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Nonlinear optical spectroscopy for studying carrier transport and recombination in scintillators for fast radiation detectors

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The current demand for materials with fast scintillation response for radiation detectors in both major application fields, future high-luminosity high energy physics experiments and medical imaging with better spatial resolution, require the characterization of processes in scintillators in picosecond domain. In particular, excitation transfer becomes important for fast scintillation response.
In this work, we adopted nonlinear optical spectroscopy to study the processes limiting the rate of excitation transfer in activated scintillators, especially in those with multicomponent crystalline matrix where the transfer might be affected by carrier trapping due to the potential fluctuations caused by compositional disorder. Transient optical absorption technique in pump and probe configuration was exploited to monitor the time evolution of nonequilibrium carrier density. Selective excitation using tuneable-wavelength pulses and measuring a wide spectrum of transient absorption (TA) at variable delay after short pulse excitation enabled the identification of the type of nonequilibrium carriers responsible for TA and revealing the peculiarities of excitation transfer. Time-resolved photoluminescence was also exploited.
Our study was focused on two families of prospective Ce-doped scintillators: lutetium yttrium oxyorthosilicates (LYSO:Ce) and gadolinium aluminium gallium garnets (GAGG:Ce). The comparison of the TA response rise time with the coincidence time resolution, which is a conventional parameter measured under gamma excitation, showed that the delay in the front of the TA response due to the population of the lowest (emitting) excited level of Ce3+ after excitation of the ion to higher excited levels reflects the electron transfer that is affected by electron trapping. The influence of aliovalent codoping on the luminescence response time was studied in LYSO:Ce,Ca and GAGG:Ce,Mg with different codoping level. It is shown that the codoping results in faster luminescence response due to elimination of trapping centers and enhances the luminescence decay rate due to introduction of quenching centers. A trade-off between luminescence efficiency and decay time is considered.

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