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Two-photon photoconductivity and luminescence in scintillators –measurement and mapping

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The radius of initial (femtosecond-scale) energy deposition around the local trajectory of a high-energy electron in a scintillator has been characterized as roughly 3 nm by several researchers, e.g. [1]. The resulting strong radial gradients of carrier density drive diffusion which can both dilute and separate carrier populations. This significantly influences the rate terms for capture, bimolecular recombination, and nonlinear quenching that govern the physical mechanisms of scintillation [2]. Particularly in halide scintillators, the holes self-trap rapidly and are initially confined near the track core, while electrons in the heavier halides persist as hot electrons for up to about 4 ps, during which time they outrun the self-trapped holes (STH) and thermalize or trap at a much larger radius than the STH. The subsequent evolution of the scintillation pulse starts from charge-separated carrier populations in a strong internal radial electric field. Thus carrier mobilities and diffusion coefficients are important characteristics of scintillator materials and essential input data for material engineering models. Inorganic scintillators most commonly are insulating crystals, and halide scintillators are often hygroscopic as well, so transport measurements can be challenging in this material class. Such data have been measured in some common alkali halides and a few oxide scintillator materials, but have yet to be measured in most of the recently developed high-performance scintillators, including rare-earth halides, alkaline earth halides, elpasolites, and materials with activator concentrations that can run to 10% or more.

This report describes experiments in which both photoconductivity and photoluminescence are measured under two-photon interband excitation of scintillator materials. Two-photon excitation enables generation of electron-hole pairs below the surface in the scintillator host. Use of sub-picosecond laser pulses allows resolving the very short carrier re-trapping times, τ, expected in doped wide-gap scintillators in order to extract the mobility, μ, from the measured $\mu\tau$ product. Using the established principles of two-photon confocal microscopy, our experiment provides depth resolution and lateral resolution determined by the microscope objective. Therefore high-resolution 3-d mapping of luminescence response and somewhat lower resolution of photoconductivity is achieved.

We have previously reported simultaneous mapping of photoconductivity and two-photon luminescence with this system in the semiconducting scintillators $LiInSe_2$ and ZnSe:Te [3]. The present report describes results on CsI and CsI:Tl as a test of our experimental method applied to a well-characterized alkali halide scintillator preparatory to applying the technique in newer scintillator materials, including their modifications and effects of heavy doping. In addition to measurements of two-photon photoconductivity in CsI:Tl, the mapping of Tl^* luminescence with 2 μm resolution in our experiment has indicated a granularity of response with feature size in the 20 μm range in standard CsI:Tl scintillators. This could indicate clustering or concentration of Tl activator ions due to aggregation in the bulk or collection along low-angle grain boundaries. We are also using similar methods to investigate possible origins of double photopeaks in defective CsI:Tl scintillators.

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[1] F. Gao et al., "Monte Carlo simulation of gamma-ray response of BaF2 and CaF2,"J. Appl. Phys., vol 114, 2013.

[2] X. Lu et al., "Energy-Dependent Scintillation Pulse Shape and Proportionality of Decay Components for CsI:Tl: Modeling with Transport and Rate Equations,"Phys. Rev. Appl., vol 14007, 2017.

[3] K. B. Ucer et al., "Dual Detection Charge Collection and Light Emission in $LiInSe_2$ and ZnSe,"Oral Presentation R08-6, IEEE: RTSD Conference, 2016.

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