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Letter

Sub-bandgap photon harvesting for organic solar cells via integrating up-conversion nanophosphors

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ABSTRACT

Lanthanide-doped up-conversion nanophosphors were employed as up-conversion converters (UC-converters) combined with organic solar cells (OSCs). The optical properties of the external up-conversion layers were investigated, and the optimized layers were laminated with OSCs. Strong photocurrents were observed when OSCs with an UC converter were illuminated by a NIR laser. An improvement of photocurrent and efficiency were also observed under AM 1.5 G sun irradiation. Our investigations show a proof-of-concept that organic solar cells could utilize sub-bandgap photons via integrating up-conversion nanophosphors, which provide us a promising approach to exceed the efficiency limit of single junction organic solar cells.

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Organic solar cells (OSCs) have been extensively investigated due to their promising properties such as solution processability, low cost and light weight over the past decade [1–3]. The photoelectric conversion efficiency of single junction OSCs has recently achieved over 9% [4]. The key part of OSCs is the active layers which are composed of conjugated polymers or small molecules as donors and fullerene derivatives or metal oxide as acceptors [5–7]. The solar radiation is harvested and converted to photocurrent by the donor–acceptor blends. Nevertheless, there is a quest for donors that can effectively harvest the sub-bandgap photons, thus increasing the photocurrent density compared to the currently used donors and also surpass the Shockley-Queisser limit of single threshold organic solar cells. The optical bandgap (E_g) of the most commonly used donors is ~1.5–2.0 eV, allowing them to absorb sunlight only up to ca. 800 nm [8–11].

In order to achieve the harvesting of sub-bandgap photons (NIR and IR regions) of solar spectrum for organic solar cells, several approaches have been developed, such as designing new low band-gap donors [12], inorganic nanocrystals sensitizers [13] or so-called ternary system consisting of two different donors, in which one donor is sensitive to the NIR photons [14]. An alternative approach for improving light-harvesting in the NIR and IR regions is the utilization of up-conversion nanophosphors (UCNPs), which can convert NIR or IR photons into visible photons [15–17]. Recently, rare earth doped up-conversion systems have been integrated into photovoltaic devices to enhance NIR light absorption and performances [18–20]. Among those up-conversion nanophosphors, due to the low phonon energy of lattices, hexagonal $NaYF_4$ (β-$NaYF_4$) crystals have been reported as the most-efficient host material for

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visible up-conversion when sensitized by Yb^{3+} and activated by Er^{3+} or Tm^{3+} [21–23]. Dionne et al. reported that their calculations show the lanthanide-doped up-converters can significantly increase efficiencies of 1.7 eV bandgap solar cell from 28% to over 34% with improved quantum yield, which show extremely promising of up-conversion for improving the efficiency of solar cells [24].

Here, we developed a simple strategy for demonstrating the up-conversion functionality of lanthanide-doped up-conversion phosphors on OSCs. We synthesized up-conversion nanophosphors $NaYF_4$: Yb/Er (20/2 mol%) nanoplates, mixed them with a polymer as matrix material and coated polymer-UCNPs composite films (UC converter). Afterwards, we combined the UC converter with semi-transparent organic solar cells (ST-OSCs). Under 975 nm laser illumination (ca. 200 mW/cm^2), the OSCs exhibit a short circuit current density of 450 μA/cm^2 and a photoelectric conversion efficiency of ~0.1%. Strikingly, we can demonstrate the up-conversion functionality and observe the performance enhancement in the ST-OSCs with UC converter under AM 1.5 G irradiation (100 mW/cm^2).

