaguchi[2]

[1]Nara Institute of Science and Technology,
8916-5 Takayama-cho, Ikoma-city, Nara 630-0192, Japan
[2]Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University,
744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan

(Received September 27, 2022; accepted November 30, 2022)

Keywords: scintillator, gamma ray, scintillation, decay

We investigated scintillation decay times at different excitation γ-ray energies for commercial luminescent materials such as Ce:$LaBr_3$, Eu:SrI_2, Ce:LYSO [$(Y,Lu)_2SiO_5$], and Ce:GAGG [$Gd_3(Al,Ga)_5O_{12}$]. Ce:$LaBr_3$ and Eu:SrI_2 did not show energy dependence, whereas Ce:LYSO and Ce:GAGG exhibited different decay curves at different excitation energies. In these materials, the energy dependence of decay correlated with the nonproportionality of scintillation light yield.

1. Introduction

Luminescent materials have been used for many applications, among which ionizing radiation detection is one of the important scientific and technological themes enriching our daily lives. Luminescent materials for radiation detection are roughly classified into two types: scintillators[(1–3)] and storage phosphors for dosimeters.[(4–7)] Since the interaction probability of radiation species and elements varies widely, many kinds of materials including single crystals,[(8–31)] glasses,[(32–50)] ceramics,[(51–56)] and other materials[(57–60)] have been researched. Requirements for properties vary considerably among applications, although high interaction probability, high luminescence intensity, and chemical stability are generally required.

In some applications, timing properties are important, especially in scintillation detectors for high-counting-rate environments. Examples are medical applications such as photon-counting computed tomography (CT)[(61)] and positron emission tomography (PET).[(62)] In these applications, the target ionizing radiation is photons with high energies of several hundred keV such as X-rays and γ-rays, and sub-ns- to ns-order timing properties are required. These applications use scintillators as the main device, and the timing property mainly depends on the scintillation decay time. Up to now, the research community has believed that the scintillation decay time is constant under excitation by high-energy photons, and differences in decay time

*Corresponding author: e-mail: t-yanagida@ms.naist.jp
https://doi.org/10.18494/SAM4135

ISSN 0914-4935
https://myukk.org/

are only caused by differences in the dopants and host used, which are based on fundamental quantum theory.[63] However, some recent studies have indicated that the scintillation decay time may have an energy dependence upon high-energy photon excitation.[64–66] The energy dependence of the decay times of Tl:NaI, Ce:Lu_2SiO_5 (LSO), $CeBr_3$, Cs_2LiYCl_6 (CLYC), and CsI has been studied in pioneering works, which showed energy dependence.[64–66]

Following such pioneering works, the current study focuses on four commercial scintillators: Ce:$LaBr_3$ (Saint-Gobain),[24] Eu:SrI_2 (Oxide Corp.),[67] Ce:$Lu_{1.8}Y_{0.2}SiO_5$ (LYSO) (Saint-Gobain),[26] and Ce:$Gd_3Ga_3Al_2O_{12}$ (GAGG) (Furukawa).[68,69]

2. Materials and Methods

The materials examined were the commercial single-crystal scintillators Ce:$LaBr_3$ (Saint-Gobain),[24] Eu:SrI_2 (Oxide Corp.),[67] Ce:LYSO (Saint-Gobain),[26] and Ce:GAGG (Furukawa).[68,69] $LaBr_3$ and SrI_2 were encapsulated, and the sample size was 1 inchΦ × 1 inch. The sample sizes of LYSO and GAGG were 5 × 5 × 5 mm^3 and 5 × 5 × 1 mm^3, respectively. In the measurement, the scintillator was optically attached to a photomultiplier tube (PMT, R7600U-200, Hamamatsu), and the signal output was directly connected to a PC-based digitizer (Pico Technology, PicoScope 6402D) with a 50 Ω termination. Scintillation decay curves of each event were recorded by the digitizer, and after averaging a sufficient number of scintillation decay curves with similar energies, the scintillation decay time was obtained by fitting each decay curve to a single exponential decay function. In this manner, the γ-ray energy dependence of scintillation decay time was evaluated. Furthermore, the nonproportionality of scintillation light yield was evaluated since a relationship between the energy dependence of decay time and the nonproportionality was reported in earlier works.[64–66] In the study of nonproportionality, its value at 662 keV was defined as 1, and the pulse height spectra were recorded by a common setup[57] using the same PMT as that used for decay time measurement. In these experiments, by using radio isotopes such as ^{22}Na, ^{55}Fe, ^{109}Cd, ^{133}Ba, ^{137}Cs, ^{152}Eu, and ^{241}Am, the energy dependences of scintillation decay time and nonproportionality were investigated.

