Hyperfine Interact
DOI 10.1007/s10751-011-0543-x

Fast scintillation detectors for high-energy X-ray region

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Received: 22 September 2011 / Accepted: 16 December 2011

Abstract We have developed fast scintillation detectors for nuclear resonant scattering experiments using synchrotron radiation and a nuclear excited level existing in >30 keV. A fast x-ray detector using an organic-inorganic perovskite scintillator of phenethylamine lead bromide (PhE-$PbBr_4$) had a dominant light emission with a fast decay time of 9.9 ns. An x-ray detector equipped with a 0.9-mm-thick PhE-$PbBr_4$ crystal (size: $\sim 8 \times 7$ mm^2) was used to detect nuclear resonant scattering in ^{61}Ni (the first excited level: 67.41 keV; half-life: 5.3 ns). We could successfully record the decaying gamma rays emitted from ^{61}Ni with a relatively high detection efficiency of 24%. A lead-doped plastic scintillator (NE142, Pb ∼5 wt% doped) had been known to have a faster decay time of 1.7 ns. Following a test of a single NE142 detector, a four-channel NE142 detector was fabricated and successfully applied to the synchrotron-radiation based Mössbauer spectroscopy experiment on ^{61}Ni.

Keywords Synchrotron Radiation · Nuclear resonant scattering · Scintillation detector · Scintillator

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Published online: 03 January 2012

1 Introduction

Fast detectors have been developed for nuclear resonant scattering (NRS) using synchrotron radiation. A silicon avalanche photodiode (Si-APD) can be used as a photon sensor by direct absorption of γ-rays in a depletion region of silicon. A Si-APD detector of direct detection for X-rays has been a standard detector in this field [1]. The reasons why the Si-APD is suitable for the NRS experiments exist in a wide dynamic rage of outputs from 10^{-2} to 10^{8} s^{-1} and in a sub-nanosecond time resolution for X-ray photons. The properties are extremely useful for separating nuclear radiation from an intense atomic scattering generated when nuclei are excited by an X-ray beam of synchrotron radiation. However, since one device of Si-APD has a thin sensitive layer of 10–150 μm thick and silicon itself has a small atomic number of 14, the intrinsic efficiency is limited to be low of several % for the photon energy of >30 keV. Since the third generation synchrotron rings have recently supplied high-energy X-ray beams of $>10^{10}$ photons s^{-1}, a lot of nuclei whose excited levels exist in >30 keV have been offered to the NRS experiments. With a stack of the APD devices, we can obtain a larger efficiency, for example, 10% even at energy of 67.4 keV [2]. In this case, a beam acceptance of the detector was limited to be a small area of 2×0.03 mm^2, less than 1 mm^2. A larger solid angle of a detector is generally needed except for nuclear forward scattering.

Before the Si-APD detectors were widely used, scintillation detectors had been tested for the NRS experiments. A scintillation detector mounting a plastic scintillator was reported as a fast detector in the year of 1990 [3, 4]. The performance was evaluated at energies of < 20 keV, at that time. The maximum count-rate of the detector was high, up to 10^7, but the detector suffered from a noise of several tens s^{-1} from a photomultiplier tube. No new scintillator suitable for the NRS experiments using high-energy X-rays had been known for these two decades. If using with a new scintillator and with a new experimental method, a scintillation detector will again give us some benefits, covering a low detection-efficiency for high energy photons when using a Si-APD detector. Here, we report on our scintillation detectors newly developed for the NRS experiments in high energy X-ray region.

2 New scintillation detectors

Our new scintillation detectors are listed in Table 1. Properties as a fast detector were investigated by using synchrotron X-rays at beamline BL-14A at the Photon Factory. Figure 1 shows measurement setups for the detectors. In (a) of Fig. 1, X-ray beam directly impinges on the detector window for measurements of energy and time spectra and (b) shows a setup for measuring a time spectrum of scintillation lights. System E is an energy spectroscopy system using a charge-sensitive preamplifier, a main amplifier and a multichannel analyzer (MCA). System T is a time spectroscopy system using a wide-band high-frequency amplifier, a constant-fraction discriminator (CFD), and a time-to-amplitude converter (TAC). In the setup of (b), scintillation light was detected by a photomultiplier tube (PMT, Hamamatsu R7400P) with a probability of less than single photon detection, provided by a distance of several tens mm, between the scintillator and the PMT having an effective entrance window of 8 mm in diameter.

