

## ARTICLE

**Temperature effect in time structure of light emission in crystalline scintillators**Toshinori NAGASAKI\*, Masahiko UEYAMA, Masakatsu YOSHIOKA, Katashi KIYOHARA,  
Yusuke KOBAYASHI, Hiroki IWAMOTO, and Yusuke UOZUMI*Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University,  
744, Motoooka, Nishi-ku, Fukuoka 819-0395, Japan*

The time structure of light emission in crystalline scintillators, NaI(Tl), GSO(Ce) and LYSO(Ce) have been investigated in a high temperature region. Main experiments were made at extremely high-temperature by heavy-ion bombardment at relativistic energies. Supplemental experiments were made through the gamma-ray incidence at temperature of 27, 40, and 51°C with a conventional heater. The measured results were compared and discussed.

**KEYWORDS:** NaI(Tl), GSO(Ce), LYSO(Ce), time structure, decay curve, temperature dependence

**I. Introduction**

The light emission variations of crystalline scintillators have been a controversial subject for a long time. For instance, a great deal of effort has been made particularly on the scintillation efficiency of NaI(Tl) with bombardments by energetic heavy-ions at different stopping power  $dE/dx$ . Experimental results have shown that the scintillation efficiency of NaI(Tl) crystals to heavy-ions decreases with increasing  $dE/dx$ . The gross structure of their  $dE/dx$  dependence has been discussed by using a universal curve of the scintillation efficiency.

It is also a long time subject that NaI(Tl) shows the temperature dependence of the light yield. This dependence has been discussed in the last decade on the shape and the amplitude of light pulses emitted from the NaI(Tl) and other scintillators for a given energy of the incident ionizing radiation. The typical tendency is seen in the slow component whose intensity increases with decreasing temperature. This tendency has been interpreted in terms of the self-trap exciton formation<sup>1)</sup>.

From a view point to understand scintillation mechanism with heavy-ion bombardment, it is interesting to investigate the temperature dependence of the temperature dependence of the time profile. We firstly observe the time structure of light emission under different  $dE/dx$  conditions. If the  $dE/dx$  dependence is significant, a new development of particle-identification can be expected. The experiment is conducted at HIMAC of the National Institute for Radiological Sciences, Japan. Secondly, we observe the temperature dependence of gamma-ray excited scintillation time profile under various temperature conditions of scintillator at a laboratory of Kyushu University. By using a commercial heater, moderate high-temperature is reached.

The purpose of the present work is to observe the time structure of the light pulse of crystalline scintillators; NaI(Tl), GSO(Ce) and LYSO(Ce) with heavy-ion and gamma-ray incidence. To achieve accurate time-profile measurements a TOF-delayed-coincidence technique was applied.

**II. Experiments**

The experiments were carried out at the Heavy-Ion Medical Accelerator in Chiba (HIMAC) of NIRS and our laboratory. Both experiments used almost same measurement techniques that draw upon the Delayed-Coincidence Method<sup>2-3)</sup>. This method was developed by Bollinger and Thomas to measure the decay time profile of scintillators.

At HIMAC experiments, the beam energies were 230 MeV/u for Helium ions, 290 MeV/u for Carbon ions, and 650 MeV/u for Argon ions. The entire set up was shown schematically in **Fig. 1**. The scintillators used were NaI(Tl), GSO(Ce), and LYSO(Ce). These scintillators were of a cubic shape having a 50.8-mm, 48-mm, and 20-mm edge length, respectively. In **Table 1** are listed the parameters of used scintillator at present experiments. All six surfaces of these scintillators were mirror polished. One of the faces was contacted tightly to a photomultiplier tube (PMT) Hamamatsu R580, from which the time of radiation incidence was provided. The opposite side of this was wrapped with aluminum foil that has a small hole at its center, where a single photon is allowed to reach at a PMT (R6427) placed apart from the crystal to pick up the time information of scintillation. The other four surfaces were wrapped completely with aluminum foils for scintillation light reflection.

**Table 1**

The parameter of scintillators used at present experiments.