3. Results and Discussion

Figure 1 summarizes the obtained scintillation decay times and the nonproportionalities of Ce:$LaBr_3$, Eu:SrI_2, Ce:LYSO, and Ce:GAGG as functions of γ-ray energy. As in previous works,[64–66] no significant changes were observed at high energies of >100 keV. In Ce:$LaBr_3$, owing to the limited dynamic range of signal intensities, the observation at energies larger than 100 keV was not possible. Since an energy dependence was observed, the data points (number of events) of Ce:LYSO and Ce:GAGG were investigated in detail with a long measurement time. In these measurements, the dominant factor determining the accuracy of decay time was the fitting error, which was within 2%.

In Eu:SrI_2, the decay time did not depend on energy, and the nonproportionality also showed no energy dependence. The typical decay curves at low and high energies appeared identical, as shown in the inset of the Eu:SrI_2 panel. The same tendency was also observed in Ce:$LaBr_3$.

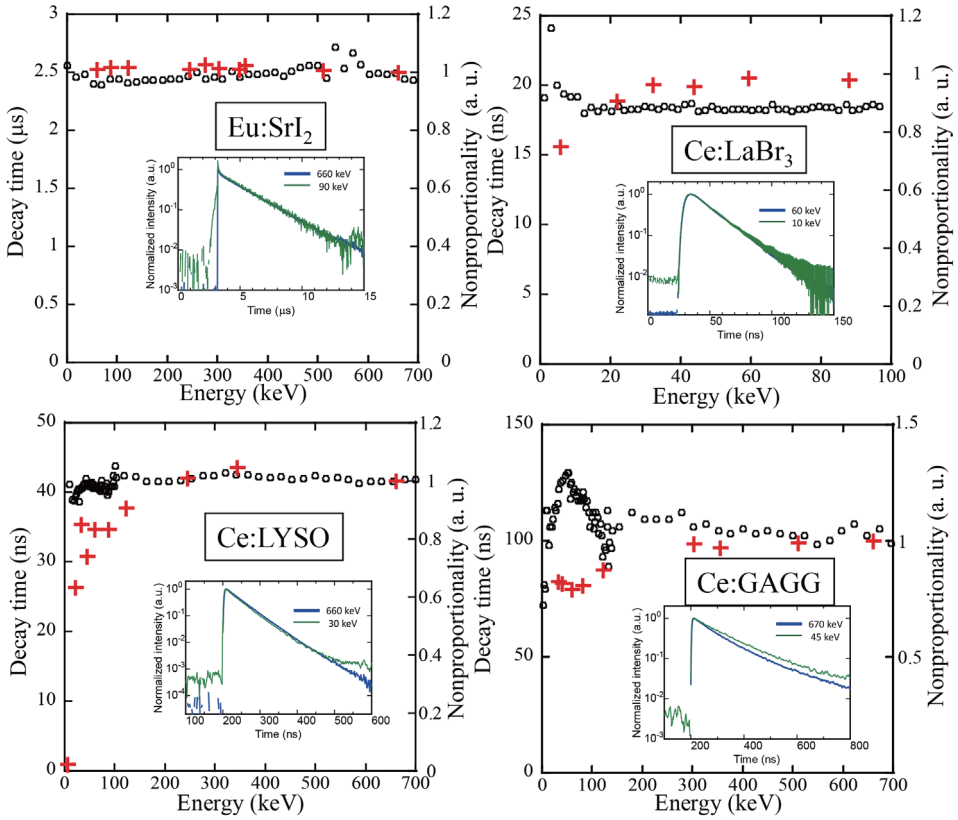

Fig. 1. (Color online) Energy dependences of scintillation decay times (circles) and nonproportionalities (crosses) of Ce:$LaBr_3$, Eu:SrI_2, Ce:LYSO, and Ce:GAGG. The inset shows typical scintillation decay curves at low and high energies.

