

Significant improvement of GAGG:Ce based scintillation detector performance with temperature decrease

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Abstract— This report presents results on the significant improvement of GAGG:Ce based scintillation detector performance with temperature decrease. When temperature of a PMT based detector is lowered to -45°C, its amplitude response at registration of γ -quanta is improved by 30%; FHHM was found to be better up to factor of 0.85, whereas scintillation kinetics become even faster in crystals co-doped with magnesium and magnesium and titanium. All this opens an opportunity for a wide application of GAGG scintillation detectors, particularly in a combination with SiPM photo-sensors, which signal-to-noise ratio would also improve with temperature decrease.

Index Terms—Inorganic scintillation material, GAGG scintillator, γ -quanta, photo-sensor

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1. Introduction

Due to a high light yield of up to 50000 phot/MeV, a short luminescence decay time (< 100 ns) [1], and good matching of the emission band with the sensitivity spectrum of conventional SiPMs, Ce doped $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}$ crystal (GAGG) is promising scintillation material for medical imaging and might compete with Ce-doped Lu_2SiO_5 crystal for PET application. Moreover, GAGG:Ce, co-doped with Mg, shows spectacular time resolution at different excitation [2,3]. GAGG is dense high light yield scintillator, hence it can be applied for a high resolution γ -radiation spectrometry similar to recently developed halide scintillators [4]. However, wide application of the material in detectors is limited. The material exhibits strong phosphorescence, both under photo-excitation and excitation by ionizing radiation. It is demonstrated that the phosphorescence might be diminished by co-doping of GAGG:Ce crystals with Mg [5]. However, contrary to many other scintillators [6-8] the co-doping of the GAGG material with the second group di-valent ions from the set of Mg, Ca, Sr results in a lower scintillation light yield.

Recently, we showed that light yield deterioration of the crystal at codoping with Mg is accompanied with a strong acceleration rate of the free carriers non-radiating recombination [9]. This effect becomes competing to a radiating recombination of free carriers via Ce^{3+} ions and, thus, results in decrease of the scintillator light yield. Non-radiative recombination occurs when hole, due to a migration, appears in the vicinity of the Mg^{2+} created defect. Moreover, some energy barrier in the vicinity of the recombination center has to be overcome by hole for a recombination. Both, migration rate and an ability to overcome barrier are dependent on the temperature. Thus, possible solution to recuperate light yield loss is a slowing down of the holes mobility. Due to a strong temperature (T) dependence of the both effects $\sim \exp(-E/kT)$, where E is the constant, dedicated to migration rate or transmission through the barrier, which are defined by the nature of the compound, k-Boltzman constant, this can be achieved by cooling of the crystal or the whole detecting unit.

Cooling of the scintillation material to gain light yield works pretty good in self-activated scintillation materials which structural units, oxy-anionic complexes, possess highly temperature quenched luminescence. There are a reasonable amount of self-activated materials which includes widely used PbWO_4 and $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ [4]. Note, last modification of PbWO_4 , PWO-II,

which is the crystal, doped with La, and Y at the total amount of less than 100ppm. Doping ions create shallow electron capturing centers in the crystal matrix and prevents free carriers capture by deep traps, significantly improving tolerance of the PWO-II to ionizing radiation. Cooling of the crystal to the - 25°C allows to triplicate its light yield at simultaneous keeping the scintillation kinetics fast enough [10].

Contrary to self-activated material, the luminescence of Ce-doped scintillation material is caused by inter-configuration d-f luminescence, having a high quantum yield and low temperature quenching effect in the vicinity of room temperature. Thus, only a minor gain of the light yield of the Ce activated scintillation material is expected with decrease of the crystal temperature. Moreover, some of oxide scintillators doped with Ce, particularly perovskites of $\text{YAlO}_3\text{-LuAlO}_3$ family, demonstrate 10-20% decrease of the light yield when temperature is lowered from room to - 20°C. Here we report an incredible improvement of GAGG:Ce,Mg and GAGG:Ce, Mg, Ti scintillation detectors performance with temperature decrease.