Scintillator	NaI(Tl)	GSO(Ce)	LYSO(Tl)
Density (g/cm <sup>3</sup> )	3.76	6.71	7.1
Decay time (nsec)*	230	56(fast) 400(slow)	41
Light output (arb.)	100	20	40
Size (mm)	50.8	43 (HIMAC) $\phi$ 25 × 5 (our lab. )	20

\*Measured at room temperature

\*Corresponding Author, E-mail:nagasaki@nucl.kyushu-u.ac.jp  
© Atomic Energy Society of Japan

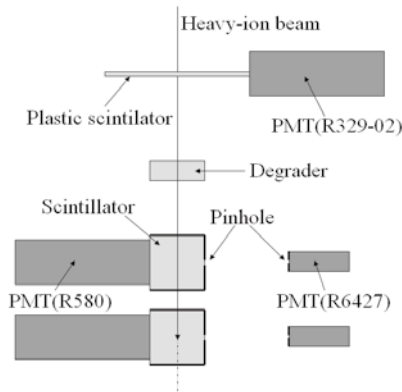


Fig. 1 Set up of the experiment at the HIMAC

Table 2

The expected specific energy losses calculated with the Bethe formula at the center of scintillator

		dE/dx (MeVcm <sup>2</sup> /g)		
		<sup>40</sup> Ar	<sup>12</sup> C	<sup>4</sup> He
NaI(Tl)	Penetrate	600	100	10
	Stop	44000	7000	940
GSO(Ce)	Penetrate	600	100	10
	Stop	34000	-	940
LYSO(Ce)	Penetrate	640	100	10
	Stop	38000	4900	770

In order to choose suitable particle energies, degraders were placed in front of the scintillation detectors, to make particles stop at the center of the second crystal. The energy of penetrate beam was measured with the front crystal and the stop beam energy was measured with the rear one. In Table 2 are listed the expected specific energy losses calculated with the Bethe formula.

The set up of experiment which was carried out at our laboratory is shown schematically in Fig. 2. The <sup>137</sup>Ce gamma-source was placed under the PMT(R580), and the GSO(Ce) scintillator (ϕ 25 × 5 mm) was optically coupled to this PMT. The opposite side of this scintillator surface was wrapped with aluminum foil that has a diameter of 1.0-cm hole, and the other surface was mirror polished and wrapped with aluminum foil. The temperature was controlled by a heater shown schematically in Fig. 2 and monitored to be stable within ~ 0.1°C preset value by a small thermometer attached on the scintillator. The range of temperature at this experiment were from room temperature to about 50°C.

Both experiments, charges of linear signals from PMT were digitized with CAMAC ADCs and taken by a PC system.

### III. Results and discussion

The representative results from present experiment at HIMAC are displayed in histogram forms in Figs. 3-5 with the <sup>137</sup>Cs gamma-ray source result at room

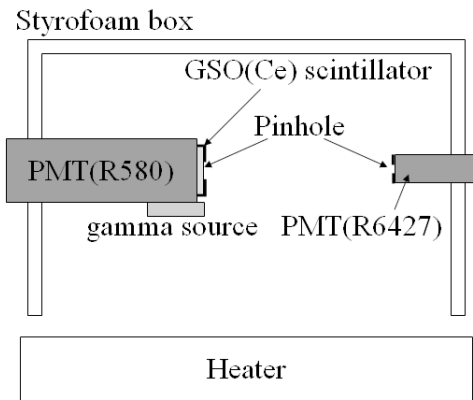


Fig. 2 Set up of the experiment at a laboratory of Kyusyu University

temperature. Each one is normalized at the peak value to make the comparison easier and clearer.

In Fig. 3 is shown a decay curve for <sup>4</sup>He incidence at an energy 230 MeV/u, which stopped at the center of NaI(Tl) scintillator(He\_stop) and that for gamma-ray(gamma-ray). The comparison of two, the decay curve of He\_stop is smaller than that of gamma-ray from 30 nsec to 1000 nsec. In Fig. 6 is shown the part which was confirmed this difference for <sup>40</sup>Ar, <sup>12</sup>C, and <sup>4</sup>He incidence data of present works. Except for the decay curve of He\_penetrate, all of decay curves are almost same in shape. By comparison the data and the value of Table 2, the decay curve of NaI(Tl) scintillator depends on the dE/dx and the curve is changing at 100(MeVcm<sup>2</sup>/g) dE/dx or less.