However, the decay time increased by 10–20% at energies lower than 20 keV, and the nonproportionality decreased in the same energy range. Although the number of data points was limited, it appears that Ce:$LaBr_3$ shows an energy-dependent scintillation decay at energies lower than 20 keV.

In the case of Ce:LYSO, the energy dependence of decay time was observed at lower energies, with a peak at ~50 keV, which coincided with the peak in the nonproportionality. In a past work on Ce:LSO,[64] the decay time increased from ~45 ns at several tens of keV to ~50 ns at ~10 keV. The same increasing tendency was observed from 100 to 50 keV in our case, and the tendency depended on the chemical composition (Lu_2SiO_5 in the past work[64] and $Lu_{1.8}Y_{0.2}SiO_5$ in this work). The difference in the tendency may also be caused by the focusing time range. In the former work,[64] only the fast component was analyzed with six data points below 100 keV, and different results may have been obtained if the slower component had been focused on. The inset shows a comparison of decay curves at different excitation energies, and a difference can be clearly observed from 500 ns.

In Ce:GAGG, a clearer energy dependence of decay time was observed, and the energy dependence curve below 100 keV had a peak at ~45 keV. The nonproportional response also had an inflection point at a similar energy. The inset shows scintillation decay curves at low and high

energies, and the difference between them is clear. When the excitation energy was low, the scintillation decay time became long. When Ce:LYSO and Ce:GAGG are used in the energy range below 100 keV, users must consider such a change in decay time.

As indicated in pioneering works, the energy dependence of decay time is related to the nonproportionality.[64] It is already widely known that the nonproportionality is correlated with energy resolution; the current study shows that it is also correlated with scintillation decay time. Although the origin of the nonproportionality is still under discussion, it is known that the inflection point of the nonproportionality appears around the K-edge energy of the heaviest element in the scintillator (e.g., Lu in Lu_2SiO_5), which was found in 2001[70] and has been confirmed for many materials. At energies around the K-edge (50.2 keV for Gd and 63.3 keV for Lu), the probability of interaction of high-energy photons markedly changes, and this phenomenon is related to energy deposition (linear energy transfer: LET). Further study is required to clarify the physics of nonproportionality, energy resolution, and decay time.

4. Conclusions

We investigated the energy dependence of the crystal scintillators Ce:$LaBr_3$, Eu:SrI_2, Ce:LYSO, and Ce:GAGG. Among these four scintillators, Ce:LYSO and Ce:GAGG showed a clear energy dependence, and the dependence coincided with the nonproportional response of the scintillation light yield as a function of energy. The scintillation decay times of Ce:LYSO and Ce:GAGG changed markedly at energies lower than 100 keV, and such dependence should be considered in the design of new γ-ray detectors.

Acknowledgments

This work was supported by Grants-in-Aid for Scientific A (22H00309), Scientific B (22H02939, 21H03733, and 21H03736), and Exploratory Research (22K18997) from Japan Society for the Promotion of Science. The Cooperative Research Project of Research Center for Biomedical Engineering is also acknowledged.

References

1 T. Yanagida: Proc. Japan Academy Ser. B **94** (2018) 75. https://doi.org/10.2183/pjab.94.007
2 T. Yanagida: J. Lumin. **169** (2016) 544. https://doi.org/10.1016/j.jlumin.2015.01.006
3 T. Yanagida, T. Kato, D. Nakauchi, and N. Kawaguchi: Jpn. J. Appl. Phys. **62** (2022) 010508. https://doi.org/10.35848/1347-4065/ac9026
4 T. Yanagida, G. Okada, and N. Kawaguchi: J. Lumin. **207** (2019) 14. https://doi.org/10.1016/j.jlumin.2018.11.004
5 T. Yanagida, G. Okada, T. Kato, D. Nakauchi, and N. Kawaguci: Radiat. Meas. **158** (2022) 106847. https://doi.org/10.1016/j.radmeas.2022.106847
6 G. Okada, Y. Koguchi, T. Yanagida, S. Kasap, and H. Nanto: Jpn. J. Appl. Phys. **62** (2022) 010609. https://doi.org/10.35848/1347-4065/ac9023
7 G. Okada, K. Hirasawa, T. Yanagida, and H. Nanto: Sens. Mater. **33** (2021) 2117. https://doi.org/10.18494/SAM.2021.3327
8 Y. Fujimoto, D. Nakauchi, T. Yanagida, M. Koshimizu, and K. Asai: Sens. Mater. **33** (2021) 2147. https://doi.org/10.18494/SAM.2021.3321
9 P. Kantuptim, H. Fukushima, H. Kimura, D. Nakauchi, T. Kato, M. Koshimizu, N. Kawaguchi, and T. Yanagida: Sens. Mater. **33** (2021) 2195. https://doi.org/10.18494/SAM.2021.3316

