A Fast X-Ray Detector Using a New Organic-Inorganic Perovskite Scintillator

We have developed a new scintillation detector for X-ray timing measurements. The organic-inorganic perovskite scintillator of phenethylamine lead bromide (PhE-$PbBr_4$) is used for sub-nanosecond time resolution and high detection efficiency. The performance of the scintillator was investigated using single-bunch operation of a Photon Factory (PF) ring. The dominant scintillation light had a fast decay time of 9.9 ns. The fast scintillation-decay was successfully applied to an X-ray detector equipped with a 0.9-mm-thick PhE-$PbBr_4$ crystal, which was used for nuclear resonant scattering measurements on ^{61}Ni (first excited level: 67.41 keV; lifetime: 7.6 ns).

In nuclear resonant scattering using synchrotron X-rays, sub-nanosecond time-resolution and nanosecond response are needed to distinguish nuclear radiation from intense prompt electronic scattering, by using time spectroscopy. A high detection efficiency for photons in which energy (E) is higher than 20 keV is also important for experiments on a nucleus for which an excited level exists at $E > 20$ keV. If a new scintillator with a short decay time of scintillation light, of less than a few nanoseconds and with a detection efficiency of > 10% at 50 keV, could be successfully produced, an X-ray detector fitted with the scintillator could be widely used instead of silicon avalanche photodiodes, which have a time resolution of $\Delta T \sim 1$ ns, but suffer from low efficiency, of a few percent at $E > 20$ keV.

In several scintillators of lead-halide-based perovskite-type organic-inorganic hybrid compounds, fast light emission caused by excitons was observed [1]. A relatively high efficiency of light emission was obtained, even at room temperature, owing to the quantum confinement effect of the low-dimensional structure consisting of metal-halide layers. The layered organic-inorganic perovskite compounds, $(n\text{-}C_mH_{2m+1}NH_3)_2PbX_4$ (m=3, 4, 6, 10, etc., X= Br or I) show a sub-nanosecond or a several-nanosecond component of scintillation light in an experiment using a short-pulsed electron beam [2]. However, a crystal of this type with a thickness greater than 0.2 mm and with a dimension larger than 3 mm^2 has not proved easy to grow. Instead, the $(C_6H_5C_2H_4NH_3)_2PbBr_4$ crystal [bis-(phenethylammonium) tetrabromoplumbate (II), abbreviated as PhE-$PbBr_4$, phenethylamine lead bromide] is expected to be a practically available scintillator. We successfully produced a single crystal of PhE-$PbBr_4$ (area: ~7 × 8 mm^2; thickness: 0.9 mm).

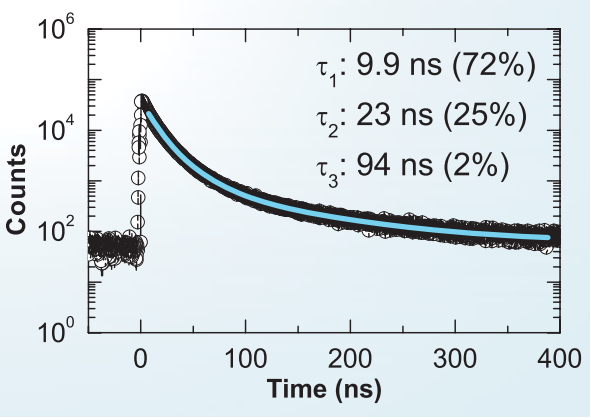

Figure 1
Scintillation decay of PhE-$PbBr_4$. The solid line was given by an exponential decay function of three decay-constants, used for least-squares fitting. Each component of the decay time is shown with a ratio of each light yield to the total emission.

The scintillation properties of PhE-$PbBr_4$ were investigated using synchrotron X-rays at BL-14A. The X-ray energy was set at 67.4 keV, which corresponds to the first excited level of ^{61}Ni. In order to measure the scintillation decay time, the scintillator crystal of PhE-$PbBr_4$ was placed 38 mm away from the entrance window of a photomultiplier tube (PMT; Hamamatsu R7400P) in a vacuum chamber. The detection probability of each scintillation light pulse was then much less than one, as the solid angle of the PMT was small enough. The decay time of scintillation light was measured during the single-bunch operation of the PF ring using a fast amplifier, a constant-fraction discriminator, and a time-to-amplitude converter. The open circles in Fig. 1 denote the scintillation decay of PhE-$PbBr_4$ obtained using the time spectroscopy system. Three components of the decay time τ were found to be 9.9 ± 0.2 ns (72 ± 1% of the total light yield), 23 ± 1 ns (25 ± 2%), and 94 ± 3 ns (2.3 ± 0.1%), by least-squares fitting using an exponential decay function (solid line).

An X-ray detector was assembled with the PMT in a cylindrical aluminum casing fitted with a beryllium window. The PhE-$PbBr_4$ scintillator was mounted onto the window of the PMT using optical grease, and then covered with Teflon tape. A charge-sensitive preamplifier was used to measure pulse-height spectra. The spectra of YAP:Ce (τ: 27 ns) having a good efficiency, and 5-wt% lead-loaded plastic scintillator (NE142, τ: 1.9 ns) having a fast response, were also obtained for reference using crystals measuring ϕ5 × 1 mm. The intrinsic efficiency for each crystal was determined by the counts detected by these crystals, compared with those detected by a NaI:Tl detector, which was fitted with a 5-mm-thick crystal. The efficiencies of PhE-$PbBr_4$, NE142, and YAP:Ce were found to be 23.7 ± 0.1%, 2.6 ± 0.1%, and 50.2 ± 0.2%, respectively. The light yields of PhE-$PbBr_4$ and NE142 were given by 22 ± 2 and 10 ± 1, respectively, when the peak channel of YAP:Ce was assumed to be 40 (that of NaI:Tl is 100).

From these results, it was confirmed that the PhE-$PbBr_4$ scintillator had a response faster than that of YAP:Ce, even though it was slower than that of NE142, keeping a good light yield and a high efficiency as well as YAP:Ce even for X-rays of 67.4 keV. With this detector, we could successfully record the decaying gamma rays emitted from ^{61}Ni with ΔT (FWHM) = 0.7 ns, which was limited by the PMT response [3]. The detector will be useful also in other high-energy X-ray experiments using high-rate counting or time spectroscopy.

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