In Fig. 4 is shown a decay curve for <sup>40</sup>Ar beam of 650 MeV/u, which penetrates at the center of GSO(Ce) scintillator(Ar\_penetrate) and a decay curve for gamma-ray(gamma-ray). The relative intensity of Ar\_penetrate is larger than that of gamma-ray from 300 to 1000 nsec. This range is enlarged and shown in Fig. 7

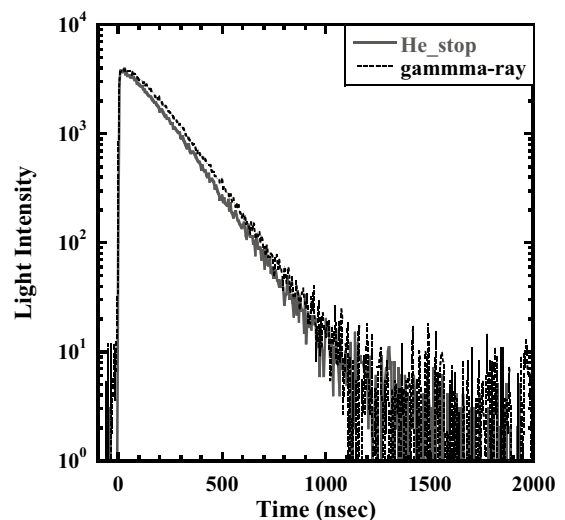


Fig. 3 Typical time structure of scintillation from NaI(Tl) for He\_stop and gamma-incidence.

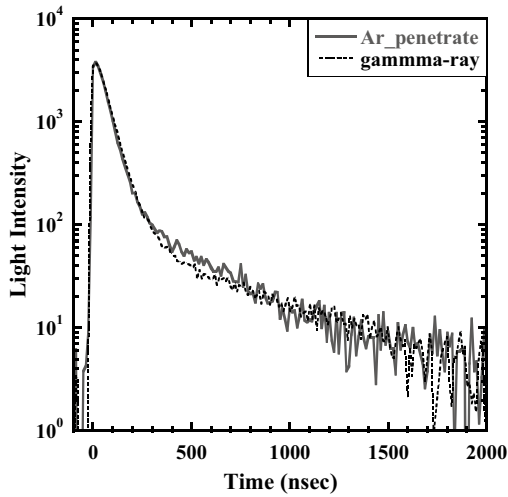


Fig. 4 Typical time structure of scintillation from GSO(Ce) for Ar\_penetrate and gamma-incidence.

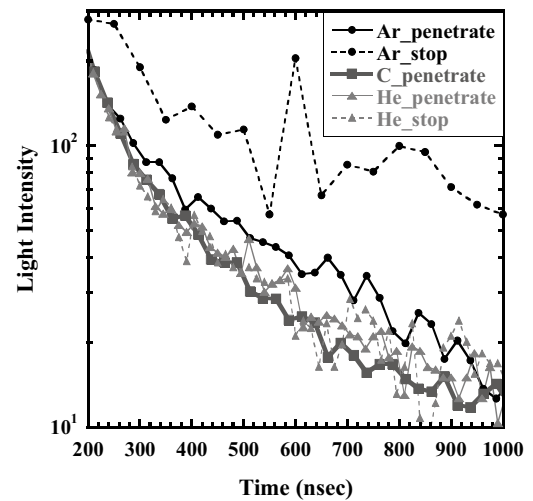


Fig. 7 Measured time structure of scintillation from GSO(Ce) in a time range between 200 and 1000 nsec.

