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Abstract title: Tailoring the ultrafast coincidence time resolution in novel highly luminous metascintillators

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Abstract

Medical imaging ToF-PET technique based on bulk crystalline scintillators has reached the maximum possible performance. However, their coincidence time resolution (CTR) of 200 ps do not allow the high contrast - high spatial resolution quality in diagnostic images. Overcoming the stagnant performance of ToF-PET medical diagnostic detectors requires a new concept of fast and highly emissive metascintillators. [1]. They consist of a dense but slow emitting material guaranteeing efficient interaction with ionizing radiations, coupled to a light but fast scintillator. Its fast emission is activated by an ultrafast energy sharing mechanism from the dense material, thus accelerating the global scintillation kinetics and therefore improving the CTR of the system. [2] This work presents two metascintillators prototypes with dimensions in the centimetre scale lengths, similar to those of ToF-PET scanners pixel and with made-to-order properties to reach both high luminescence characteristics in terms of light yield (LY), light output (LO) and timing performances towards the CTR goal. In the first prototype, the heterostructure is made by layers of standard BGO and HfO₂ - nanocomposites. [3] Importantly, this nanocomposite scintillator was engineered ad hoc in terms of elements' composition and electronic properties to maximize the stopping power and energy deposition of g-ray, involving its conversion to emissive molecular excitons, for boosting the LY up to 60 000 ph/MeV while preserving fast timing, according to [4]. In the resulting metascintillator and upon interaction with 511 keV g-rays, the energy sharing among the layers triggers the nanocomposite's fast emission allowing an excellent ultrafast CTR of 120 ps. A second smart approach to build metascintillators and further approach the goal of CTR <100 ps is presented. We used MOF films as fast scintillating part grown on dense scintillating dense crystals to optimize the energy sharing and the optical coupling between the layers. MOF were synthesized by using one (homo-ligand MOF) or two ligands (hetero-ligand MOF). [5,6]. Thus, the extreme versatility of MOF enabled to maximize the LO of the metascintillator by matching the best efficiency spectral window of photodetectors and obtaining a largest Stokes shift, thus avoiding reabsorption effects. Importantly, the architecture of MOF allowed to tune the ultrafast donor-acceptor energy transfer processes within the crystalline framework, achieving sub-nanosecond ultrafast scintillation pulses with LY that resembles and also exceeds the ones of best commercial fast emitting plastic scintillators [1] Lecoq, IEEE Trans Radiat Plasma Med Sci. 1, 473–485, 2017 [2] Orfano, Adv. Mater. Technol. 2302075, 2024 [3] Sala, Advanced Functional Materials, 2421434, 2024 [4] Villa, Nano Lett. 24, 8248, 2024 [5] Perego, Nat Photonics, 15 (5), 2021 [6] Orfano, Adv Funct Mater, 34 (44), 2024