Temperature dependence of scintillator nonproportionality

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Using highly monochromatic synchrotron X-rays in the energy range from 10.5 keV to 100 keV the temperature dependence of nonproportionality and energy resolution of $LaBr_3$ scintillators doped with 5% Ce^{3+} and SrI_2 doped with 5% Eu^{2+} were studied at 80K, 295K, 450K, and 600K. For the first time improvement of the proportionality and better energy resolution was observed on changing the temperature. This discovery suggests that the already outstanding energy resolutions of $LaBr_3$:Ce and SrI_2:Eu can be improved even further. It also may provide new clues to better understand the processes that cause nonproportionality of inorganic scintillator response.

Attempts to measure the photon nonproportional response (photon-nPR) as a function of temperature were made before by changing the temperature of both the scintillator and PMT over a relatively narrow temperature range from -30ºC to +60ºC. In our experiments the PMT remains at room temperature while the scintillator temperature can be changed. Thus, we do not need to consider the effect of temperature on the characteristics of the PMT, and we measure the intrinsic properties of the scintillator only.

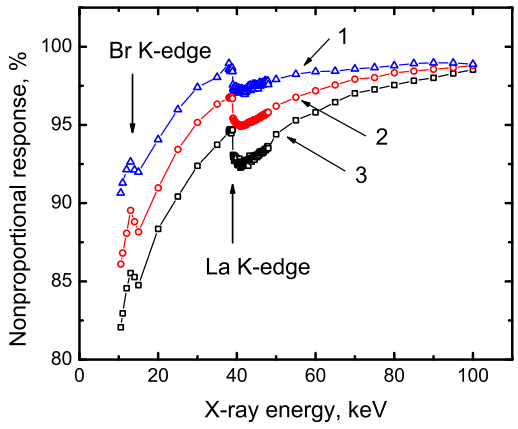

Fig. 1: Nonproportional response of $LaBr_3$:Ce as a function of x-ray energy (E_X) at curve 1) 80K, 2) 295K, and 3) 450K.

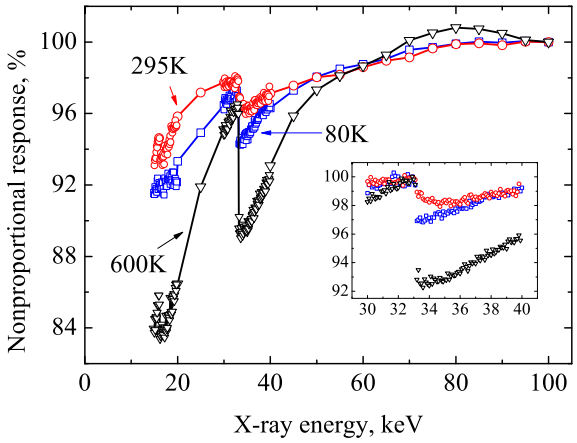

Fig. 2: Nonproportional response of SrI_2:Eu as a function of x-ray energy (E_X) at 1) 80K, 2) 295K, and 3) 600K.

To measure the pulse height spectra at many finely spaced energy values between 10.5 keV and 100 keV, experiments were carried out at the X-1 beam line at HASYLAB. A highly monochromatic pencil x-ray beam in the energy range 10.5 – 100 keV was used as an excitation source. To record scintillation pulse height spectra as a function of temperature, a sample was fixed at the bottom of a parabolic-like stainless steel cup covered with reflective Al foil, mounted onto the cold finger of a liquid nitrogen bath cryostat. The cup directed the scintillation light through the quartz window towards a PMT situated outside the cryostat chamber. The Hamamatsu R6231-100 PMT at -680V remained at room temperature and observed about 20% of the emitted scintillation light. To collect as much of the PMT charge pulse as possible, the shaping time of an Ortec672 spectroscopic amplifier was set at 10 μs. The temperature of the crystal was controlled by two thermocouples attached to different parts of the sample holder.

We define the photon-nPR of a scintillator at x-ray energy (E_X) as the light output per MeV observed at energy E_X divided by the light output per MeV observed at E_X = 662 keV. Figure 1 shows the photon-nPR of $LaBr_3$:Ce as a function of E_X at 80K, 295K and 450K, and Figure 2 shows the photon-nPR of SrI_2:Eu as a function of E_X at 80K, 295K and 600K. The shape of the response curves has been discussed by us in detail [1]. Note that for $LaBr_3$:Ce in Fig.1 we observe discontinuities in the photon-nPR curves not only at the lanthanum K-electron binding energy E_{KLa} = 38.925 keV, but also at the bromine K-electron binding energy E_{KBr} = 13.474 keV. For SrI_2:Eu, we observed discontinuities' at E_{KSr}=16.105 keV and at E_{KI}=33.169 keV. The size of the drop in *nPR* at the K-edge is called the K-dip magnitude.

The most important observation is that the photon-nPR reveals strong temperature dependence. For $LaBr_3$:Ce, at 100 keV the values of nPR at all three temperatures are almost the same 98.8%. The situation is quite different at low energy. At 10.5 keV the nPR at 80 K is 90.7%, at 295 K it falls to 86.1% and at 450 K it has decreased to 82.1%. Therefore, a clear improvement in the photon-nPR occurs as the temperature decreases from 450K to 80K. Since the typical error in photon-nPR does not exceed 0.1% at 10.5 keV these are significant improvements. This result indicates that electron-hole recombination losses in the high ionization density part of the ionization track in $LaBr_3$:Ce have a strong temperature dependence. In contrast to $LaBr_3$:Ce, the *nPR* in SrI_2:Eu first improves on heating from 80K to 295K and then worsens with further increase in temperature to 450K and 600K. The inset in Fig. 2 shows the K-dip on an enlarged scale, normalized to 100% at E_{KI}. As in $LaBr_3$:Ce, in SrI_2:Eu the K-dip magnitude decreases with improved proportionality.

We have found that by reducing the temperature of $LaBr_3:Ce^{3+}$ scintillator from 450 K to 295 K to 80 K its proportionality improves and as a consequence a better energy resolution for X-ray photon detection is obtained [2]. SrI_2:Eu proportionality and energy resolution first improves on heating from 80 K to 295 K and then worsens on further heating to 450 K and 600 K [3]. These behaviors are evidence that the yet unknown parameters responsible for the nonproportional response of $LaBr_3$:Ce and SrI_2:Eu depend on temperature. Apparently, with decreasing temperature, the amount of electron hole recombination losses in the dense parts of the ionization track decreases and as a consequence the efficiency of the scintillator increases. Recent simulation data suggest that electron and hole mobilities are important parameters that determine nonproportionality, and our finding is a manifestation of changing mobilities with changing temperature. It is known that electron mobility is strongly temperature dependant due to phonon scattering. Although, the cause of proportionality improvement is not yet established in this work, our findings for the first time do demonstrate that nonproportionality is a property that can be improved. If it can be improved by changing temperature one may hope to improve it also by other means like activator concentration, crystal quality, co-dopants etc.

This work was funded by the Netherlands Technology Foundation (STW), and supported by Saint-Gobain Crystals & Detectors, France and by the European Community's Seventh Framework Programme (FP7/2007-2013) under grant agreement n° 226716. We thank the scientists and technicians of the X-1 beamline at the Hamburger Synhrotronstrahlungslabor (HASY-LAB) synchrotron radiation facilities for their assistance.

References

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