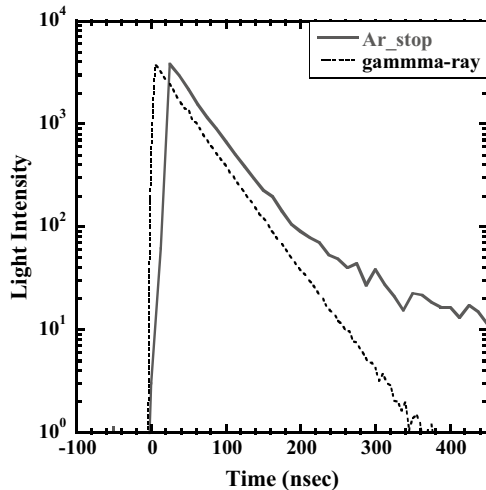


Fig. 5 Typical time structure of scintillation from LYSO(Ce) for Ar\_stop and gamma-incidence.

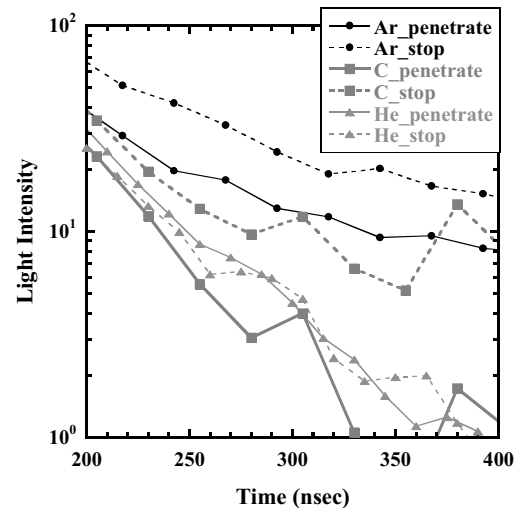


Fig. 8 Measured time structure of scintillation from LYSO(Ce) between 200 and 400 nsec

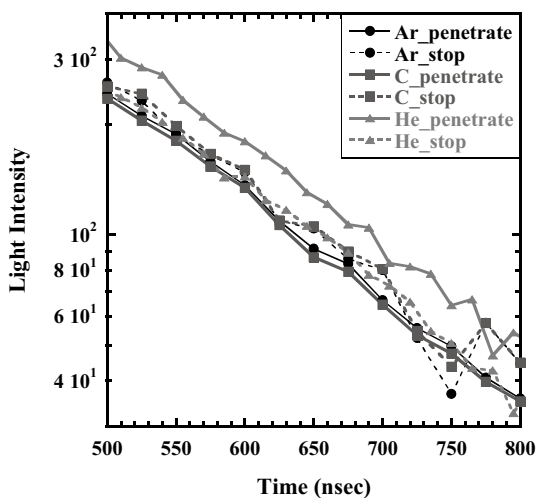
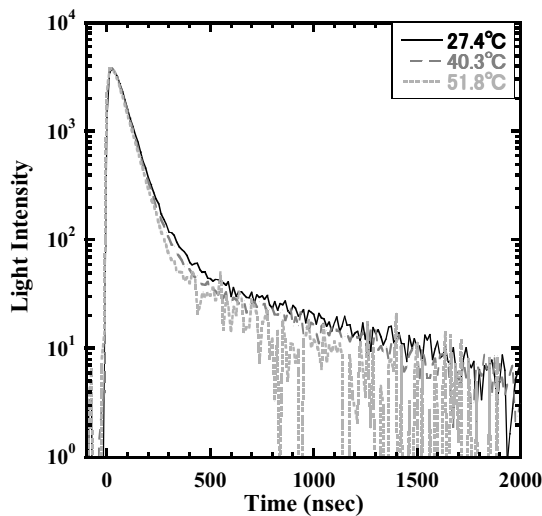


Fig. 6 Measured time structure of scintillation from NaI(Tl) in a time range between 500 and 800 nsec

for all experimental data. The relative intensity for Ar\_stop is the largest in this slow component range and that for Ar\_penetrate is the second. The others, He\_penetrate, He\_stop and C\_penetrate are of almost the same in this time range. As shown in **Table 2**, the  $dE/dx$  value of He\_stop is larger than that of Ar\_penetrate. The present result should imply that the decay constant depends on not only  $dE/dx$ . Actually, the scintillation efficiency is widely known to depend on the particle identity as well as  $dE/dx$ .