2. Samples

All the garnet samples under study were cut from single crystal boules, grown by the Czochralski method from iridium crucibles by Fomos Crystals from the melt close to $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}$ stoichiometric composition. To compensate gallium leakage from the melt during the crystal growth, samples were grown with excessive Ga_2O_3 added into the melt in the crucible. The reference sample (S1) was doped with Ce (0.5 at.%) whereas two other studied crystals, S2 and S3, were co-doped by Mg (0.1 at.%) and Mg (0.05 at.%) and Ti (0.01at.%) correspondingly. Pulse height spectra of ^{137}Cs source with 662keV energy of gamma-quanta have been measured with bialkali green-extended PMT Hamamatsu R329 in a thermostat within temperature range from +20°C to -45°C, with temperature stabilization accuracy 0.1°C. Light was measured with bialkali PMT Hamamatsu R2059 in different time gates of charge sensitive gated ADC. Stability of PMT - photocathode sensitivity and PMT gain in the temperature range was controlled by a reference green LED light pulse source. Sample size was 15x18x7mm. We used Basyon[®] optical grease and Teflon[®] light reflectors.

3. Experimental results and discussion

Change of the photo-peak position (i.e. light yield change), and change of energy

resolution with temperature were measured in different time gates. Figure 1 represents pulse height spectrum of ^{137}Cs gamma-source measured with S3 sample at four temperatures: $+20^\circ\text{C}$, 0°C , -20°C , -45°C . Cooling of the detecting unit results in improvement both, peak position and energy resolution as well. Figure 2 shows change of the normalized to room temperature and measured in 1000 ns gate FWHM of 662keV photo-peak with temperature. The most prominent improvement has been achieved with the sample co-doped with Mg, whereas Ce doped and Ce doped and co-doped with Mg and Ti showed similar behavior.

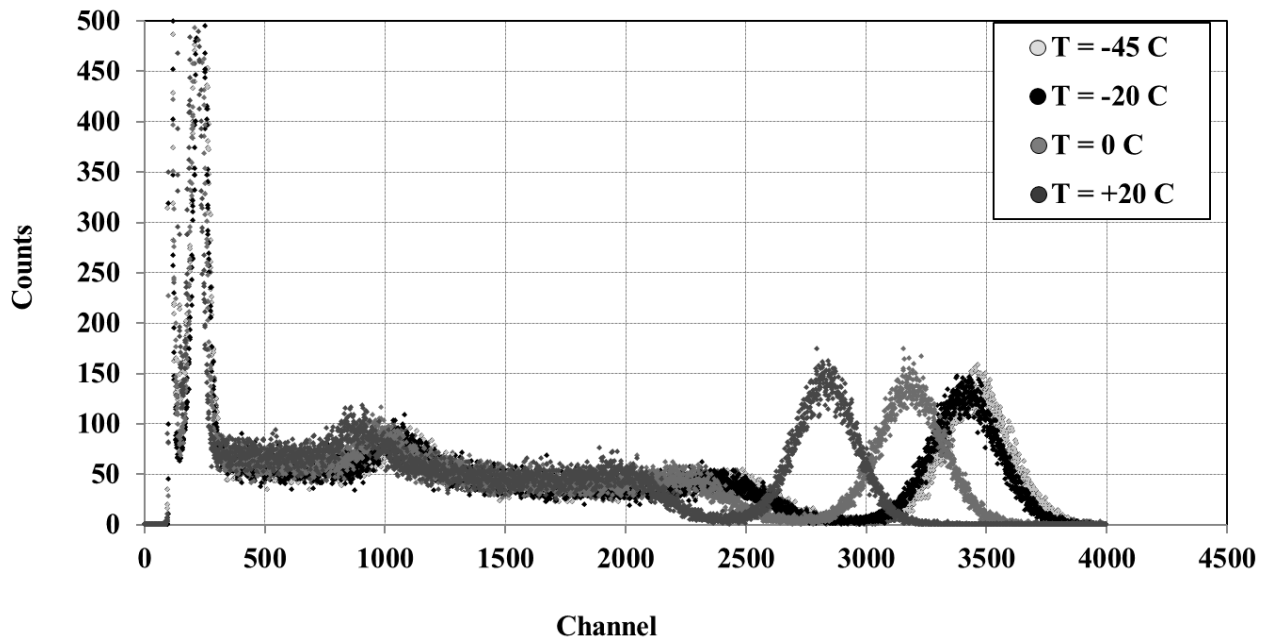


Fig.1. Pulse height spectrum of ^{137}Cs gamma-source measured with S3 sample at four temperatures: $+20^\circ\text{C}$, 0°C , -20°C , -45°C .

