

Free carrier absorption for study of fast excitation transfer in scintillation crystals

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New requirements for timing of scintillator-based radiation detectors for future high energy physics experiments and medical imaging applications necessitate demand for novel tools to study the non-equilibrium carrier dynamics in scintillators on picosecond time scale. In this report, we summarize our current results on the application of free carrier absorption to study the carrier dynamics in several scintillators prospective for fast ionizing radiation detectors.

Our experiments have been carried out in pump and probe configuration. The non-equilibrium carriers were photoexcited by 200-fs-long laser pulses in visible or UV. The capability of photoexcitation to change the energy of excitation photons by using harmonics of the main laser radiation or gradually tune it by application of parametric oscillators was exploited for the resonant excitation of activator ions or structural units in the host crystal. The change in optical absorbance induced by the excitation was probed in the infrared region. The change is caused by free carrier absorption. Its absolute value reflects the density of non-equilibrium carriers, while the spectral features might be exploited to characterize the density of states in the bands. Both intrinsic (PWO) and Ce-doped scintillators (GAGG:Ce, YAGG:Ce) have been studied. The technique has been exploited to compare the excitation transfer in GAGG:Ce with and without codoping with divalent magnesium.

In self-activated lead tungstate (PbWO₄, PWO), the nonlinear optical response due to free carrier absorption occurs immediately after the short-pulse excitation. The possibility to use this fast response to provide precise timing capabilities in novel radiation detectors is under discussion.

In GAGG:Ce, the signal is dominated by the absorption of free holes. The response peak occurs with a delay of few picoseconds due to hole delocalization from Gd³⁺ ground state to the valence band. The response in the garnet scintillator YAGG:Ce containing no Gd occurs immediately after the short-pulse excitation. The comparison of the results on free carrier absorption and those obtained by time-resolved photoluminescence spectroscopy enabled us to compile a electronic energy-level diagram of the main structural units of GAGG:Ce and GAGG:Ce,Mg to explain the influence of codoping with divalent Mg on the time characteristics and light yield of this scintillator.

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