In **Fig.5** is shown a decay curve for  $^{40}\text{Ar}$  beam of energy 650 MeV/u, which stopped at the center of LYSO(Ce) scintillator (Ar\_stop) and that for gamma-ray (gamma-ray). Rather large differences can be seen between these two cases. The rise time for the Ar\_stop is longer than that for gamma-rays, as known widely; however, the Ar\_stop result reveals two decay

components. **Fig. 8** shows the enlarged part of decay curves for all experimental data. In this time range, the relative intensity of Ar\_stop is largest. Those of Ar\_penetrate and C\_stop are the next. And the others, He\_penetrate, He\_stop, and C\_penetrate follow with similarity in this range. Along with GSO(Ce) scintillator, the value of  $dE/dx$  of He\_stop is larger than that for Ar\_penetrate in the LYSO(Ce) scintillator. But the value of the decay curve for He\_stop is lower than that for Ar\_penetrate. We expected that the decay curve of these scintillators depends on not only  $dE/dx$  of incident particle but also the characteristics of the incident particle.



**Fig. 9** Measured time structure of scintillation from GSO(Ce) at 27.4°C, 40.3°C, and 51.8°C

The temperature dependence of the gamma-ray excited scintillation time profile of GSO(Ce) scintillator, which was measured at a laboratory of Kyusyu University, is shown in **Fig. 9**, where the comparison is made between temperatures of 27.4°C, 40.3°C, and 51.8°C. As the temperature of the GSO(Ce) scintillator increases from room temperature, the slow component appears to be suppressed. No difference is seen in the fast component. It is the opposite tendency compared with that of heavy-ion incidence providing different  $dE/dx$  conditions, if one considers the larger  $dE/dx$  corresponds to the higher temperature.

#### IV. Conclusion

We observed the time structure of light emission of inorganic scintillator NaI(Tl), GSO(Ce), and LYSO(Ce) under different conditions. The  $dE/dx$  dependence was investigated at HIMAC by using heavy-ion beams. The temperature dependence was also studied for gamma-ray incidence at temperature 27°C, 40°C and 51°C.

The scintillation time profiles of all crystals have appeared to change with both  $dE/dx$  and temperature. From the pulse shape analyses, we have found that these results cannot be interpreted by the decay time change of a single component, but they should be attributed to the change of the slow component intensity. The first component is insensitive to  $dE/dx$  and temperature.

In the case of the GSO(Ce) scintillator for instance, the results of ion-bombardment experiments indicate that the slow component becomes smaller with  $dE/dx$  larger. The temperature dependence of the slow component has been found to decrease with increasing temperature. These results are rather unreasonable because the  $dE/dx$  dependence would be similar to the temperature dependence, because a local heating is expected around the ion track. It should be noticed that other crystals show different dependences from each other. The variation of absolute scintillation yields cannot be discussed, since we measured just the relative scintillation intensity.

#### Acknowledgment

Authors gratefully acknowledge Dr. M. Imamura for useful information and discussion. The present study was conducted under the HIMAC program number 20P239.

#### References

- 1) B. S. Alexandrov, K. D. Ianakiev, P. B. Littlewood, "Branching transport model of NaI(Tl) alkali-halide scintillator," Nuclear Instruments and Methods in Physics Research Section A, 586, 432 (2008).
- 2) L. M. Bollinger, G. E. Thomas, "Measurement of the Time Dependence of Scintillation Intensity by a Delayed-Coincidence Method," The Review of Scientific Instruments, 32, 1044 (1961).
- 3) N. Tsuchida, M. Ikeda, T. Kamae, et al., "Temperature dependence of gamma-ray excited scintillation time profile and light yield of GSO, YSO, YAP and BGO," Nuclear Instruments and Methods in Physics Research Section A, 385, 290 (1997)
- 4) G. F. Knoll, "Radiation Detection and Measurement third edition," 2001