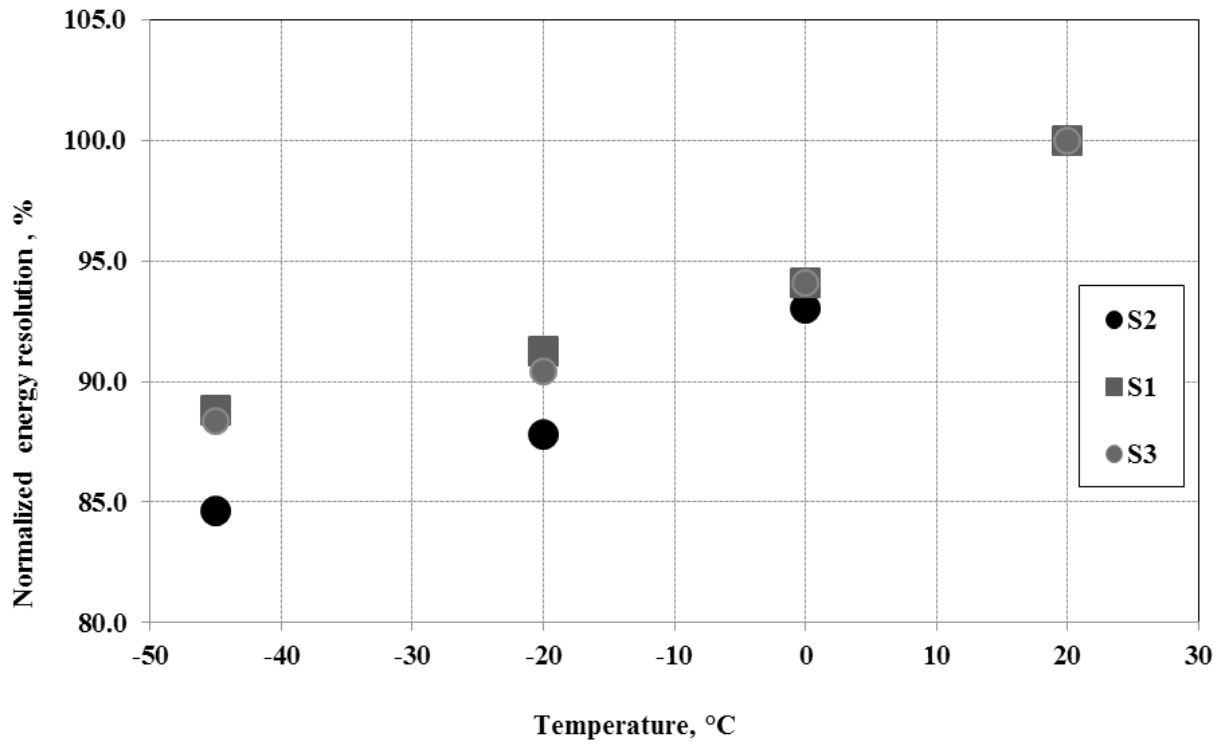


Fig.2. Temperature change of energy resolution FWHM at 662 keV normalized to room temperature. Measurements have been performed with 1000 ns time gate.

Figures 3-5 show light yield of the samples in various time gates, measured at different temperatures. It worth to note that largest improvement of the LY at cooling is observed in the crystals, co-doped with Mg and Mg, Ti. Figures 6-8 show relative change of the light yield, normalized to 20°C, versus time gate at different temperatures.

