# CALIBRATED PHOTOLUMINESCENCE MEASUREMENTS OF THE UPCONVERTER $NaYF_4$:20% $Er^{3+}$ FOR SILICON SOLAR CELLS

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ABSTRACT: Upconversion (UC) of subband-gap photons is promising to enhance solar cell efficiency. We explore the properties of $\beta$-$NaYF_4$:20% $Er^{3+}$ for this application. This material shows efficient UC suitable for semiconductor based solar cells with band gaps up to 1.25 eV. In this work we investigate the optical UC efficiency by calibrated photoluminescence measurements. The values were obtained in dependence of the excitation wavelength as well as the irradiance. The highest optical UC quantum efficiency of 4.3% was obtained for monochromatic irradiance of 1390 $Wm^{-2}$ at 1523 nm. Because these data from optical measurements do not account for any losses associated with the application of the upconverter to a solar cell, they represent the upper limit of energy conversion for an optimized solar cell UC device.

## 1 INTRODUCTION

Upconvertion (UC) materials can generate higher energy photons from (a larger number of) lower energy photons. One interesting potential application of such materials is photovoltaics. To decrease photovoltaic electricity costs, it is important to both reduce production costs and to increase solar cell efficiency. Silicon is the dominant material for commercial solar cell production. However, silicon solar cells absorb less than 55% of the incident photons. More than 45% of all photons of the solar spectrum are not utilized, because the energy of these photons is less than the band-gap energy of silicon. These photons carry about 20% of the whole energy of the sun's radiation. In consequence, transforming these sub-band-gap photons into higher energy photons that can be utilized by the solar cell could significantly enhance the efficiency of the solar cell [1, 2]. Analysis shows that the theoretical upper efficiency limit of a silicon solar cell is raised from near 30% [3] up to 40.2% [4] for a silicon solar cell with upconverter illuminated by non-concentrated light.

Hexagonal sodium yttrium fluoride doped with trivalent erbium ($\beta$-$NaYF_4$:20% $Er^{3+}$), is known to show the highest UC efficiencies in the near infrared spectral region relevant for UC in silicon solar cells [5, 6]. $Er^{3+}$ has energy levels conveniently spaced for UC, see Figure 1 [7]. The $NaYF_4$ host lattice has low phonon energies [8] which reduces non-radiative losses. Several sites and the disorder in $\beta$-$NaYF_4$ [9] results in a broader absorption range of the $^4I_{13/2}$ energy level compared to other $Er^{3+}$ materials [10].

UC in rare earth ions occurs by different mechanisms. The dominant mechanisms are ground state absorption (GSA), followed by either excited state absorption (ESA) or energy transfer upconversion (ETU) [11]. Generally stated, the absorption of more than one photon populates higher energy levels of the upconverter via intermediate levels or cooperative processes. Subsequently, a photon with more energy than the absorbed ones can be emitted by radiative transition to the ground state. The UC photon can produce a free carrier in the solar cell. In consequence, the transmission losses are reduced. Since the sub-band-gap photons are transmitted through the semiconductor, the upconverter can be placed on the backside of the solar cell. This is a big advantage because the upconverter does not interfere with the electronic properties of the solar cell or cause optical losses at its front side. As the photons used by the upconverter have been transmitted by the solar cell anyway, every UC photon absorbed by the cell can create an extra free carrier that increases the current of the solar cell.

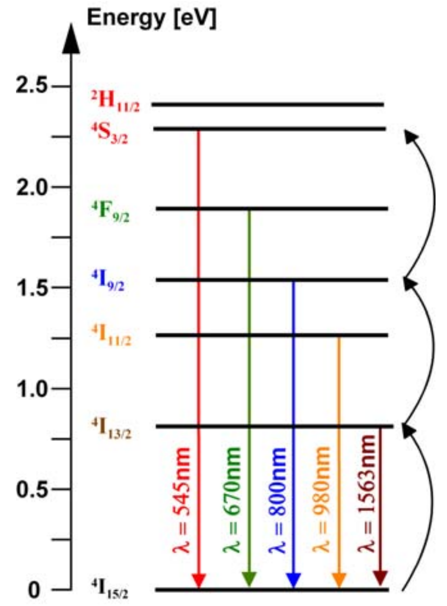

**Figure 1:** Energy levels and corresponding wavelengths of the radiative transitions to the ground state of $Er^{3+}$. The population of higher energy levels by the absorption of several 1.5 μm photons is illustrated by the curved arrows. The data was taken from [7].