Table 1 Fast scintillation detectors for nuclear resonant scattering

Detector	Scintillator	PMT	Experiment
A	PhE-$PbBr_4$ 8 × 7 mm × 0.9 mm thick	R7400P	Ni-61
B	NE142 9 mm dia. × 5 mm thick	R7400P	I-127
C	NE142 17 mm dia. × 10 mm thick 4 elements	R7400P 4 channels	Ni-61

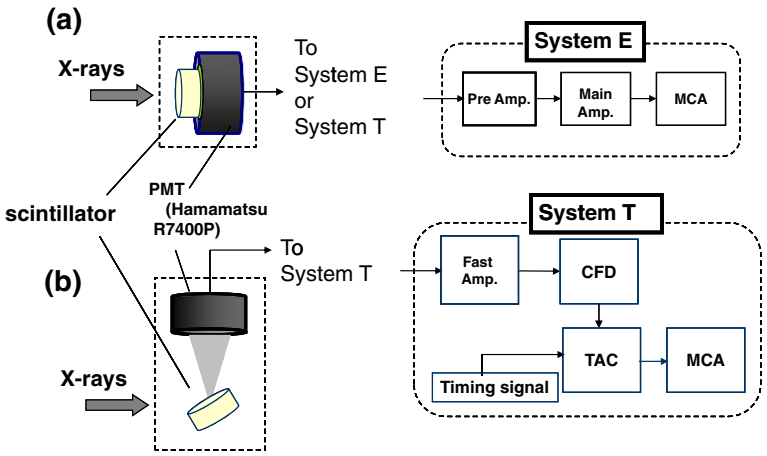

Fig. 1 Experimental setups: (**a**) for direct detection of X-rays and (**b**) for scintillation detection with a single-photon probability. System (E) is used for measuring an energy spectrum and System (T) is for time spectrum

2.1 The PhE-$PbBr_4$ detector

A fast X-ray detector using an organic-inorganic perovskite scintillator of phenethylamine lead bromide (PhE-$PbBr_4$) (Detector A in Table 1) was recently reported [5, 6]. The name of PhE-$PbBr_4$ is abbreviation of $(C_6H_5C_2H_4NH_3)_2PbBr_4$ crystal (bis-(phenethylammonium) tetrabromoplumbate (II). The crystal has the low-dimensional structure consisting of metal-halide layers. This structure causes relatively high efficiency of a fast light emission by exciton, even at room temperature owing to the quantum confinement effect [7]. We produced single crystals of PhE-$PbBr_4$ (area: 10–100 mm^2; thickness: up to 1.7 mm). Although sub-nanosecond light was not observed, the scintillator had a dominant light emission with a fast decay time of 9.9 ± 0.2 ns (72 ± 1% of the total light yield). Other components were estimated by 23 ± 1 ns (25 ± 2%), and 94 ± 3 ns (2.3 ± 0.1%) ns. An X-ray detector equipped with the PhE-$PbBr_4$ crystal was used to detect NRS in ^{61}Ni (the first excited level: 67.41 keV; half-life: 5.34 ns [8]). The Ni-61 NRS measurement was carried out at beamline BL09XU of the SPring-8. Figure 2 shows the PhE-$PbBr_4$ crystal (size: ∼8 × 7 mm^2, thickness: 0.9 mm) and a photomultiplier tube (PMT, Hamamatsu R7400P) of the detector used for the Ni-61 experiment. A sample of

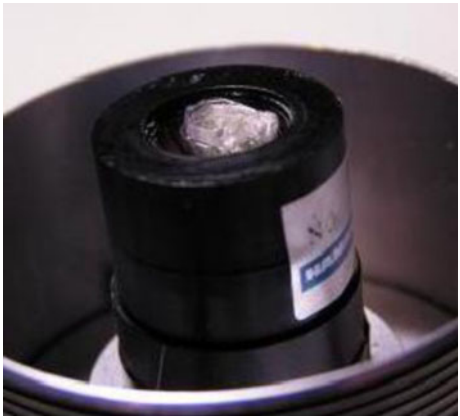

Fig. 2 Photograph of a PhE-$PbBr_4$ crystal and a photomultiplier tube (Hamamatsu R7400P)

Ni metal foil (thickness: 0.38 mm; ^{61}Ni: 95% enriched) was attached to a 3-mm-thick aluminum plate in order to absorb fluorescent X-rays from the Ni foil. We could successfully record the decaying gamma rays emitted from ^{61}Ni with a time resolution of 0.71 $\pm$ 0.05 ns (FWHM) and a relatively high detection-efficiency of 23.7 $\pm$ 0.1%. Although the scintillator had a good efficiency for high-energy X-rays and relatively fast scintillation light, the slow components prolonged the tail of output pulse, unfortunately.

