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Purcell Enhanced Perovskite Nanocrystals for Ionizing Radiation Detection

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Abstract - Scintillators convert ionizing radiation into visible light and are critical in medical diagnostics and radiation detection. Enhancing their efficiency through nanophotonic strategies - particularly by exploiting the Purcell effect - has gained interest but has typically been constrained to nanoscale or ultrathin geometries due to the highly localized nature of plasmonic field enhancements. In this work, we present an approach to extend the influence of nanoplasmonic enhancement to bulk scintillating materials. By embedding 100 nm plasmonic nanoparticles (spheroidal and cuboidal in shape) into a polymer matrix containing $CsPbBr_3$ perovskite nanocrystals, we achieve significant improvements in scintillation performance within a 5-mm-thick composite. Photoluminescence measurements reveal enhancements in emitted power and decay rates by factors of up to (3.20 ± 0.20) and (4.20 ± 0.31), respectively, depending on nanoparticle geometry. These results are supported by theoretical modeling, which predicts enhancement factors of up to (2.26 ± 0.31) and (3.02 ± 0.69). Additionally, scintillation yield under ^{241}Am γ-ray excitation exhibits a measurable increase of (2.07 ± 0.39). This method is not emitter-specific; it remains effective as long as the plasmonic nanoparticle scattering spectrum aligns with the emitter luminescence. Our approach addresses a critical limitation of prior nanoplasmonic scintillators by enabling enhancement in millimeter-scale materials. This advancement offers a generalizable pathway for integrating Purcell-enhanced nanocrystal scintillators into large-volume detectors, supporting emerging demands in medical imaging and high-energy radiation monitoring.

Keywords**:** $CsPbBr_3$, nanocrystals, scintillation, Purcell effect

1. Introduction

Scintillator performance is not solely determined by inherent material properties. By altering the local density of optical states (LDOS) [1-5], nanophotonic and nanoplasmonic methods [1,2,6,7], such as the Purcell effect [8], are being investigated to improve emission characteristics. The Purcell effect can greatly increase radiative decay rates by optimizing the LDOS, which could lead to an increase in scintillation efficiency. Since this impact is strongly localized, thin-layer materials were the focus of earlier research [2-4]. The main question remains, how these improvements might be scaled-up to bulk scintillators - whose thickness can reach millimetres on scale. By embedding $CsPbBr_3$ [9] perovskite nanocrystals (NCs) with silver (Ag) nanospheres (SNPs; 100 nm diameter) and nanocubes (CNPs; 100 nm side length) in a polydimethylsiloxane (PDMS) matrix, we overcome this challenge. Here we show that it is possible to achieve nanoplasmonic Purcell enhancement in bulk, stable, bright, and scalable scintillating material that is appropriate for security and medical imaging applications. Silver is chosen since it can offer the greatest improvements and the least losses for our NCs' green emission. This hybrid system approach increases the light yield and decay rate of $CsPbBr_3$ NCs without being constrained by thickness. This is the first practical implementation of Purcell enhancement in materials thicker than a few μm.

In this work, the Purcell effect is assessed by analysing changes in both luminescence intensity (radiated power, P) and photoluminescence decay dynamics (quantified by the inverse of the decay time, Γ) of $CsPbBr_3$ nanocrystals. These NCs are incorporated into silver nanoparticle–PDMS (Ag NP–PDMS) composite structures and compared to reference samples embedded solely in a PDMS matrix, denoted with a superscript "0". The enhancement is characterized by the Purcell factor (F_p), which is defined as [12]:

$$F_p = \frac{P}{P_0} = \frac{\Gamma}{\Gamma_0} \tag{1}$$

where, both the power and decay rate ratios serve as equivalent representations of F_p, depending on the quantum efficiency (QE) of the NCs [13].

2. Results

A schematic overview of the experimental approach is presented in Fig. 1. High quality $CsPbBr_3$ NCs were prepared by a modified hot-injection method [14] and embedded into PDMS matrix. Such system served as a reference for Ag co-doped composites. PVP shelled Ag NPs were purchased from Nanocomposix. The PVP shell acts as a surfactant, preventing direct contact between NCs and NPs.

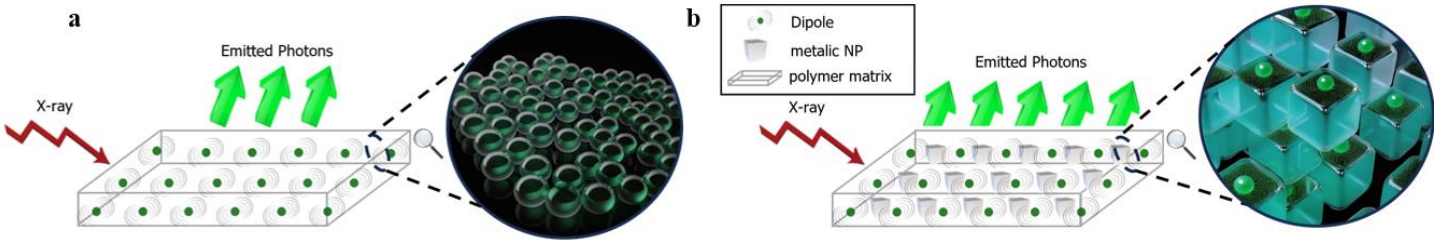

Fig. 1. Schematic descriptions of scintillator nanocrystals (NCs) (**a**) and NCs co-doped with metallic nanoparticles (**b**) embedded into the polymer matrix.