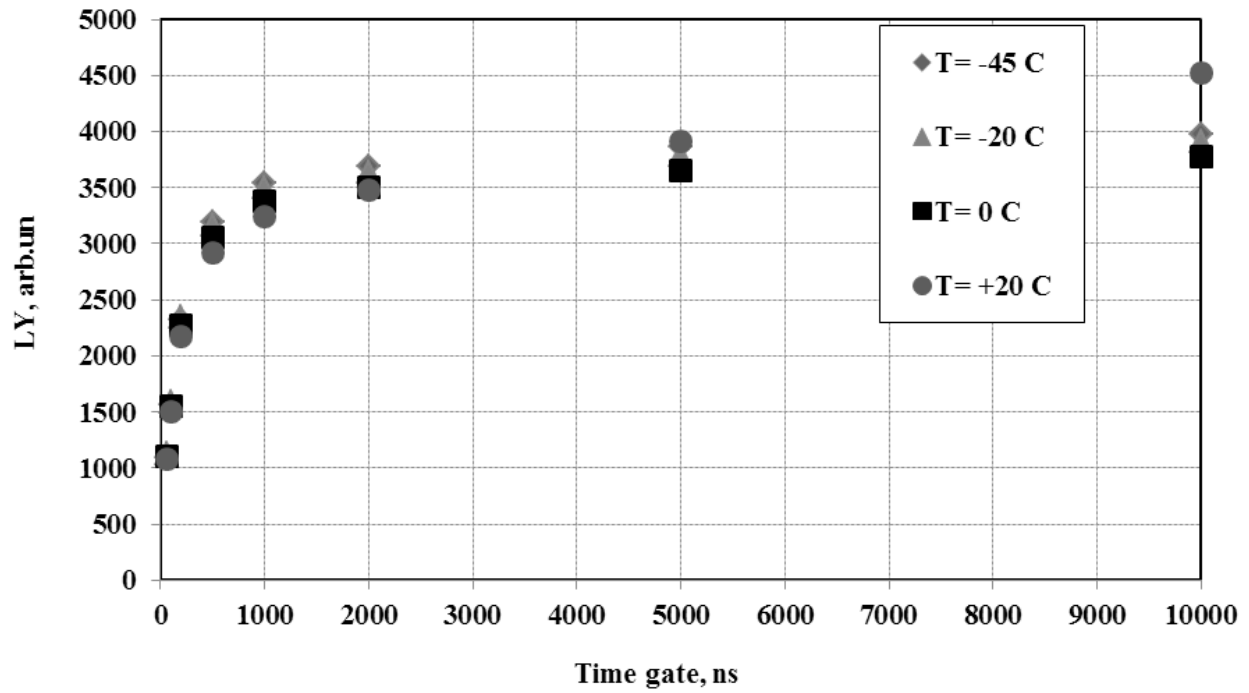


Fig.3. Light yield of GAGG:Ce (S1) sample measured in different time gates in temperature range.