For the application in photovoltaics, the efficiency of the UC process over the spectral range of upconvertible photons is the key figure of merit. In many previous works the UC efficiency has been measured indirectly by applying the upconverter to solar cells, which served as detector [4, 6, 10, 12-15]. While these measurements yield the efficiency of the solar cell UC device, as is relevant for the later application, the actual UC efficiency could only be calculated by estimating the different loss mechanisms occurring in the solar cell UC device. For example, the transmission of sub-band-gap photons of the solar cell reduces the amount of photons that eventually reach the upconverter. Because of the non-linear characteristic of the upconverter, a reduction of irradiance on the upconverter reduces the UC efficiency itself. Furthermore, there are losses of the light emitted by the upconverter until it reaches the solar cell. The solar cell itself has an external quantum efficiency smaller than one for the light it receives from the upconverter, which is mostly due to reflection at the surface of the solar cell. As the experiments were performed with spot laser illumination, it is reasonable to assume that the solar cells were not fully illuminated during the experiments. Unfortunately, not all publications give the geometric dimensions of their devices. Inhomogeneous illumination results in parts of the solar cell being illuminated while other parts remain dark. This can reduce the external quantum efficiency of the solar cell below the level determined by external quantum efficiency measurements under homogeneous illumination with additional bias-light, because silicon solar cells do not respond linearly at low illumination (below 0.5 suns) [16]. Moreover, the UC material is usually embedded in a binding agent for its application on the solar cell. Both optical properties of these materials and potential chemical processes during the embedding process can affect the UC material [17]. Consequently, the determination of the optical UC efficiency from UC solar cell devices is plagued by several uncertainties, as not all these loss-mechanisms can be easily quantified.

Hence it is advantageous to know the optical UC efficiency of the unmodified material, as this is the upper limit that can be achieved with an optimized device and processing. Therefore, optical measurements of the UC material are an attractive alternative. Many previous works using optical methods, however, remained on a qualitative level and no optical UC efficiency was determined [13, 14, 18]. Only a few publications discussed the UC efficiency as determined by optical measurements [11, 19, 20], but excitation intensities had been much higher than expected for photovoltaic applications.

To overcome these limitations, we present the UC efficiency of $\beta$-NaYF$_4$:20% Er$^{3+}$, determined by calibrated photoluminescence measurements. The dependence of the UC efficiency on the excitation wavelengths and the irradiance is investigated as well.

## 2 MEASUREMENT SETUP AND CALIBRATION

Because of reported [6] high UC efficiencies we used $\beta$-NaYF$_4$:20% Er$^{3+}$ in our investigation. This material is a microcrystalline powder. Therefore, it was filled in a powder cell with an optical window on the front. The thickness of the compacted powder layer is more than 3 mm in order to achieve complete absorption of the incident photons within this layer. The upconverter was illuminated through the optical window with an IR-Laser ECL-210 from Santec. The incident wavelength of the laser $\lambda_{inc}$ can be tuned from 1430 nm to 1630 nm and the power of the laser diode can be varied up to approximately 8 mW, depending on the operating conditions. Since UC is a non-linear process, it is very important to know the irradiance on the sample. To determine the irradiance we measured the incident photon flux of the excitation $\Phi_{inc}$ at different laser powers with a germanium solar cell. The external quantum efficiency (EQE) of the germanium solar cell is known from characterization with standard solar cell characterization equipment with an uncertainty of 3% relative. The diameter of the laser beam was $3.2 \pm 0.1$ mm$^2$. With these data and the geometrical properties of the experiment, the irradiance $I$ of the laser on the sample could be calculated.

