Nonproportional response of inorganic scintillators to synchrotron X-ray irradiation

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The widespread use of inorganic scintillators for applications in science and society is the driving force behind the search for new high performance compounds. The most important requirement imposed on new scintillators is a high energy resolution for gamma ray detection. There are two key factors that determine the energy resolution, i.e., Poisson statistics in the number of detected photons and the nonproportionality of the light yield of scintillators with gamma-ray energy. Nonproportionality means that the total light output of a scintillator is not precisely proportional to the energy of the absorbed gamma-ray photon. This has a deteriorating effect on energy resolution. For example, based on Poisson statistics alone $LaBr_3$:Ce, should display an energy resolution of 2.1% when 662 keV gamma-ray photons are detected with a standard type PMT. However in reality it is about 2.8%. Because the light yield and the PMT performance is already close to optimal we need to reduce nonproportionality in order to improve the energy resolution.

Nonproportionality is due to electron-hole recombination losses during the scintillation process. It is currently believed that those losses occur inside parts of the ionization track where the ionization density is high. That density increases when the gamma-ray energy decreases. The scintillation yield per energy unit in $LaBr_3$:Ce scintillator at 10 keV energy is for example 15% smaller than at 662 keV (Fig.1) [1]. The origin of this decrease in efficiency, i.e., the true cause of electron-hole recombination losses, and the related deterioration in energy resolution is not known. It is a mystery to the scintillation community why some scintillators reveal poor proportionality while others appear reasonably good [2]. To elucidate the true origin of nonproportionality, we have continued our study of nonproportionality of inorganic scintillators. The nonproportional photon response of more than 20 different scintillators was measured using highly monochromatic synchrotron irradiation.

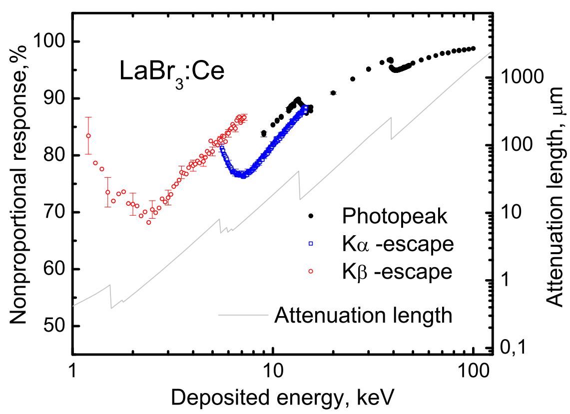

Figure 1: Photon nonproportional response of $LaBr_3$:Ce as a function of deposited energy. Black solid circles, photopeak-nPR; blue open squares, K_α escape-nPR; red open circles, K_β escape-nPR .The solid curve shows the calculated X-ray attenuation length for $LaBr_3$.

To estimate the photon response, pulse height spectra at many finely spaced energy values between 9 keV and 100 keV were measured. The experiment was carried out at the X-1 beamline at the Hamburger Synhrotronstrahlungslabor (HASYLAB) synchrotron radiation facility in Hamburg, Germany. Special attention was paid to the X-ray fluorescence escape peaks [1-3] as they provide us with additional information about photon response in the range 1.0 – 10.0 keV. A rapid variation of the photon response curve is observed near the K- electron binding energy for all scintillators. A dense sampling of data is performed around this energy and this is used to apply a method, which we call K-dip spectroscopy [2]. This method allows us to derive the electron response curves down to energies as low as 0.1 keV.

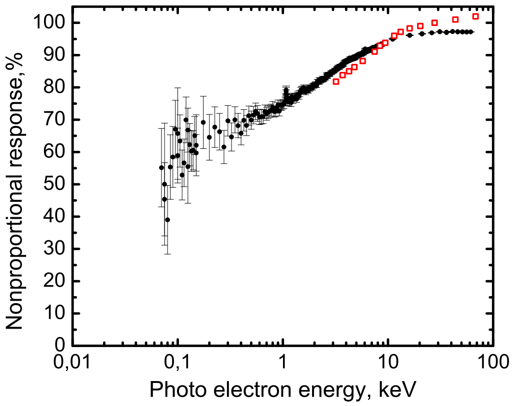

Figure 2: Black solid circles, electron nonproportional response of $LaBr_3$:Ce as a function of K-photoelectron energy obtained from K-dip spectroscopy. Red open squares, electron-nPR obtained with Compton Coincidence Technique.

In principle, scintillation light yield nonproportionality can be characterized as a function of either photon or electron energy. The scintillation response as a function of X-ray and gamma photon energy, is in general easy to measure and is an indication of scintillator quality (Fig.1). However, the scintillation nonproportional response as function of electron energy, is more fundamental (Fig.2). For a better understanding of the true cause of nPR, measurements of both the photon and the electron response of the scintillator in question are needed. The most dramatic changes in the nPR occur in the 0.1 keV-10 keV energy range, where the ionization density along the track is higher than at energies of say 100 keV to 1 MeV. To study the nonproportional response in the 0.1 keV – 10 keV range we applied escape peak analysis and K-dip spectroscopy [1-3].

The K-dip spectroscopy method can be briefly described as follows. An X-ray that photoelectrically interacts with the lanthanum K-shell leads to the creation of a K-shell photoelectron plus several Auger electrons. The response of a scintillator is then equivalent to the sum of two main interaction products: 1) the K-shell photo electron response plus 2) the response from the electrons emitted due to the sequence of processes following relaxation of the hole in the K-shell, the so-called K-cascade response. Our strategy is to employ X-ray energies just above E_K. The K-cascade response is assumed independent from the original X-ray energy. This response is found by tuning the X-ray energy to just above E_K. By subtracting the K-cascade response from the total X-ray response we are left with the response in photoelectrons from the K-shell photoelectron alone with energy E_X - E_K. The K-electron-nPR curve is then obtained from the number of photoelectrons/MeV at the energy of the K-photoelectron divided by the number photoelectrons/MeV measured at 662 keV.

References

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