10 T. Yanagida, M. Sakairi, T. Kato, D. Nakauchi, and N. Kawaguchi: Appl. Phys. Exp. **13** (2020) 016001. https://doi.org/10.7567/1882-0786/ab5ffa
11 D. Nakauchi, T. Kato, N. Kawaguchi, and T. Yanagida: Sens. Mater. **33** (2021) 2203. https://doi.org/10.18494/SAM.2021.3323
12 H. Fukushima, M. Akatsuka, H. Kimura, D. Onoda, D. Shiratori, D. Nakauchi, T. Kato, N. Kawaguchi, and T. Yanagida: Sens. Mater. **33** (2021) 2235. https://doi.org/10.18494/SAM.2021.3324
13 D. Nakauchi, G. Okada, N. Kawaguchi, and T. Yanagida: Jpn. J. Appl. Phys. **7** (2018) 100307. https://doi.org/10.7567/JJAP.57.100307
14 M. Akatsuka, H. Kimura, D. Onoda, D. Shiratori, D. Nakauchi, T. Kato, N. Kawaguchi, and T. Yanagida: Sens. Mater. **33** (2021) 2243. https://doi.org/10.18494/SAM.2021.3319
15 M. Koshimizu, N. Kawano, A. Kimura, S. Kurashima, M. Taguchi, Y. Fujimoto, and K. Asai: Sens. Mater. **33** (2021) 2137. https://doi.org/10.18494/SAM.2021.3314
16 T. Yanagida, G. Okada, T. Kato, D. Nakauchi, and S. Yanagida: Appl. Phys. Exp. **9** (2016) 042601. https://doi.org/10.7567/APEX.9.042601
17 K. Okazaki, D. Onoda, D. Nakauchi, N. Kawano, H. Fukushima, T. Kato, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 575. https://doi.org/10.18494/SAM3678
18 T. Yanagida, Y. Fujimoto, K. Fukuda, and V. Chani: Nucl. Instrum. Methods Phys. Res. A **729** (2013) 58. https://doi.org/10.1016/j.nima.2013.07.010
19 D. Onoda, M. Akatsuka, N. Kawano, T. Kato, D. Nakauchi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 585. https://doi.org/10.18494/SAM3679
20 T. Yanagida, T. Kato, D. Nakauchi, and N. Kawaguchi: Sens. Mater. **34** (2022) 595. https://doi.org/10.18494/SAM3684
21 C. van Aarlea, K. W. Krämer, and P. Dorenbos: J. Lumin. **251** (2022) 119209. https://doi.org/10.1016/j.jlumin.2022.119209
22 P. Kantuptim, D. Nakauchi, T. Kato, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 603. https://doi.org/10.18494/SAM3690
23 D. Nakauchi, H. Fukushima, T. Kato, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 611. https://doi.org/10.18494/SAM3696
24 E. V. D. van Loef, P. Dorenbos, and C. W. E. van Eijk: Appl. Phys. Lett. **79** (2001) 1573. https://doi.org/10.1063/1.1385342
25 M. Akatsuka, N. Daisuke, K. Takumi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 619. https://doi.org/10.18494/SAM3692
26 L. Pidol, A. Kahn-Harari, B. Viana, E. Virey, B. Ferrand, P. Dorenbos, J. T. M. de Haas, and C. W. E. van Eijk: IEEE Trans. Nucl. Sci. **51** (2004) 1084. https://doi.org/10.1109/TNS.2004.829542
27 Y. Fujimoto, D. Nakauchi, T. Yanagida, M. Koshimizu, and K. Asai: Sens. Mater. **34** (2022) 629. https://doi.org/10.18494/SAM3693
28 J. Blevins and G. Yang: Mater. Res. Bull. **144** (2021) 111494. https://doi.org/10.1016/j.materresbull.2021.111494
29 M. Koshimizu, S. Kurashima, A. Kimura, M. Taguchi, T. Yanagida, Y. Fujimoto, and K. Asai: Sens. Mater. **34** (2022) 637. https://doi.org/10.18494/SAM3694
30 M. N. Singh and R. K. Gartia: Mater. Today Proc. **65** (2022) 2657.
31 Y. Takebuchi, T. Kato, D. Nakauchi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 645. https://doi.org/10.18494/SAM3685
32 H. Masai, T. Ina, H. Kimura, N. Kawaguchi, and T. Yanagida: Sens. Mater. **33** (2021) 2155. https://doi.org/10.18494/SAM.2021.3326
33 Y. Miyamoto, H. Nanto, T. Kurobori, Y. Fujimoto, T. Yanagida, J. Ueda, S. Tanabe, and T. Yamamoto: Radiat. Meas. **71** (2014) 529. https://doi.org/10.1016/j.radmeas.2014.08.007
34 T. Kato, D. Shiratori, M. Iwao, H. Takase, D. Nakauchi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **33** (2021) 2163. https://doi.org/10.18494/SAM.2021.3318
35 B. Yang, Y. Zhang, B. Xu, and H. Xia: Acta Optica Sinica **33** (2013) 0216001 (in Chinese).
36 N. Kawaguchi, H. Masai, M. Akatsuka, D. Nakauchi, T. Kato, and T. Yanagida: Sens. Mater. **33** (2021) 2215. https://doi.org/10.18494/SAM.2021.3410
37 J. K. Cao, X. Yao, X. Li, Y. Wei, L. Chen, and H. Guo: J. Lumin. **170** (2016) 207. https://doi.org/10.1016/j.jlumin.2015.10.049
38 K. Ichiba, Y. Takebuchi, H. Kimura, D. Shiratori, T. Kato, D. Nakauchi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 677. https://doi.org/10.18494/SAM3680
39 L. Teng, W. Zhang, W. Chen, J. Cao, X. Sun, and H. Guo: Ceram. Int. **46** (2020) 10718. https://doi.org/10.1016/j.ceramint.2020.01.079
40 G. Ito, H. Kimura, D. Shiratori, D. Nakauchi, T. Kato, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 685. https://doi.org/10.18494/SAM3681

