

# Comparative analysis of upconversion efficiencies in fluoride materials for photovoltaic application



Elena Favilla<sup>b,\*</sup>, Giovanni Cittadino<sup>a</sup>, Stefano Veronesi<sup>b</sup>, Mauro Tonelli<sup>b</sup>, Stefan Fischer<sup>c</sup>, Jan Christoph Goldschmidt<sup>c</sup>, Arlete Cassanho<sup>d</sup>, Hans P. Jenssen<sup>e</sup>

<sup>a</sup> Dipartimento di Fisica, Università di Pisa, Largo B. Pontecorvo 3, I-56127 Pisa, Italy

<sup>b</sup> NEST Istituto Nanoscienze-CNR Pisa, Piazza San Silvestro 12, 56127 Pisa, Italy and Dipartimento di Fisica dell'Università di Pisa, Largo B. Pontecorvo 3, Pisa, Italy

<sup>c</sup> Fraunhofer Institute for Solar Energy Systems ISE, Heidenhofstrasse 2, 79110 Freiburg, Germany

<sup>d</sup> AC Materials, Inc., 756 Anclote Road, Tarpon Springs, FL 34689, USA

<sup>e</sup> CREOL, The College of Optics and Photonics, University of Central Florida, 4000 Central Florida Boulevard, Orlando, FL 32816, USA

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## ABSTRACT

Infrared to visible upconversion (UC) is a promising way to enhance the efficiency of silicon based solar cells. In this paper, the spectral conversion and recovery of sub-band gap photons of the solar spectrum, from NIR-IR to the VIS-NIR wavelength region, is investigated in two fluorides hosts doped with trivalent erbium ions ( $\text{Er}^{3+}$ ). The efficiency gain due to upconversion in silicon solar cells is compared for single crystal samples of  $\text{BaY}_2\text{F}_8:\text{Er}^{3+}$  and  $\text{LiYF}_4:\text{Er}^{3+}$  in a dedicated upconverter solar cell device (UCSCD) with monochromatic excitation in the  $1.5\ \mu\text{m}$  spectral region.

The highest external quantum efficiency due to upconversion was found for the UCSCD using the  $\text{BaY}_2\text{F}_8:30\ \text{at}\% \text{Er}^{3+}$  single crystal, reaching an EQE of  $6.8 \pm 0.2\%$  for  $(1.10 \pm 0.12) \cdot 10^5\ \text{W m}^{-2}$  spectral irradiance at  $1494\ \text{nm}$ .

We present a comprehensive spectroscopic study of the crystal samples also taking into account the effects of the different crystal symmetry as well as the different phonon energies. Our findings enable us to explain the higher efficiency of the  $\text{BaY}_2\text{F}_8:\text{Er}^{3+}$  compared to the  $\text{LiYF}_4:\text{Er}^{3+}$  upconverter in terms of both static and dynamic properties.

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## 1. Introduction

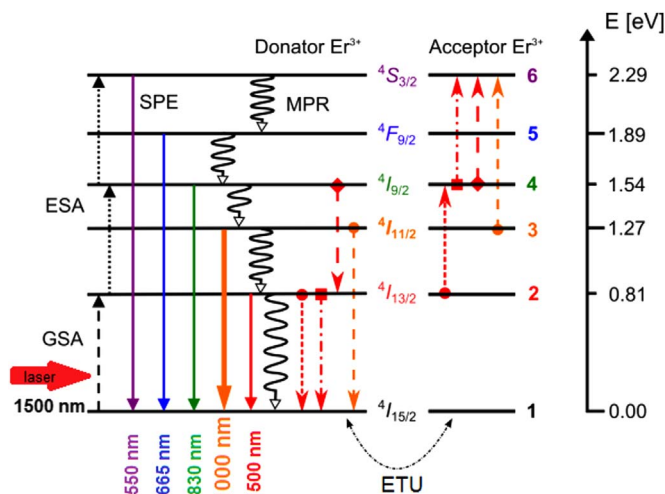
One of the most important limiting factors for the efficiency of a solar cell is related to photons with energy below the band gap of the solar cell material, which are not utilized. This low energy part of the solar spectrum can be accessed using upconversion processes where two or more incoming photons, interacting with the upconverter, are transformed in one photon having an energy that is higher than the single incoming photon one.

The upconverting property of rare earth (RE) materials is widely used in many applications such as solid state lasers [1], light emitting devices like infrared indicator cards, bio-labels [2] or for three-dimensional displays [3].

