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High-energy-resolution scintillator: Ce^{3+} activated $LaCl_3$

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The scintillation properties of $LaCl_3$ doped with 10% Ce^{3+} are presented. Under optical and gamma ray excitation, Ce^{3+} emission is observed to peak at 330 and 352 nm. The scintillation light output is 46 000 ± 1000 photons/MeV at 662 keV. Forty percent is emitted with a decay time of 26 ns, 30% with 210 ns, and 30% with about 1000 ns. An energy resolution (full width at half maximum over the peak position) of 3.3 ± 0.3% was observed for the 662 keV full absorption peak. © *2000 American Institute of Physics.* [S0003-6951(00)03936-X]

High detection efficiency, good energy resolution, and a fast timing resolution are highly desirable properties of x-ray and gamma ray radiation detectors. The inorganic scintillation crystal NaI:Tl, discovered by Hofstadter in 1948,[1,2] read by means of a photomultiplier combines all three properties to reasonable satisfaction. This and the fact that large crystals can be grown without problems at relatively low costs makes NaI:Tl still the most popular scintillator today.

Many more scintillators have been discovered that found application in detector systems. The high density of $Bi_3Ge_4O_{12}$ ($\rho = 7.13\ g/cm^3$) gives an excellent detection efficiency of high-energy gamma radiation but the time resolution (scintillation decay time, $\tau = 300$ ns) and especially energy resolution are worse than those of NaI:Tl are. Both the time resolution ($\tau = 40$ ns) and detection efficiency are very good for $Lu_2SiO_5{:}Ce^{3+}$ ($\rho = 7.4\ g/cm^3$), but the energy resolution is rather poor. Furthermore this material is quite expensive and technological problems in the growth of uniform crystals still have to be solved.

In the search for the ''ideal scintillator'' most efforts have been and still are directed towards oxide materials. Undoubtedly, oxides have the potential to provide very dense scintillators. However in order to obtain a high light output attention should be directed towards small band gap materials. We have reported already on $K_2LaCl_5{:}Ce^{3+}$ (Refs. 3 and 4) and $RbGd_2Br_7{:}Ce^{3+}$.[5,6] These compounds show a scintillation light output close to the theoretical maximum and an unequalled energy resolution down to 4% can be obtained. Unfortunately, K_2LaCl_5 presents a rather long decay component and both K_2LaCl_5 and $RbGd_2Br_7$ contain radioactive isotopes, i.e., ^{40}K and ^{87}Rb.

Here we report on the scintillation properties of $LaCl_3{:}Ce^{3+}$. Previously in the work by Guillot-Noël *et al.*[7] a sample doped with 0.57% Ce^{3+} was studied. At this concentration there appears to be a competition between slow host related self-trapped-exciton (STE) emission and fast Ce^{3+} emission. In order to favor the fast Ce emission we decided to further increase the Ce concentration and small single crystals with 2%, 4%, 10%, 30%, and 100% Ce^{3+} were studied.[8] Additionally, a relatively large crystal (∅8 mm × 5 mm) with 10% Ce^{3+} was grown, the excellent scintillation properties of which are presented in this letter. The more elaborated results on all crystals will be published elsewhere.

The $LaCl_3$:Ce 10% crystal was grown from $LaCl_3$ and $CeCl_3$ by the Bridgman technique. Starting materials were prepared from La_2O_3 (Heraeus, 99.999%), CeO_2 (Heraeus, 99.999%), NH_4Cl (Merck, reinst) and HCl (Merck, pro-analysis) by the ammonium halide method.[9,10] $LaCl_3$ crystallizes in the UCl_3 type structure, space group $P63/m$.[11] Based on structure and lattice parameters, $LaCl_3$ has a calculated density of 3.86 g/cm^3. With a Z_{eff} of 59.5, $LaCl_3$ is comparable to NaI with respect to high-energy radiation detection efficiency.

Figure 1 shows the pulse height spectra of radiation from a ^{55}Fe source, a ^{241}Am source, and a ^{137}Cs source, respectively, obtained with a Hamamatsu R1791 photomultiplier tube (PMT). An elaborate description of the experimental setups for light yield and scintillation decay measurements can be found in the paper by Guillot-Noël.[7] The absolute light yield for $LaCl_3$:Ce 10% is 46 000 ± 1000 photons per MeV (ph/MeV) of absorbed γ ray energy at 662 and 60 keV. A lower absolute light yield was obtained employing a ^{55}Fe source: 39 000 ± 1000 ph/MeV at 6 keV. The energy resolu-