The luminescence spectra of the upconverter were measured with a grating monochromator H25 from Jobin Yvon, a silicon photodiode detector from OEC, and a lock-in-amplifier 7265 from Signal Recovery. The detector is thermo-electrically cooled to minimize thermal noise. The chopper was placed between sample and monochromator and operated at a frequency of 15 Hz. Because of the long lifetimes of the involved Er$^{3+}$ states in the range of ms, it is unfavorable to place the chopper in the excitation beam [21]. A schematic graph of the measurement setup is shown in Figure 2.

The monochromator is optimized for infrared radiation and features a gold grating. Because of the absorption properties of the gold, only luminescence with wavelengths longer than approximately 600 nm could be observed. The systematic error introduced by this restriction is much lower than the uncertainty of the calibration. The emission from the transition of the $^4S_{3/2}$ to the $^4I_{15/2}$ with a wavelength of 545 nm contributes only a fraction of 0.3% to all emitted photons [8], while the transitions of higher energies contribute even less.

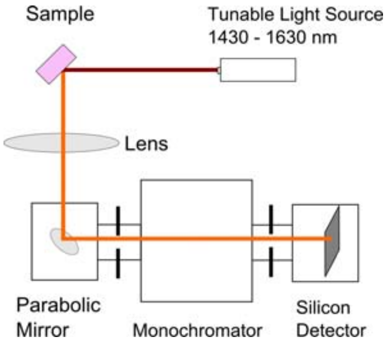

**Figure 2:** Scheme of the measurement setup.

To determine the calibrated efficiency of the UC on an absolute scale, the efficiency of the used detection unit must be known. To achieve this goal, the relative spectrum of a tungsten halogen lamp was determined at ISE Callab. This lamp was placed in the excitation light path of the setup. The total number of photons at the sample position was determined subsequently with the aforementioned germanium solar cell. Afterwards, a white reflector made from BaSO$_4$ coated roughened aluminum was placed at the sample position. Goniometer measurements on this reflector confirmed that the reflected light had an angular characteristic of a Lambertian reflector. By combining these results, the reflected photon flux per wavelength and solid angle from the light source in the direction of the detection system is known. By comparing with the output of the detection unit, the detection efficiency could be determined. Details on the calibration can be found in [22].

Throughout this paper we will state the effective efficiency of the system of the powder cell with the UC powder. Overall an error of 8% is estimated for the uncertainty of this efficiency. The main uncertainties stem from the inaccuracy of the calibration of the photoluminescence setup and the external quantum efficiency of the reference solar cell to calculate the photon flux of the excitation.

Throughout this paper, we define the spectral optical UC efficiency $\eta_{UC,spectral}(\lambda_{inc}, \lambda_{UC}, I)$ at a certain luminescence wavelength $\lambda_{UC}$ under the excitation with a wavelength $\lambda_{inc}$ and an irradiance $I$ as

$$\eta_{UC,\,spectral}(\lambda_{inc}, \lambda_{UC}, I) = \frac{\Phi_{UC}(\lambda_{UC}, I)}{\Phi_{inc}(\lambda_{inc}, I)}, \qquad (1)$$

where $\Phi_{UC}(\lambda_{UC}, I)$ is the photon flux of the UC photons with a wavelength $\lambda_{UC}$ and $\Phi_{inc}(\lambda_{inc}, I)$ the total incident flux of photons with a wavelength $\lambda_{inc}$ onto the powder cell.

With this definition, the maximum optical UC efficiency that can be reached is 50%, because at least two sub-band-gap photons must be absorbed to generate one UC photon. The maximum optical UC efficiency for UC involving three incoming photons is correspondingly lower.

## 3 PHOTOLUMINESCENCE MEASUREMENTS

### 3.1 Dependence on the excitation wavelength

The excitation wavelength was varied between 1430 nm and 1630 nm in 2 nm steps and the photoluminescence spectrum of the $NaYF_4$:20% $Er^{3+}$ was recorded at each excitation wavelength. Figure 3 shows the spectral optical UC efficiency at a constant irradiance of 880 $Wm^{-2}$. Three peaks from the transition of the states $^4F_{9/2}$, $^4I_{9/2}$ and $^4I_{11/2}$ to the ground state $^4I_{15/2}$ are visible. The corresponding luminescence peaks are centered at the wavelengths of 660 nm, 810 nm and 980 nm respectively. By integrating over the luminescence wavelength $\lambda_{UC}$, the integrated optical UC efficiency $\eta_{UC}(\lambda_{inc}, I)$ at a certain excitation wavelength $\lambda_{inc}$ was calculated:

$$\eta_{UC}(\lambda_{inc}, I) = \int \eta_{UC,spectral}(\lambda_{UC}, \lambda_{inc}, I)\, d\lambda_{UC} \qquad (2)$$