2.2 The NE142 detectors

2.2.1 The single-channel detector

Detector B in Table 1 was fabricated, expecting the time response faster than that of Detector A. A lead-doped plastic scintillator of NE142 (Pb ∼5 wt% doped; equivalent to Saint Gobain BC-452) was equipped to the detector and applied to nuclear quasi-elastic scattering on ^{127}I (the first excited level: 57.60 keV; half-life: 1.9 ns [9]). A cylindrical scintillator of 9 mm in dia. and 5 mm thick was mounted on a small metal-package PMT (Hamamatsu R7400P), which was the same as that of the PhE-$PbBr_4$ detector. By containing lead in plastic base of the scintillator, absorption of X-rays increased to a relatively high value. The intrinsic efficiency was measured in the arrangement of direct beam incident, which was a setup of (a) in Fig. 1, using the system E with a charge-sensitive preamplifier of Canberra 2005 and the PMT voltage of −650 V. The obtained result was 15.3% at 57.6 keV. A simulation result by Geant4 (http://geant4.cern.ch/) indicated that a lead component of 4.2 wt% best fitted to the experimental energy spectrum of Fig. 3. Prepared for an experiment of nuclear inelastic or quasi-elastic scattering, the detection efficiency was estimated to be 3.4% by the simulation, considering with its solid angle of 30%, when X-rays were isotropically emitted from a point 1.9 mm apart from the center of the scintillator's surface. Using the system T in Fig. 1, an output pulse from the 300 MHz-bandwidth fast amplifier ORTEC VT120A was recorded, as shown in Fig. 4. The pulse width (FWHM) was 3 ns at the PMT voltage of −500 V. Count-rate property of the detector was measured at 57.6 keV with the fast counting system using a 500 MHz counter,

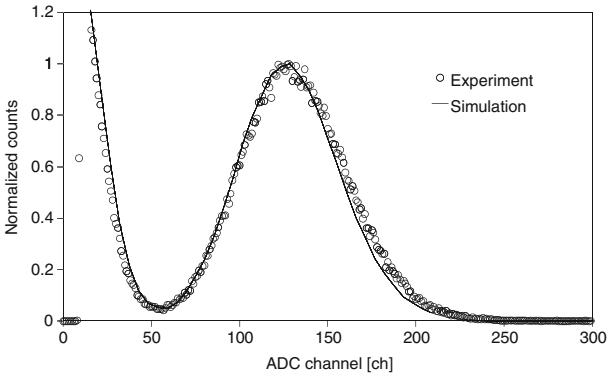

Fig. 3 Energy spectra of the single NE 142 detector: ○ measured at 57.6 keV and − calculated by Geant4

