

Scintillation Mechanisms in Cerium Fluoride

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This paper investigates the mechanisms responsible for the scintillation of cerium fluoride (CeF_3) and cerium–doped lanthanum fluoride ($\text{LaF}_3:\text{Ce}$), a recently discovered family of scintillators of interest for gamma ray detection in both medical imaging and high energy physics [1, 2, 3, 4]. The observed scintillation spectra and decay kinetics of these materials are not simple and depend upon the particular crystals and experimental conditions used. Interest in understanding and improving the scintillation properties of CeF_3 is currently quite high, for at least one high energy physics collaboration is proposing to build an electromagnetic calorimeter containing as much as 60 cubic meters of CeF_3 [5, 6]. The motivation for this study is a better understanding of the fluorescence mechanism with the objective of improving the scintillation light yield and decay time.

A variety of experimental measurements were made to investigate the scintillation in samples of cerium fluoride and of lanthanum fluoride doped with cerium in concentrations between 0.01% and 50% mole fraction cerium. Ultraviolet photoelectron spectroscopy was used to determine the energies (with 0.2 eV resolution) of the populated electron energy bands lying between 0 and 30 eV binding energy, including the Ce^{3+} 4f band. Optical transmission was used to determine the positions of the unpopulated bands that this optically active Ce^{3+} 4f electron could be promoted to, namely the five Stark split Ce^{3+} 5d levels. Fluorescence excitation spectroscopy (with excitation energies between 4 and 27 eV) was used to associate observed emission lines with features noted in the UPS spectra (and hence specific transitions), and times resolved spectroscopy used to observe the decay kinetics of these emission lines.

In $\text{LaF}_3:\text{Ce}$ the absorption of either optical or ionizing radiation directly or indirectly results in excitation of the Ce^{3+} 4f electron to the lowest 5d level followed by $5d \rightarrow 4f$ fluorescence at 284–300 nm. Whereas for optical excitation the fluorescence has a 20 ns decay time, for ionizing radiation there is an additional faster (2–10 ns) initial decay component. As the cerium concentration increases, another band appears that partially absorbs the 284–300 nm emission and re–radiates it in a broad band peaking at 340 nm and having a longer (~30 ns) decay time. In the limit of 100% CeF_3 , this radiation trapping is very pronounced. The additional absorption and emission bands present at large Ce concentrations are attributed to cerium ions that have been perturbed in some way, either by impurities, crystal defects, or interactions with other cerium ions.

Because the 340 nm light is emitted with a slower decay time than the 290 nm light, this absorption and re–emission increases the effective decay time, which degrades the performance of CeF_3 as a scintillator. The data imply that the scintillation properties of CeF_3 can be improved if perturbed sites can be eliminated. This may be possible if the perturbation is caused by an impurity or introduced in the manufacturing process, but is impossible if it is due to Ce–Ce interactions.

References:

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