41 H. Kimura, T. Kato, D. Nakauchi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 691. https://doi.org/10.18494/SAM3687
42 Z. Gu, C. Chen, and Y. Zhang: Vacuum **169** (2019) 108832. https://doi.org/10.1016/j.vacuum.2019.108832
43 H. Masai, Y. Yanagida, H. Kawamoto, Y. Koguchi, M. Koshimizu, and M. Yamawaki: Sens. Mater. **34** (2022) 699. https://doi.org/10.18494/SAM3688
44 R. Nakamori, N. Kawano, A. Takaku, D. Onoda, Y. Takebuchi, H. Fukushima, T. Kato, K. Shinozaki, and T. Yanagida: Sens. Mater. **34** (2022) 707. https://doi.org/10.18494/SAM3689
45 J. Zhao, L. Huang, S. Zhao, and S. Xu: Opt. Mater. Exp. **9** (2019) 576. https://doi.org/10.1364/OME.9.000576
46 H. Fukushima, D. Shiratori, D. Nakauchi, T. Kato, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 717. https://doi.org/10.18494/SAM3691
47 N. Kawaguchi, D. Nakauchi, T. Kato, Y. Futami, and T. Yanagida: Sens. Mater. **34** (2022) 725. https://doi.org/10.18494/SAM3705
48 J. Zhao, L. Huang, T. Liang, S. Zhao, and S. Xu: J. Lumin. **205** (2019) 342. https://doi.org/10.1016/j.jlumin.2018.09.045
49 D. Shiratori, Y. Takebuchi, T. Kato, D. Nakauchi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 745. https://doi.org/10.18494/SAM3695
50 S. Qian, L. Huang, S. Zhao, and S. Xu: J. Rare Earths **35** (2017) 787. https://doi.org/10.1016/S1002-0721(17)60977-3
51 H. Kimura, T. Kato, D. Nakauchi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **33** (2021) 2187. https://doi.org/10.18494/SAM.2021.3322
52 J. E. Bell, A. C. Stowe, A. Burger, and K. G. Stassun: Opt. Mater. **112** (2021) 110798. https://doi.org/10.1016/j.optmat.2020.110798
53 T. Kato, D. Nakauchi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 653. https://doi.org/10.18494/SAM3682
54 J. Xu, L. Cao, Y. Feng, Y. Huang, Y. Wang, L. Qin, and H. J. Seo: J. Lumin. **183** (2017) 166. https://doi.org/10.1016/j.jlumin.2016.11.021
55 T. Kunikata, T. Kato, D. Shiratori, D. Nakauchi, N. Kawaguchi, and T. Yanagida: Sens. Mater. **34** (2022) 661. https://doi.org/10.18494/SAM3683
56 J. Xie, Y. Shi, L. Fan, and Z. Xu: Opt. Mater. **35** (2013) 744. https://doi.org/10.1016/j.optmat.2012.08.013