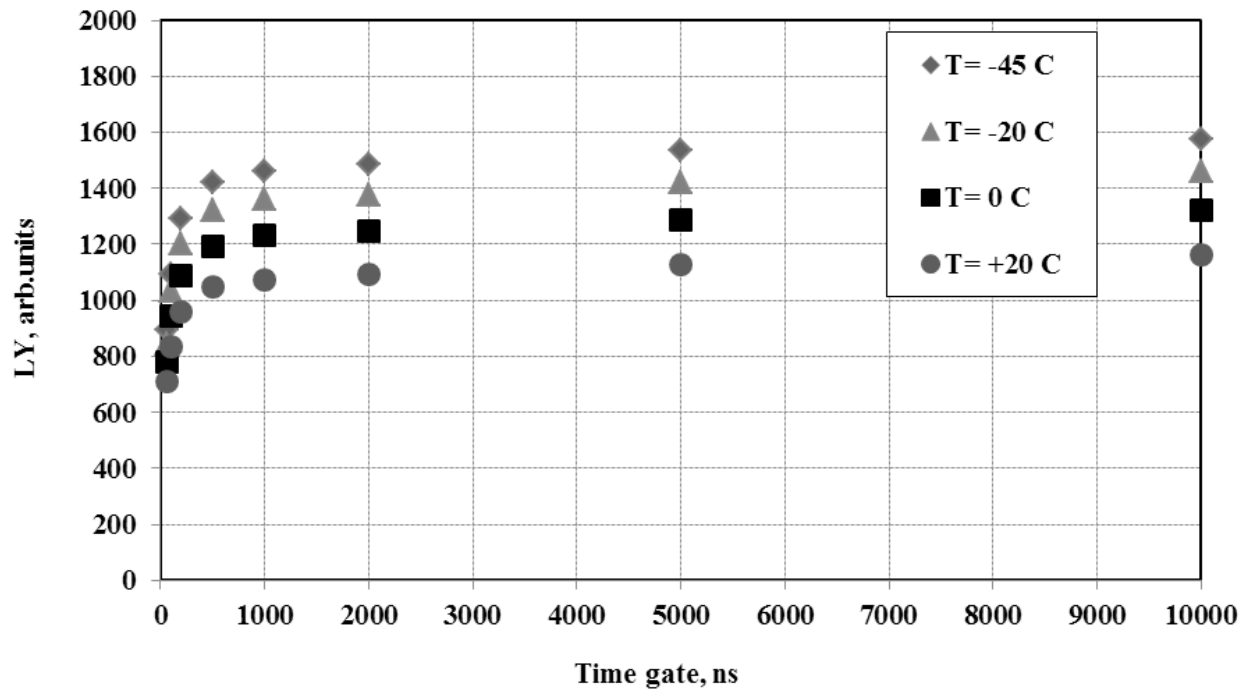
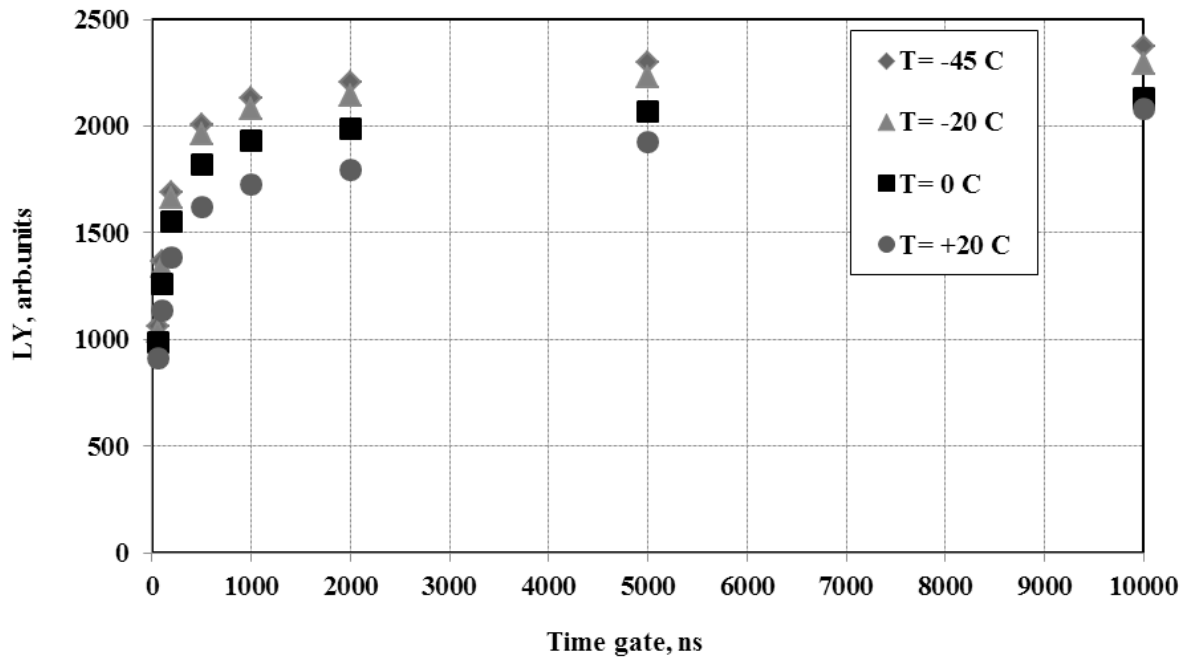
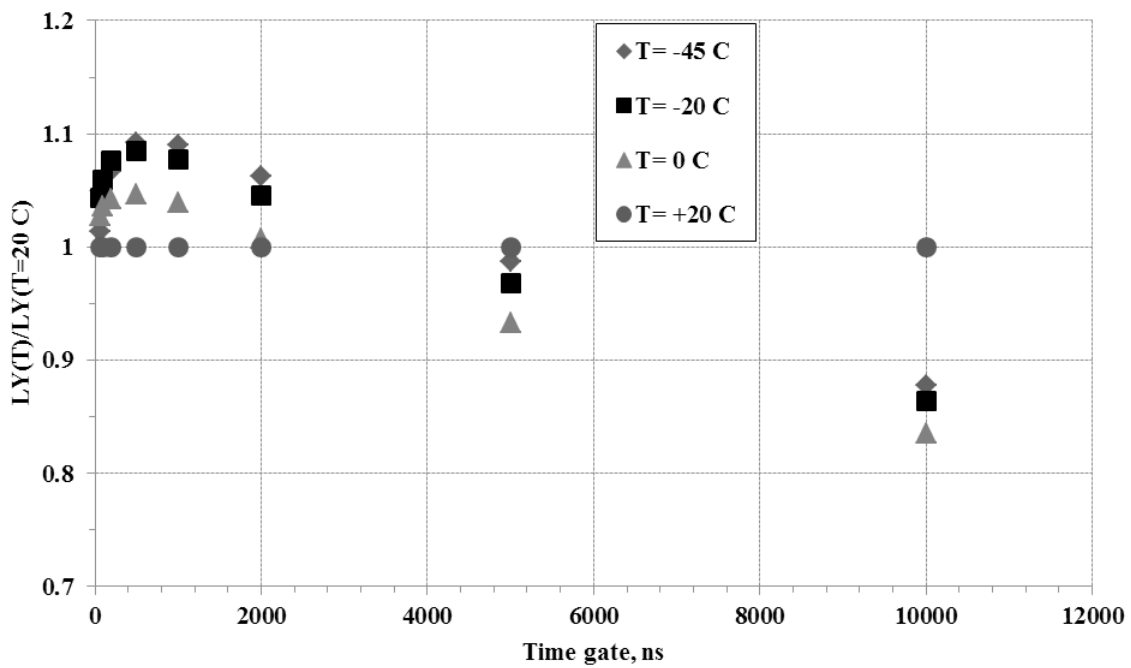