Among the possible upconverters to be used with silicon solar cells, erbium is one of the most promising due to the  $^4\text{I}_{15/2} \rightarrow ^4\text{I}_{13/2}$  ground state absorption around  $1500\ \text{nm}$  and emissions in the green ( $\sim 540\ \text{nm}$ ), red ( $\sim 650\ \text{nm}$ ) and NIR ( $\sim 980\ \text{nm}$ ), within the

useful range for this kind of solar cell (see Fig. 1). In fact, the dominant emission at  $980\ \text{nm}$  can be efficiently used by a silicon solar cell. The pioneering studies of this approach were performed using sodium yttrium fluoride ( $\text{NaYF}_4$ ). It has been successfully applied as upconverter in the hexagonal form ( $\beta\text{-NaYF}_4$ ), doped with trivalent erbium ions ( $\text{Er}^{3+}$ ) on the rear side of a bifacial silicon solar cell as reported by Shalav et al. [4,5] and Fischer et al. [6]. In addition to  $\text{NaYF}_4$ , gadolinium oxysulfide ( $\text{Gd}_2\text{O}_2\text{S}$ ) host lattice doped with erbium has been studied intensively [7–12]. It shows an intense UC luminescence and it has demonstrated to be a good converter in biological imaging application, where a monochromatic excitation is needed, while for silicon solar cells, where a broader excitation spectrum is present,  $\beta\text{-NaYF}_4:25\ \text{at}\% \text{Er}^{3+}$  powders showed a better performance as upconverter [13]. More recently upconverter solar cell devices (UCSCDs) with  $\beta\text{-NaYF}_4:25\ \text{at}\% \text{Er}^{3+}$  have shown a relative enhancement of  $0.16\%$  in the short-circuit current under broad band excitation ranging from  $1450$  to  $1600\ \text{nm}$  with a correspondent EQE of  $1.28\%$  for an irradiance of  $2570\ \text{W m}^{-2}$  [14]. A relative increase in the solar cell's efficiency of  $0.19\%$  was reported for illumination from a solar

\* Corresponding author.



**Fig. 1.** Energy level diagram of  $\text{Er}^{3+}$ , depicting the five lowest energy levels and the most important processes involved in the upconversion. These are especially ground state absorption (GSA), energy transfer upconversion (ETU), multi-phonon relaxation (MPR) and spontaneous emission (SPE).

simulator, where the light was concentrated with a lens by a factor of 210 [13].

Because of its good performance relative to other upconverter materials, up to now most of the work concerning PV-UC applications was done using  $\beta\text{-NaYF}_4$  microcrystalline powders doped with erbium as upconverter. However, the achieved enhancements of the solar cell efficiency are still not sufficient for a wide-spread application of upconversion in photovoltaic. Consequently there is a need for exploring new materials that are able to significantly enhance the total efficiency of photovoltaic devices. The family of fluorides, due to their favourable optical properties both as single crystals or microcrystalline powders, is a promising area to search candidates for this application.

Due to a stronger Stark splitting, doped fluorides show excitation bands much broader than other host materials such as chlorides [15]. Hence they absorb a larger fraction of the incident solar spectrum. A key point for the efficiency of the UC process is related to phonon energy which strongly influences the performances by non-radiative losses, especially due to multiphonon decay. Therefore, the phonon cut off energy of the medium plays a vital role: higher phonon energy leads to a higher rate of non-radiative transitions and typically to a lower UC efficiency. Due to their low phonon energy, most fluoride crystals are attractive hosts to realize upconverters based on RE trivalent ions. Furthermore, their good thermo-mechanical and chemical properties guarantee their durability, small aging effects, and they allow a large doping concentration which consequently leads to strong absorption of the incident light. Furthermore, large doping results in small average distances between the RE ions, which enhances energy transfer processes, such as energy transfer upconversion (ETU), and therefore increases the upconversion quantum yield at least for the low irradiance conditions that are relevant in photovoltaic applications. However, for very high RE doping the UCQY decreases due to concentration quenching. The optimal concentration is a result of balance between the beneficial ETU processes and the concentration quenching for high RE doping, which also depends on the irradiance of the excitation [11].

