Abstract—Pr:LuAG has interesting properties of high density, high light yield and a very fast 5d-4f emission decay time. Recently we have developed 2-inch-diameter Pr:LuAG single crystal for scintillator applications such as medical imaging and high-energy physics. In this report, we study uniformity of the light yield, energy resolution, decay time and γ-ray response in the range from 59.5 keV (241Am) to 1.4 MeV (241Am).

I. INTRODUCTION

Pr3+ ion shows fast 5d-4f emission in several host materials and such systems can be candidate for high figure-of-merit scintillator[1,2]. Recently our group intensively examined scintillation properties of several Pr-doped compounds [3-7]. Among those materials, Pr:Lu3Al5O12 (Pr:LuAG) was found to have interesting properties of high density (6.7g/cm3), high light yield (three times higher than Bi4Ge3O12 (BGO)), good energy resolution and a very fast 5d-4f emission decay time (20ns)[5,6]. Owing to these excellent scintillation properties, Pr:LuAG scintillator can be used for applications in medical imaging and high-energy physics. In order to apply Pr:LuAG scintillators to such applications, we must establish mass production of Pr:LuAG single crystals with uniform scintillation performance in whole crystal.

In this report, we establish single crystal growth of 2-inch-diameter Pr:LuAG by the Czochralski (CZ) method. We report their scintillation characteristics such as light yield, energy resolution, γ-ray response and decay time.

II. EXPERIMENTAL PROCEDURE

1) Crystal Growth

Starting materials used in this study were Pr6O11, Lu2O3 and α-Al2O3 with a purity of 99.99%. Pr6O11 was added to the starting material for luminescent centers as Pr3+. Nominally, Lu3+ was substituted by Pr3+ according to the formula of (Pr0.025 Lu0.975)3Al5O12.

Pr:LuAG single crystals were grown by CZ method with an RF heating system. The rotation rate was 4-12 rpm and the growth rate was 1.0 mm/h. An automatic diameter control system by crystal weighing was applied to control growth parameters. Crystals were grown from an Ir crucible 100mm in diameter and 100mm in height. Ar atmosphere was used to prevent oxidization of the crucible. The seed crystal was [100] oriented Pr:LuAG. After the completion of growth, the grown crystal was taken off from the melt and was gradually cooled down to room temperature.

2) Light output measurements

Several pieces with 2.45x5.1x15mm size were cut along the growth axis. Every surface was mechanically polished. Because generally used reflectors, such as ESR film or Teflon, are not suited to collect UV photons, we coated sample pieces with BaSO4 reflector, which is improved to be suited well for Pr:LuAG. The 2.45x5.1 face of the pieces was coupled with the PMT using optical grease (OKEN, 6262A). To determine light yield, the energy spectra were collected under 662 keV γ-ray excitation ( 137Cs source), using an amplifier with a shaping time of 0.5 μs, a photomultiplier (Hamamatsu R6331) and a multichannel analyzer in the pulse height mode.

3) Rising and Decay Time Measurements

Starting rise and decay spectra were measured by using the conventional single photoelectron technique with a pulsed X-ray source for the excitation. The X-ray pulses were produced in a light-excited X-ray tube N5084 (Hamamatsu Photonics) with a tungsten anode at a high voltage of 20 kV between photocathode and anode. The excitation light pulse was made in a laser diode with a width of ~40 ps (in FWHM) at a repetition rate of 100 Hz. The generated X-rays were collimated on to the single crystal sample. The scintillation light was detected by a photomultiplier (PMT) R4998 (Hamamatsu, bialkali photocathode, borosilicate glass window). The light intensity at the PMT was made much smaller than the level of one photoelectron per pulse so as to avoid the distortion of the decay spectrum. The timing of the
PMT output signal was registered in a CAMAC TDC with respect to the initial clock pulse which triggered the laser-diode using InGaN: Picosecond Injection Laser (PiLAS, Advanced Laser-diode Systems GmbH, Berlin). The wavelength is nominally 409.5 nm while the pulse width is ~40 ps in FWHM.

4) γ-ray Response Measurements

Several pieces with 2mmx2mmx15mm size were cut along the growth axis. Every surface was mechanically polished. These pieces were coated with BaSO₄ reflector. Figure 1 shows Pr 0.22 mol% doped LuAG sample and APD S8664-55.