Fig.4. Light yield of GAGG:Ce, Mg (S2) sample measured in different time gates in

128 temperature range.



129
130 **Fig.5.** Light yield of GAGG:Ce, Mg, Ti (S3) sample measured in different time gates in
131 temperature range.



132
133 **Fig.6.** Relative change of the light yield of GAGG:Ce (S1) sample normalized to 20°C
134 versus time gate at different temperatures.

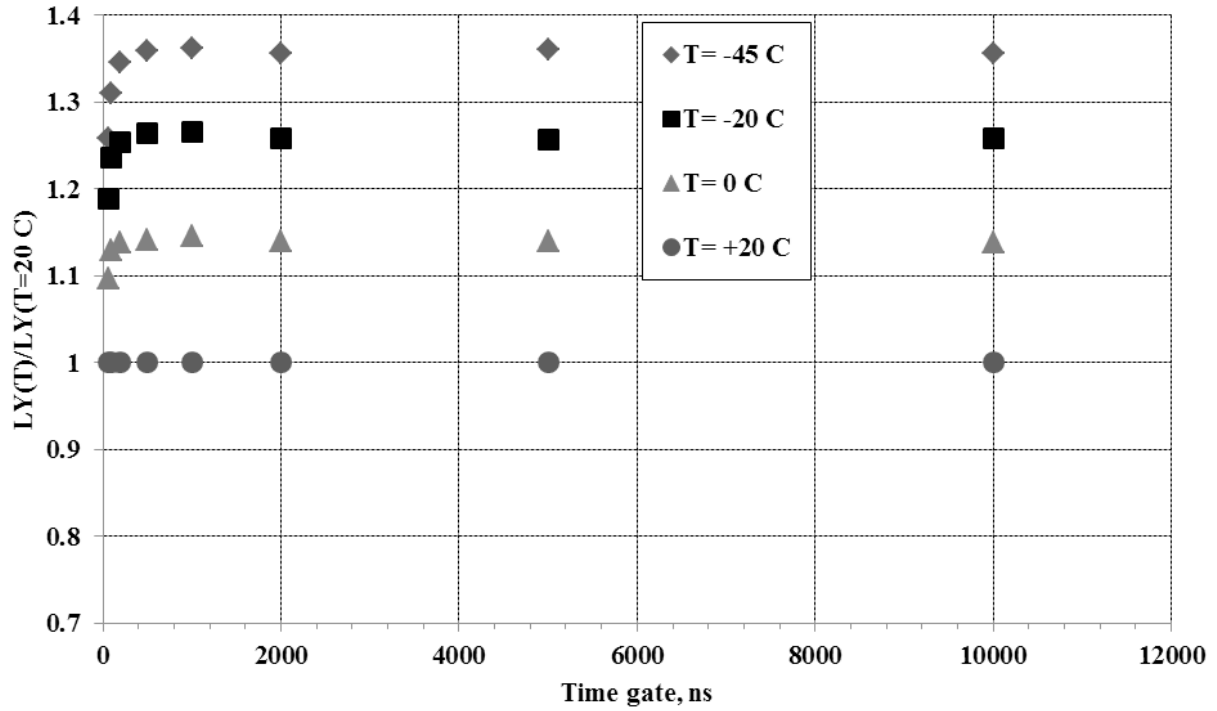


Fig.7. Relative change of the light yield of GAGG:Ce (S2) sample normalized to 20°C versus time gate at different temperatures.

