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Picosecond absorption spectroscopy of self-trapped holes, self-trapped excitons, and transient Ce states in LaBr_3 and LaBr_3:Ce

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We report excitation-induced picosecond absorption over an extended spectral range from 320 nm to 2700 nm in $LaBr_3$, $LaBr_3$:Ce(4%) and $LaBr_3$:Ce(20%). Preliminary identification of a V_k band near 3.6 eV, self-trapped exciton (STE) hole transition near the same energy, and STE bound-electron transitions spanning 0.46 eV to 1.1 eV have been made. Comparison to recent calculations of V_k and STE structure in $LaBr_3$ by A. Canning and M. Del Ben [private communication] is helpful and in rough agreement on several points. We also observe transient induced absorption bands whose strength increases with Ce concentration, and they are tentatively attributed to carrier capture and/or excited states involving Ce. Strong Ce-correlated transient absorption bands are found at 2.8 eV, 2.2 eV, and with weaker Ce correlation at about 1.25 eV. We are working to establish identification with expected charge-transfer (CT) electron and hole transitions of the Ce^{3+*} excited activator and CT transitions of Ce^{4+} activator-trapped holes. Together with the identified STH and STE transitions noted above, these should constitute the main species in $LaBr_3$:Ce scintillation. The excitation in these experiments is two-photon absorption of 300 fs pulses producing total transition energies of 5.9 eV, only slightly above the band gap of $LaBr_3$, and 8.86 eV, capable of creating hot electrons with almost 3 eV excess energy. Use of the corresponding two pump photon energies at 2.95 eV and 4.43 eV allows distinguishing effects of direct absorption of the pump photons by Ce dopant. Assembling information on the picosecond-scale sequential populations of trapped carriers and excited states that are main participants in scintillation, along with quantitative rates of capture, is necessary for a material engineering model of $LaBr_3$:Ce. The present work follows our similar picosecond-measurement program in CsI, CsI:Tl [1] which supplied a number of key rate coefficients for successful modeling of pulse shape, proportionality of decay components, and light yield in CsI:Tl [2].

References

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