The up-conversion nanophosphors $NaYF_4$: Yb/Er (20/2 mol%) nanoplates (UCNPs) were synthesized according to a method reported in literature (see experimental section in Supplementary data) [22]. Bright-field (BF) and high-resolution (HR) transmission electron microscopy (TEM) images of the $NaYF_4$: Yb/Er (20/2 mol%) nanoplates with hexagonal shapes and ca. 270 nm in diameter are shown in Fig. 1a. The selected-area electron diffraction (SAED) (Fig. 1a) and powder X-ray diffraction (XRD) patterns (Fig. 1b) confirm that all the $NaYF_4$: Yb/Er (20/2 mol%) nanoplates are of pure β-$NaYF_4$ phase. The $NaYF_4$: Yb/Er (20/2 mol%) nanoplates exhibit up-conversion luminescence under 975 nm laser excitations under ca. 200 mW/cm^2 power density (Fig. 1c). Three visible emission bands centered at 527, 541, and 660 nm are observed, attributable to the radiative transitions from the ($^2H_{11/2}$, $^4S_{3/2}$) (green) and from the $^4F_{9/2}$ (red) excited states to the $^4I_{15/2}$ ground state of Er^{3+}, respectively. The activator Yb^{3+}, capable of absorbing the ca. 976 nm near-infrared light efficiently (Fig. 1d), transfers its energy sequentially to nearby Er^{3+} through the $^2F_{5/2}$ (Yb^{3+}) → $^4I_{11/2}$ (Er^{3+}) process, pumping the Er^{3+} to its emitting levels (Fig. S2). The up-conversion emission spectrum of UCNPs powder under low power density (ca. 4 mW/cm^2 similar to the density of 975 nm in sunlight) also was shown (Fig. S1), which could demonstrate that the UCNPs could show obvious up-conversion emission signal under NIR sunlight excitation.

Since $NaYF_4$: Yb/Er (20/2 mol%) nanoplates (UCNPs) are a nanocrystalline powder, a matrix material is needed to form a solid up-conversion layer. We have tested two matrix materials: Polymethyl methacrylate (PMMA) and Polystyrene (PS). In addition to the requirement to form a solid layer, the optical properties of the matrix materials are important. An ideal binding agent must have low absorption as well as emission in the spectral region of

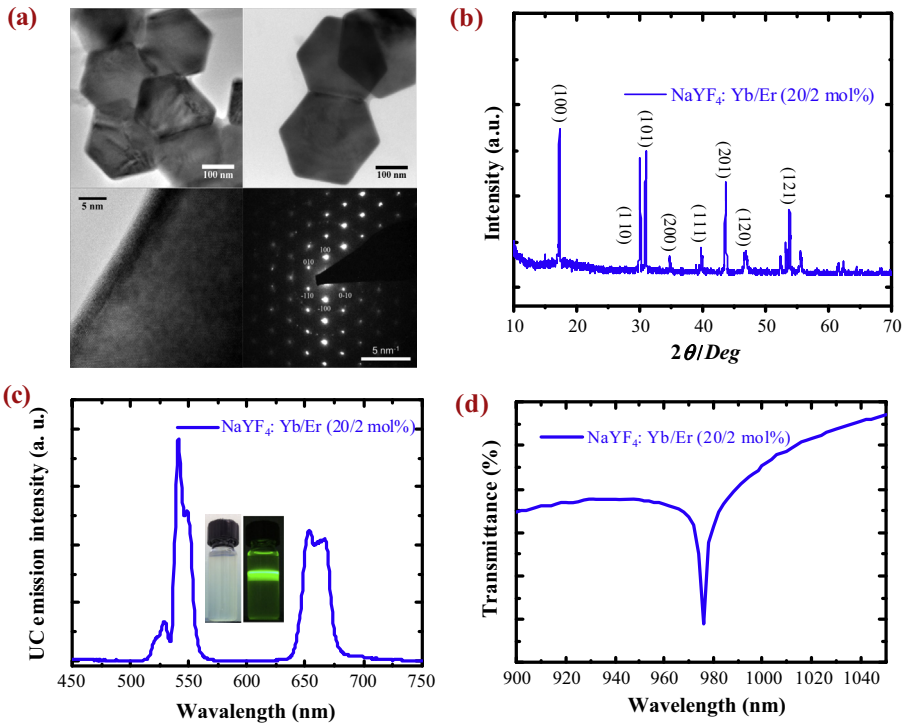

Fig. 1. (a) BF-TEM images, HRTEM image and SAED pattern of $NaYF_4$: Yb/Er (20/2 mol%) nanoplates (UCNPs); (b) powder XRD pattern of (UCNPs). The SAED (view along close to [001] zone axis) and XRD patterns confirm the β-NaYF4 phase of the nanoplates; (c) room temperature up-conversion emission spectrum of the UCNPs powder, the insert of (c) clearly show the strong green emission of the UCNPs in toluene under 975 nm laser excitation; (d) absorption spectrum of the UCNPs in film, the absorption band at ca. 977 nm originates from the absorption of Yb^{3+} in the UCNPs. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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