One fluoride host material which is a good alternative to  $\beta\text{-NaYF}_4$  is  $\text{BaY}_2\text{F}_8$ .  $\text{BaY}_2\text{F}_8$  was used already in a preliminary work on a PV-UC device with a monofacial silicon cell [16] and showed a potential EQE comparable to the “reference”  $\beta\text{-NaYF}_4$  materials used for this type of application. In another work,  $\text{BaY}_2\text{F}_8$  showed that it can even outperform  $\beta\text{-NaYF}_4$  as a host material for erbium as

upconverter in photovoltaic applications. Indeed, at an illumination with a solar simulator and solar concentration of  $94 \pm 17$  suns, the upconverter solar cell device using a  $\text{BaY}_2\text{F}_8:30 \text{ at\% Er}^{3+}$  sample showed a record relative enhancement of the short-circuit current of  $0.55 \pm 0.14\%$  [17]. Another, yet unexplored option is  $\text{LiYF}_4$ . This crystalline host is a very well-known material that is often used as a reference in laser sources development, because of its good thermo-mechanical properties, low phonon energies and negative  $dn/dT$  benefiting low thermal lensing [18]. It is at present widely used in commercial devices (even operating at high power), and it has been used as an active laser medium doped with almost all rare earth ions. However, it has only rarely been used in photovoltaic [19] and no good quantitative data exists.

In this paper, we report a comparison of two fluoride hosts, the current champion material single crystalline  $\text{BaY}_2\text{F}_8$  with the new option of single crystalline  $\text{LiYF}_4$ , for the application in photovoltaics. Samples were realized from high optical quality crystal boules grown with the Czochralski technique. The single-crystal samples of  $\text{BaY}_2\text{F}_8:30 \text{ at\% Er}^{3+}$  were attached to the rear side of a planar bifacial silicon solar cell that features broad-band anti-reflection coatings and was optimized for the UC application [20]. The effect of reabsorption by the crystal was evaluated to achieve the best performances, by utilizing several thicknesses correspondent to different absorbed pumping powers. The results are then compared with those for  $\text{LiYF}_4$  single crystal doped with 25 at%  $\text{Er}^{3+}$ . Our results show the appealing performances of these materials to realize innovative photovoltaic devices.

## 2. Material and methods

### 2.1. Upconverter materials

In this work, we explored and compared two materials as upconverter for silicon solar cell devices:  $\text{BaY}_2\text{F}_8$  (BYF in the following) doped with 30 at%  $\text{Er}^{3+}$  and  $\text{LiYF}_4$  (YLF in the following) doped with 25 at%  $\text{Er}^{3+}$ . The Er:BYF single crystal was grown in a homemade furnace with conventional resistive heating and automatic optical diameter-control. All the raw powders utilized in the growth, having 5 N purity, were provided by AC Materials (Tarpon Springs, FL, USA). The chosen doping concentration was achieved by adding a proper amount of  $\text{ErF}_3$  and of  $\text{BaF}_2$  to the  $\text{BaY}_2\text{F}_8$  powders [21].

The BYF host lattice has a monoclinic structure belonging to the space group C2/m, each primitive cell has two molecules while lattice constants are  $a=6.972 \text{ \AA}$ ,  $b=10.505 \text{ \AA}$ ,  $c=4.260 \text{ \AA}$ , and angles between crystallographic axes are  $\alpha=90^\circ$ ,  $\beta=90^\circ$  and  $\gamma=99.76^\circ$ . Additionally, it has a refractive index  $n \cong 1.5$  and low phonon energy ( $\cong 350 \text{ cm}^{-1}$ ) [22]. The monoclinic structure is an important feature of this crystal because it introduces a low symmetry structure, which leads to wider absorption and emission bandwidths.

The Er:YLF single crystal was provided by AC Materials (Tarpon Springs, FL, USA).

$\text{LiYF}_4$  is an uniaxial crystal with a tetragonal structure belonging to the Scheelite ( $\text{CaWO}_4$ ) family and having C64h (I41/a) space group. The primitive cell has four molecules and its dimensions are:  $a=b=5.160 \text{ \AA}$ ,  $c=10.85 \text{ \AA}$  [23]; rare earth ions substitute  $\text{Y}^{3+}$ , in a site having point symmetry S4. Its transparency region extends from the visible up to infrared ( $5 \mu\text{m}$ ). It has a refractive index  $n \cong 1.5$ .

The structural investigation on the boules by X-ray Laue technique allowed for identifying the crystallographic axes of the crystals and confirms the single crystal character of the boules. Moreover an optical analysis with a laser beam probe confirmed

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