Silicon solar cells use photons up to a wavelength of approximately 1150 nm. Therefore, the integrated optical efficiency including these three peaks is a good estimation of how efficiently sub-band-gap photons can be upconverted into photons that can be used by a silicon solar cell. In Figure 4 the integrated optical efficiency in dependence on the excitation wavelength is plotted. The $\eta_{UC}(\lambda_{inc}, I)$ peaks at a wavelength of 1523 nm, reaching 3.0% for a constant irradiance of 880 $Wm^{-2}$. In the spectral range from 1492 nm to 1547 nm the efficiency of the UC is higher than 1.5%, which constitutes a full width at half maximum (FWHM) of 55 nm.

This excitation spectrum features distinctive peaks. They result from the crystal field splitting of the $Er^{3+}$ energy levels and the spectral overlap of the UC steps between these levels [7]. The UC luminescence and especially the UC efficiency not only depend on the excitation wavelength, but also on the irradiance of the sample. This dependency is reviewed in the following section.

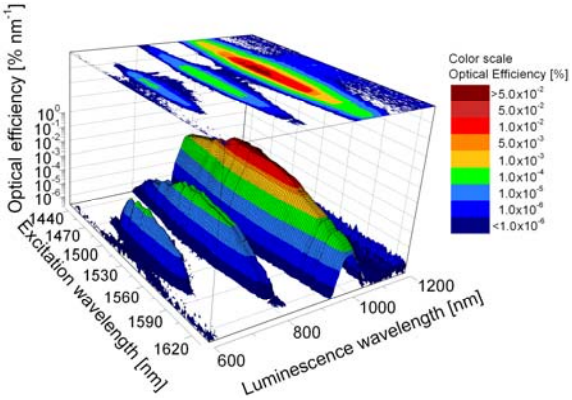

**Figure 3:** Spectral optical UC efficiency $\eta_{UC,spectral}(\lambda_{inc}, \lambda_{UC}, I)$ of the $NaYF_4 : 20\, Er^{3+}$ in a logarithmic scale calculated from calibrated photoluminescence measurements. The irradiance $I$ was kept constant at 880 $Wm^{-2}$. Emission peaks occur at 660 nm, 810 nm and a dominating one at 980 nm, cf. Figure 1.

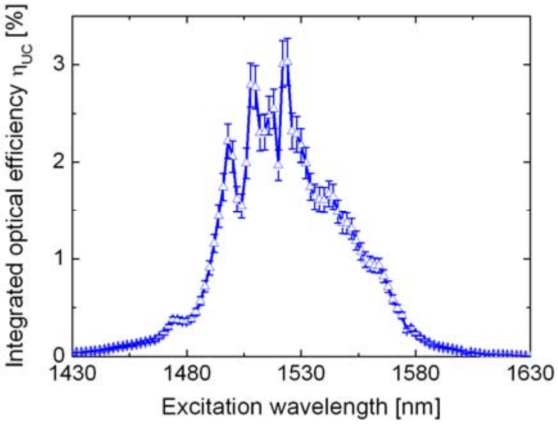

**Figure 4:** The integrated optical UC efficiency $\eta_{UC}(\lambda_{inc}, I)$ is calculated by integration over the luminescence wavelength $\lambda_{UC}$ in Figure 3. Photons with a wavelength of $\lambda_{UC}$ = 1523 nm are most efficiently upconverted. The FWHM is 55nm from 1492 nm to 1547 nm.