Physical, optical and scintillation properties of prepared samples were examined by electron microscopy, photo- and X-ray luminescence as well as their time-resolved counterparts. Results of such examination are presented in Fig. 2.

To understand the physical origin of the observed enhancements, we employ FDTD simulations using a classical electromagnetic model for a dipole emitter placed near metallic nanoparticles. Simulations account for rounded-edge geometries and PVP shells, with emitter-NP separation set to 9 nm. The local density of optical states (LDOS) increase is most prominent near corners of CNPs, explaining their superior performance. Monte Carlo simulations of 100,000 emitter-NP configurations further confirm that CNPs consistently achieve higher LDOS enhancements compared to SNPs, reaching (7.85 ± 1.80) and (5.87 ± 0.01) for CNPs and SNPs, respectively. EDS evaluation of NCs-NP coupling percentage

(employing Sorensen-Dice similarity method [15]) and QE measurements reduced the expected FDTD LDOS enhancement values to (3.02 ± 0.69) and (2.26 ± 0.31) for CNPs and SNPs, respectively. Interestingly the analytical approach, yields a perfect match with FDTD simulations, reaching (3.03 ± 0.23) and (2.43 ± 0.64) for CNPs and SNPs, respectively. The enhanced performance of Ag CNPs compared to their spheroidal counterparts is primarily due to their angular morphology, which supports the formation of localized plasmonic hotspots with higher field intensities. These localized enhancements significantly increase the LDOS surrounding the emitter. As a result, the radiative decay pathways of $CsPbBr_3$ nanocrystals are more effectively promoted, leading to a pronounced boost in scintillation efficiency within the bulk composite material.

Both experimental data and FDTD simulations consistently confirm Purcell-enhanced luminescence and scintillation performance of $CsPbBr_3$ nanocrystals. As presented in Fig. 2**c** our results are in a good agreement with theory with CNPs reaching the highest enhancement values.

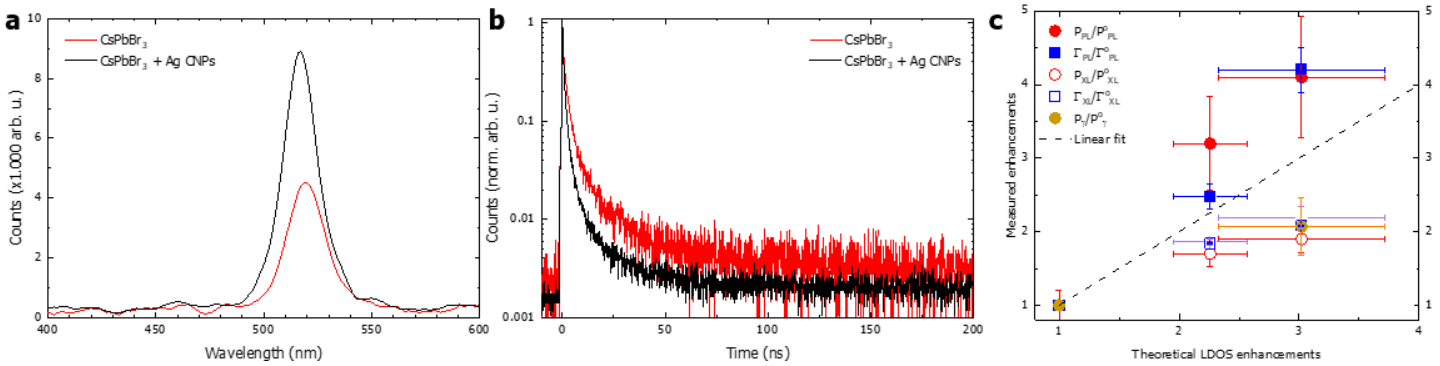

Fig.2. The X-ray luminescence spectra (**a**) and time-resolved XL decay curves (**b**). (**c**) The measured vs theoretical LDOS enhancements, derived from FDTD calculations with correction factors. All measurements were performed at room temperature.

4. Conclusion

In this work, we demonstrate the first practical implementation of the Purcell effect in a bulk material for enhanced scintillation efficiency. Our approach provides an alternative strategy to overcome the limitations of thin-film geometries and achieves stable performance improvements. Furthermore, our millimetre-scale solution, which relies on a scalable self-assembly process, addresses the growing demand for large-area, high-performance nanophotonic scintillators in advanced medical imaging and security applications. We present enhancements of up to (4.10 ± 0.20), (1.92 ± 0.13), and (2.07 ± 0.39) for Ag CNP-doped samples under optical, X-ray, and γ-ray excitation, respectively. Time-resolved measurements revealed enhancements of emission rates up to (4.20 ± 0.31) and (2.08 ± 0.06) in TRPL and TRXL. This study introduces a scalable and broadly applicable strategy for extending Purcell enhancement into thicker nanocrystal scintillator volumes. Using $CsPbBr_3$ nanocrystals embedded in a PDMS matrix as a model system, we demonstrate that the approach is not restricted to a specific emitter, provided the scattering spectrum of the plasmonic nanoparticles is spectrally matched with the emitter's photoluminescence. This generality is particularly relevant for practical applications, where scintillator layers must meet specific thickness requirements to ensure efficient detection [16].

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