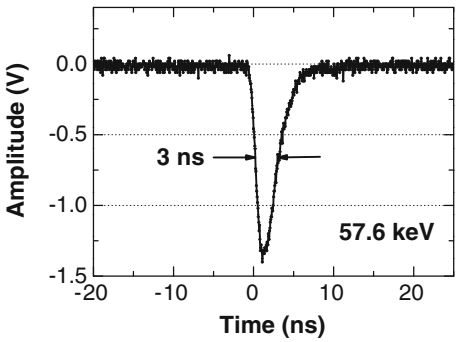

Fig. 4 An output pulse of the single NE142 detector, observed at 57.6 keV

instead of the TAC in System T of Fig. 1. Figure 5 shows the result of the output rates vs. the input photon rates. The input photon rate was controlled with filters of tungsten foil. A detector mounted with a 5-mm thick NaI(Tl) scintillator and the tungsten filters were used to convert a low-rate count less than 10^3 s^{-1} into the input photon rate. The threshold level of the CFD (ORTEC 935) was set to be −0.11 V. In Fig. 5, open circles indicate the output rate and a solid curve means the paralyzable model which has an output width of 26 ns [10]. The observed output rate was saturated at 1.39×10^7 s^{-1}, which was similar to the maximum of the paralyzable model of 26-ns pulse width. Since the pulse width was 7–8 ns on the 0-V baseline, the count limit was caused not only by the output width, also maybe by a PMT current. The time property of the detector was measured at a single-bunch operation of the PF ring, which mode had an X-ray pulse-interval of 624 ns. The scintillation light from the plastic scintillator had a fast decay time of 1.7 ns (96% of the total light yield), and others of 6.4 ns (3.3%) and 41 ns (0.3%), from the fitted result shown in Fig. 6. Each uncertainty given by one standard deviation was at most 7% on both the decay constant and the light yield. This spectrum was measured with the setup of (b) in Fig. 1 and a faster 1.5 GHz amplifier of Phillips Scientific 6954, instead of VT120A. Time resolution of 0.54 ± 0.01 ns (FWHM) was obtained from a peak profile at the setup (a) in Fig. 1, by setting a threshold level of −0.2 V and the PMT voltage of −500 V, as shown in Fig. 7. The single NE142 detector was thus applied

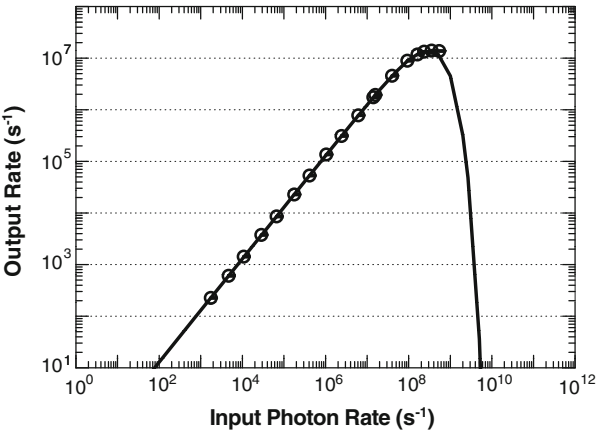

Fig. 5 Output count-rates vs. input photon rates, measured with the single NE142 detector at 57.6 keV

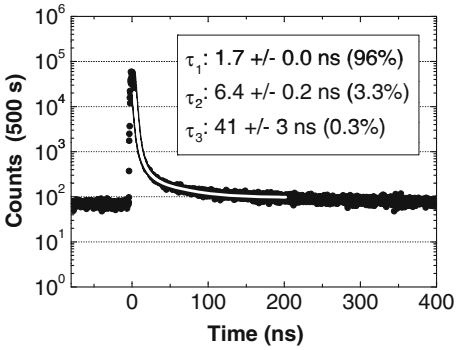

Fig. 6 Scintillation decay of NE142, recorded at 57.6 keV using a single-bunch mode of the PF ring

to the nuclear quasi-elastic scattering experiment on a potassium iodide solution, carried out at BL09XU of the SPring-8 (proposal No. 2009B1070). This fast response could separate delayed nuclear events from huge electronic scattering in a shorter period of ∼5 ns. The result in detail will be reported in another place.

2.2.2 The four-channel detector

For the Synchrotron Radiation (SR) based Mössbauer spectroscopy experiment [11], a multiple detector mounted with four plastic scintillators of NE142 (Detector C in Table 1) was developed. The scintillator size of one element was 17 mm in diameter and 10 mm thick. In order to obtain a larger solid angle, the scintillator size increased by a factor of about 14.3 in total area, compared with that of the Detector B. The thickness of the one element scintillator was also two times as large as that of the Detector B, to be more sensitive for high-energy X-rays. Figure 8 shows the arrangement of the detector elements and its measurement system. An aluminum window of 25 μm thick and 50 mm in diameter was put at the front of the brass cylindrical case.