We optically coupled scintillators to APD (Hamamatsu, S8664-55) with silicone grease (OKEN, 6262A). The signal is fed into pre-amplifier (CP580K), shaping amplifier (CP 4417), pocket MCA (Amptec 8000A), and finally to the PC. The bias for APD is supplied by CP 6641. Because APD is very sensitive to an environmental temperature, we control the temperature by a heat bath within ± 0.5 K.

III. CRYSTAL GROWTH OF 2-INCH-DIAMETER Pr:LuAG

Growth conditions such as corn angle, rotation rate, and heat insulation design in the furnace were optimized for obtaining crack-free 2-inch-diameter Pr:LuAG crystals with uniform light yield in whole crystal. I referred to some reports for the optimization of the growth conditions [8-11].

2-inch-diameter Pr2.5%:LuAG single crystal with a length of 160 mm was grown with a nominal composition of (Pr₀.₀₇₅Lu₂.₉₂₅)Al₅O₁₂ (Fig. 2). The solidification fraction of Pr:LuAG was about 45% of the melt of raw material in the crucible.

IV. SCINTILLATION PROPERTIES

1) Uniformity of Light yield

Typical energy spectra of Pr2.5%:LuAG excited by ¹³⁷Cs at room temperature are shown in Fig.3. photo-absorption peak was fit with a single Gaussian function to estimate an energy resolution. The energy resolution was 7.8%. The light yield of

![Typical energy spectrum of Pr2.5%:LuAG excited by ¹³⁷Cs](image)

Pr:LuAG was around three times higher than that of a conventional BGO sample with 2.45mmx5.1mmx15mm size. The dependence of light output on solidification fraction (g) is shown in Fig.4. The standard deviation of the light yield was
2) Rising and Decay Time

Rising time spectra of Pr2.5%:LuAG sample excited by pulsed X-ray at room temperature are shown in Fig. 5. Pr2.5%:LuAG shows very fast rising time of around 0.4ns. Decay time spectra of Pr2.5%:LuAG are also given in Fig.6. The dominant component was about 13.1 ns, which is much shorter than the scintillation decay time of other oxide scintillators (BGO:300 ns, LSO:40 ns). This together with a noticeable presence of slower decay components (55.0 ns) points to retrapping processes and delayed radiative recombination at Pr\(^{3+}\) emission centers.

3) \(\gamma\)-ray Response

An adequate avalanche gain to achieve the best energy resolution is ~20 for this APD, we supplied high voltage of 310V (avalanche gain =19.4) at 20°C. A shaping time of a shaping amplifier is set to be 2 \(\mu\)s through this work.

The sample was excited by \(^{241}\)Am, \(^{22}\)Na, \(^{137}\)Cs, \(^{133}\)Ba, and \(^{152}\)Eu. Then, each photo-absorption peak was fit with a single Gaussian function, and estimate an energy resolution at each energy. As shown in fig. 7, 662 keV photo-absorption peak is clearly detected by using 2.2mm x2.2mm x10mm size Pr:LuAG sample. This sample shows 5.7% of energy resolution. The peak was fit with a single Gaussian function to estimate an energy resolution. The energy resolution was
V. CONCLUSION

We demonstrated 2-inch-diameter Pr:LuAG single crystal with a length of 160mm by the Cz method. The solidification fraction was about 45% of the melt of raw material in the crucible. The grown Pr:LuAG crystals have homogeneous light yield which is around three times higher than that of a conventional BGO. The 2.45mmx5.1mmx15mm size sample shows energy resolution of 7.8% measured by photomultiplier (Hamamatsu R6331) using 137Cs excitation. Pr2.5%:LuAG shows very fast rising time of around 0.4ns excited by pulsed x-ray at room temperature. Decay time was about 13.1 ns, which together with a noticeable presence of slower decay components (55.0 ns decay time). Using 2.2mm x2.2mm x10mm size sample coupled with high QE APD, energy resolution of 5.7% at 662 keV is achieved, and this value is better than PMT use. Pr:LuAG shows good linearity between energy and pulse height within around 2% of the standard deviation in the range of 59.5 keV (241Am) to 1.4 MeV (241Am).