57 T. Yanagida, N. Kawaguchi, Y. Fujimoto, K. Fukuda, K. Watanabe, A. Yamazaki, and A. Uritani: J. Lumin. **144** (2013) 212. https://doi.org/10.1016/j.jlumin.2013.07.016
58 A. Watanabe, A. Magi, M. Koshimizu, A. Sato, Y. Fujimoto, and K. Asai: Sens. Mater. **33** (2021) 2251. https://doi.org/10.18494/SAM.2021.3411
59 S. Matsumoto, Ta. Watanabe, and A. Ito: Sens. Mater. **34** (2022) 669. https://doi.org/10.18494/SAM3698
60 T. Matsubara, T. Yanagida, N. Kawaguchi, T. Nakano, J. Yoshimoto, M. Sezaki, H. Takizawa, S. P. Tsunoda, S. Horigane, S. Ueda, S. Takemoto-Kimura, H. Kandori, A. Yamanaka, and T. Yamashita: Nat. Commun. **12** (2021) 4478. https://doi.org/10.5281/zenodo.4964867
61 T. Flohr, M. Petersilka, A. Henning, S. Ulzheimera, J. Ferda, and B. Schmidt: Physica Medica **79** (2020) 126. https://doi.org/10.1016/j.ejmp.2020.10.030
62 M. Tamal: Heliyon **6** (2020) e05267. https://doi.org/10.1016/j.heliyon.2020.e05267
63 T. Yanagida, Y. Fujimoto, A. Yamaji, N. Kawaguchi, K. Kamada, D. Totsuka, K. Fukuda, K. Yamanoi, R. Nishi, S. Kurosawa, T. Shimizu, and N. Sarukura: Radiat. Meas. **55** (2013) 99. https://doi.org/10.1016/j.radmeas.2012.05.014
64 L. Swiderski, M. Moszynski, A. Syntfeld-Kazuch, M. Szawlowski, and T. Szczesniak: Nucl. Instrum. Methods Phys. Res. A **749** (2014) 68. https://doi.org/10.1016/j.nima.2014.02.045
65 X. Wen and A. Enqvist: Nucl. Instrum. Methods Phys. Res. A **853** (2017) 9. https://doi.org/10.1016/j.nima.2017.02.019
66 W. Wolszczak and P. Dorenbos: Nucl. Instrum. Methods Phys. Res. A **886** (2018) 30. https://doi.org/10.1016/j.nima.2017.12.080
67 T. Yanagida, M. Koshimizu, G. Okada, T. Kojima, J. Osada, and N. Kawaguchi: Opt. Mater. **61** (2016) 119. https://doi.org/10.1016/j.optmat.2016.05.030
68 T. Yanagida, K. Kamada, Y. Fujimoto, H. Yagi, and T. Yanagitani: Opt. Mater. **35** (2013) 2480. https://doi.org/10.1016/j.optmat.2013.07.002
69 T. Yanagida, Y. Fujimoto, M. Koshimizu, K. Watanabe, H. Sato, H. Yagi, and T. Yanagitani: Opt. Mater. **36** (2014) 2016. https://doi.org/10.1016/j.optmat.2013.12.043
70 Y. Uchiyama, M. Kouda, C. Tanihata, N. Isobe, T. Takahashi, T. Murakami, M. Tashiro, K. Makishima, Y. Fukazawa, and T. Kamae: IEEE Trans. Nucl. Sci. **48** (2001) 379. https://doi.org/10.1109/23.940084