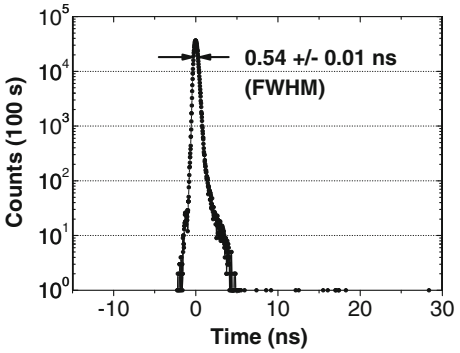

Fig. 7 Time spectrum of 57.6-keV X-ray beam profile, measured with the single NE142 detector. A time resolution (FWHM) was 0.54 ± 0.01 ns

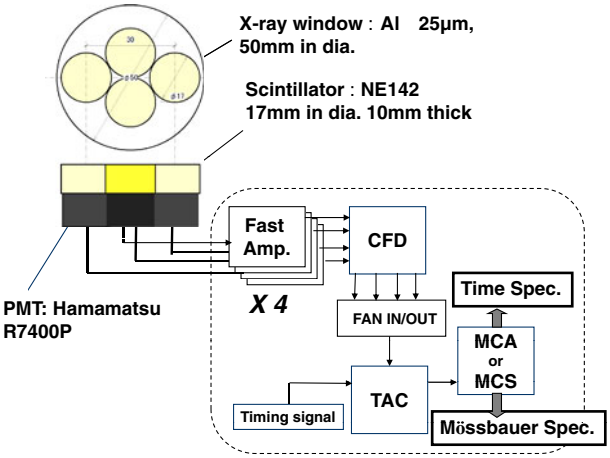

Fig. 8 Scintillator arrangement of the four-channel NE142 detector and its measurement system

The four-channel detector was applied to the Ni-61 SR based Mössbauer experiment on iron pnictide [12] at beamline BL09XU of the SPring-8 (Proposal No. 2010B1369). An enriched Ni doped iron pnictide of $Ba(Fe_{1-x}Ni_x)As_2$, x = 3% and 5% was a sample for our experiment. Coexistence of magnetism and superconductivity was reported on this compound [13]. Non-doped $BaFe_2As_2$ show antiferromagnetism below 130 K. The Neel temperature decreased with Ni doping, and superconductivity was appeared. At 5% Ni doping, the antiferromagnetism was disappeared and then only superconductivity was observed. The microscopic state of Fe and Ni atoms on this coexistence of magnetism and superconductivity is very interesting to elucidate the mechanism of the superconductivity.

Each of the samples x = 3% (150 mg) and 5% (107.5 mg), shaped with 7 mm in diameter, was set to a cryostat for liquid-helium flow cooling, as a transmitter. A piece of $Ni_{84}V_{16}$ alloy, 3.8 μm thick was used as a scatterer because the alloy show a single absorption peak at Mössbauer measurement. The scatterer was mounted on a copper block 30° inclined to the horizontal plane in the cryostat and was moved in the constant-acceleration mode using a transducer for Mössbauer spectroscopy. 86.2%-enriched Ni-61 was doped to all the samples.

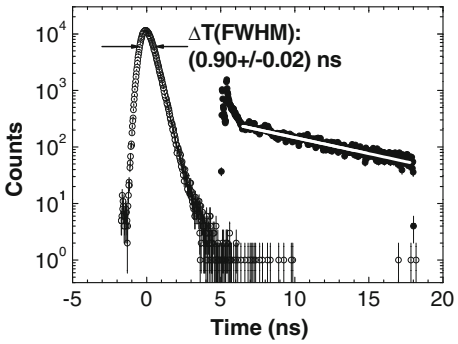

Fig. 9 Time spectra measured on the resonance with the prompt peak (open circles) and without the prompt peak (closed circles). The white line indicates a 7.7-ns decay curve fitted to the data

Table 2 A comparison of the Si-APD detector and the four-channel NE142 detector

	[A]Efficiency ([B]Int. eff.)	Delayed signals (cps), Time region (ns)
Si-APD (1 × 2 mm^2, 0.03 mm thick, 8 pixels)	[C]1.8% (42.1%)	13 cps, 5–18 ns
NE142 (17 mm dia., 10 mm thick, 4 elements)	9.3% (29%)	60 cps, 6.5–18 ns

[A]Considering each solid angle (4.3% for Si-APD, 35% for NE142).

[B]Absorption by an X-ray beam of 1 mm in diameter.

[C]This is given by Ni-K X-ray Absorption.