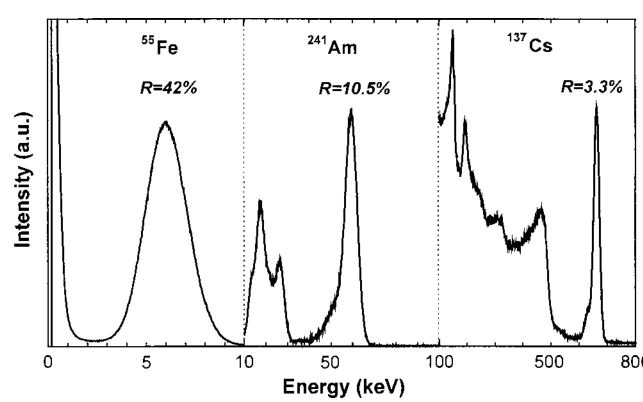

FIG. 1. Energy resolutions obtained with a single crystal of $LaCl_3$:Ce 10% for x/γ rays from (a) ^{55}Fe, (b) ^{241}Am, and (c) ^{137}Cs sources.

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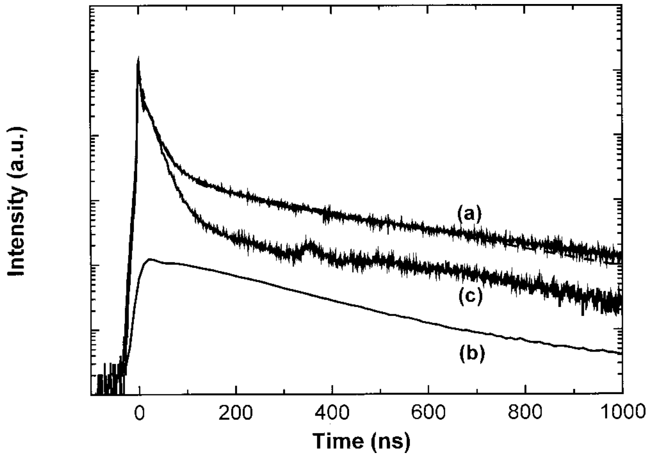

FIG. 2. Decay curves of (a) $LaCl_3$:Ce 10%, (b) NaI:Tl, and (c) $LaCl_3$:Ce 30%. The dashed curve represents the fit of decay curve (a).

tion R [full width at half maximum (FWHM) over the peak position] of the 662 keV full absorption peak is $3.3\pm0.3\%$. This energy resolution is the best ever reported in the literature with scintillation detectors. Features like the characteristic La K x-ray escape peak at 629 keV and a weak edge around 550 keV due to double Compton scattering events may be observed as a result of this excellent energy resolution.

With a shaping time of 10 μs, on average, 7800 photoelectron are collected at the first dynode of the PMT after absorbing a 662 keV γ quantum. For γ rays from a ^{241}Am source, 10 times less photoelectrons are collected. Then, according to Poisson statistics,[12] the energy resolution R would be 10.4%. We experimentally determined an energy resolution of $R=10.5\pm0.9\%$, which is in good agreement with expectations. In this line of reasoning, it was anticipated that the energy resolution R of the 6 keV full absorption peak is in the order of 30%–35%. However, an energy resolution of $42\pm1\%$ was found.

The scintillation decay curve of $LaCl_3$:Ce 10% is depicted in Fig. 2, curve (a). For comparison, the scintillation decay curve of NaI:Tl is shown as well [Fig. 2, curve (b).] In order to analyze these curves, a simple exponential decay function with three decay components was used.

$LaCl_3$:Ce 10% has a short decay component of 26 ± 1 ns which represents 40% of the total light yield. A relatively long decay component of $\sim$210 ns accounts for 30%. If the Ce concentration is increased to 30%, the total light yield remains the same ($46\,000\pm1000$ ph/MeV) but the short decay component now represents 61% of the total light yield; see decay curve (c) in Fig. 2. In the $LaCl_3$ sample doped with 0.57% Ce^{3+} part of the decay curve of $LaCl_3$:Ce^{3+} luminescence could be explained by assuming energy transfer by diffusion of STE to Ce^{3+}. An increase in Ce^{3+} concentration would increase the transfer efficiency of the excitation energy from STE to Ce^{3+} and enhance Ce^{3+} luminescence.[7] Indeed, for $LaCl_3$:Ce^{3+}10% and even more for $LaCl_3$:Ce^{3+}30%, Ce^{3+} luminescence is favored over STE luminescence. The contribution of the short decay component to the total light yield is significantly larger in these samples than in $LaCl_3$:Ce 0.57%.

If we compare the scintillator properties of $LaCl_3$:Ce with those of NaI:Tl it is obvious that $LaCl_3$:Ce shows a much better energy resolution and, depending on the Ce concentration, a much faster response. Although hygroscopic, $LaCl_3$:Ce is easy to grow. For applications where a good energy resolution is required, $LaCl_3$:Ce seems to be a very promising scintillator and competitive with NaI:Tl.

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