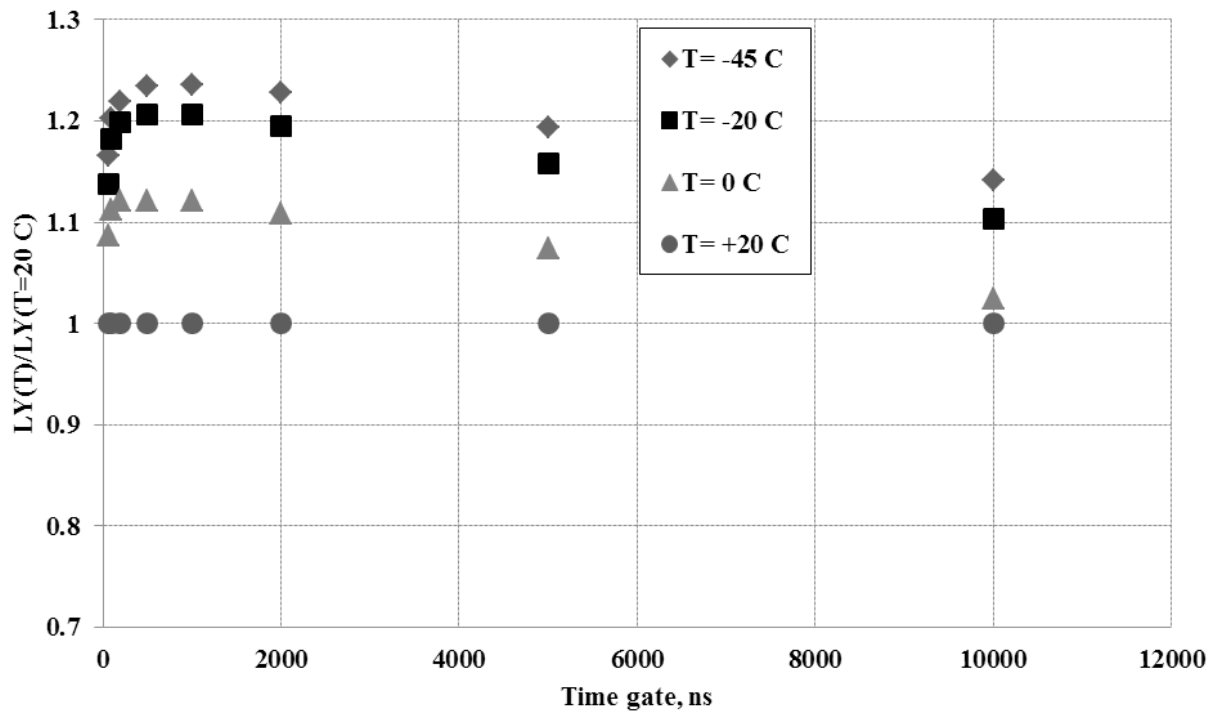


Fig.8. Relative change of the light yield of GAGG:Ce (S3) sample normalized to 20°C versus time gate at different temperatures.

As it is seen, crystal doped only with Ce shows the lesser relative light yield increase at cooling. Moreover, behavior of the gated light yield shows that with temperature decrease a redistribution of the scintillation in the favour of slow components and phosphorescence occurs. Due to this reason, number of the detected photons in a long gate, for example 10 microseconds, is 20% less at -45°C than at room temperature. Situation is drastically improved in co-doped crystals. More than 95% of the scintillation is collected in 300 ns, a weak dependence of the light yield on the gate is observed. This indicates that electron traps have a minor contribution to the farther stage of the scintillation kinetics and phosphorescence as well.

4. Conclusions

We observed a significant improvement of the detecting properties of GAGG/PMT based scintillation detectors at their cooling. Improvement that is even more spectacular is expected at the application of SiPM for the scintillation photon detection. SiPM noise is reduced by factor two for each 10°C of temperature decrease, therefore combining of multidoped GAGG and SiPM readout opens an opportunity to create advantageous detectors, particularly for medical imaging applications.

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