Time spectra on the resonance without the prompt peak (closed circles) and with the prompt peak (open circles) were recorded by radiation from the scatterer, as shown in Fig. 9. By using the timing system, detector counts were inhibited for the period of the prompt scattering, before 5 ns, as the spectrum of closed circles. A time resolution (FWHM) of the detector was known to be 0.90 ± 0.02 ns from the prompt peak in Fig. 9. A decay structure by nuclear radiation from the Ni-61 excited state was well observed without the intense prompt peak. A decay curve of 7.7-ns time constant, the white line in Fig. 9, well fitted to the measured data after 6.5 ns. Using the counts after 6.5 – 18 ns, a peak profile of the Ni-61 nuclear radiation around 67.4 keV was measured, while scanning energy of the incident X-ray beam. An FWHM of 7.6 ± 0.3 eV was observed for the peak profile, as shown in Fig. 10. This value means an energy width of the selected nuclear radiation, determined mainly by optics of the beamline. The net count-rate of the delayed nuclear radiation reached more than 50 s^{-1}, four times than that for a Si-APD detector (1 × 2 mm^2, 30 μm; 8 pixels) at almost the same experimental condition. Detection efficiency was calculated for the Si-APD and the present four-channel detectors by the simulation code of Geant4. The results are listed in Table 2. The distance between the center of detector window and the scattering point was assumed to be 5 mm in the calculation. The efficiency including geometry, of the NE142 detector was obtained to be 9.2% for 67.4-keV γ rays, while that of the APD detector was 1.8% for Ni KX-rays. These values well explained the higher count-rate for the delayed nuclear radiation with the detector C. With the multi-channel scaling of the delayed counts, we observed a Mössbauer absorption spectrum in Fig. 11, where open circles indicate the resonant absorption by the sample of x = 3%. The measured temperature was 12 ± 0.1 K

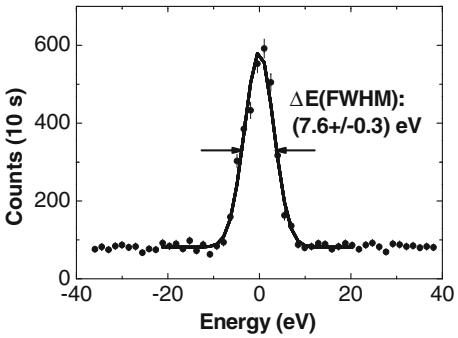

Fig. 10 Peak profile of nuclear resonant scattering, measured with scanning the incident photon energy around 67.4 keV. An energy width (FWHM) was observed to be 7.6 ± 0.3 eV

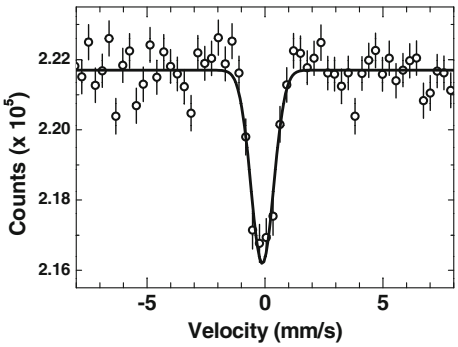

Fig. 11 Observed Mössbauer absorption spectra on a sample of $Ba(Fe_{1-x}Ni_x)As_2$, x = 3%

for the samples as the transmitter and 20.5 ± 0.6 K for the scatterer. The measuring time was about 52 hours. From Gaussian fitting to the absorption peak, drawn by the solid curve in Fig. 11, the peak width (FWHM) was obtained to be 1.1 ± 0.1 mm/s. Comparison with the result for the sample of x = 5% and the discussion will be presented in a future article by Y. Kobayashi et al..

3 Conclusions

Performances of our scintillation detectors have been reviewed and discussed for the NRS experiments. Fast decay time of the scintillation light is possible to select nuclear radiation from a sample to analyze with the time spectroscopy using synchrotron X-ray pulse. Compared with silicon avalanche photodiodes, scintillation detectors are useful for high-energy X-rays, especially in the measurements which need a larger solid angle and detection efficiency. The multi-element NE142 detector actually proved successful in the Ni-61 experiment with the SR-based Mössbauer spectroscopy. Further we are studying to obtain a scintillator with both a fast decay time and a larger absorption of X-rays. Simulation by Geant4 is quite effective for expectation of the detector performance and also for its evaluation. For improving noise rate from the existing photomultiplier tube, new development for a photosensor will also be important.

Acknowledgements The work was partly supported by the Japan Science and Technology Agency, CREST. The study for the NE142 detectors was performed with the approval of Japan Synchrotron Radiation Research Institute (JASRI) (Proposal No. 2009B1070 and 2010B1369) and of the Photon Factory Advisory Committee (Proposal No. 2008G104 and 2010G179).

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