### 3.2 Dependence on the irradiance

The power dependence of the UC luminescence of $NaYF_4$:20% $Er^{3+}$ was measured at its most efficient wavelength of 1523 nm. The laser power was varied from 0.1 mW to the maximum stable output power of 7.9 mW which corresponds to an irradiance of 17 $Wm^{-2}$ to 1370 $Wm^{-2}$ in our setup. The dependence of the integrated UC efficiency on the irradiance is shown in Figure 5. The UC efficiency increases with the irradiance. For higher irradiance values the slope decreases due to saturation. The maximum $\eta_{UC}(\lambda_{inc}, I)$ in the present setup was 4.3% for an irradiance of 1370 $Wm^{-2}$.

From theory, it is expected that $I$ follows a quadratic power law for low irradiances, hence with a characteristic exponent $m = 2$ [11]. As the integrated optical UC efficiency is derived from the luminescence data by dividing by the irradiance, one expects a power law with an exponent $m-1$ for the dependence of the UC efficiency on the irradiance. To determine the characteristic exponent $m$, we performed a least-square fit to the data of the integrated optical UC efficiency with the function

$$\eta_{UC}(\lambda_{inc}, I) = c_1 \times I^{m-1} + c_2\,, \tag{3}$$

where $c_1$ and $c_2$ are additional fitting parameters. We determined the characteristic exponents for two different ranges of irradiance values. For the lower range between 0 $Wm^{-2}$ and 490 $Wm^{-2}$ the characteristic exponent is $m = 1.86$. In the higher range between 440 $Wm^{-2}$ and 1050 $Wm^{-2}$ the characteristic exponent is $m = 1.35$. The decrease of m reflects saturation effects as excitation of higher energy levels [23].

This nonlinear characteristic of the upconverter makes it difficult to compare efficiency values obtained at different irradiances. Therefore, Auzel [11] established the notation $\eta_{UC,norm}(\lambda_{inc}, I)$ for a *normalized* UC efficiency, which explicitly connects the irradiance $I$ with the UC efficiency:

$$\eta_{UC,norm}(\lambda_{inc}, I) = \frac{\eta_{UC}(\lambda_{inc}, I)}{I} \tag{4}$$

For an ideal two photon UC process in a three level system, the characteristic exponent $m$ would be 2 and the UC efficiency would depend linearly on the incident irradiance at low irradiances. Following the definition, the normalized UC efficiency $\eta_{UC,norm}(\lambda_{inc}, I)$ would therefore be constant. However, in a real system, the *normalized* values are not constant, because the characteristic exponents are not exactly 2 and vary with the irradiance. Since some publications in the past named only the normalized efficiency, we present the normalized values in this work as well for comparison.

The obtained values for $\eta_{UC}(\lambda_{inc}, I)$ of 4.3% at 1370 $Wm^{-2}$ yield a $\eta_{UC,norm}(\lambda_{inc}, I)$ of 0.31 $cm^2W^{-1}$. Recently an absolute UC efficiency of 12.7% for an Er-doped fluoride glass was presented at an irradiance of approximately $10^9$ W/m² [19], resulting in a $\eta_{UC,norm}(\lambda_{inc}, I)$ of approximately $10^{-4}$ $cm^2W^{-1}$ which is considerably lower than our values. In [6] a *normalized* UC efficiency of 0.07 $cm^2W^{-1}$ (16.7% at 24000 $Wm^{-2}$) was presented, which was not measured directly, but calculated from electrical measurements on solar cell devices with attached upconverter by considering estimated losses. This comparison shows that our values are the highest reported values for the normalized efficiency $\eta_{UC,norm}(\lambda_{inc}, I)$. This result, however, must be interpreted with care. As we have seen in Figure 5, the slope of the efficiency curve and the characteristic exponent decrease. Therefore, the *normalized* UC efficiency $\eta_{UC,norm}(\lambda_{inc}, I)$ is higher at low excitation irradiances used in our experiments and lower at higher excitation irradiances used in the cited works. In consequence, comparing efficiency values obtained at different irradiances remains a challenge. The *normalized* notation proposed by Auzel can help to compare values but as a linear behavior of the efficiency is assumed, its usage should be limited to regions of the irradiance where $m$ does not differ significantly from 2. Instead, extrapolations with the help of fitted parameters can be used for comparison. Using equation (3) and the corresponding fitting parameters from the least-square fit in the higher irradiance range, we calculated a $\eta_{UC,norm}(\lambda_{inc}, I)$ of 0.07 (17.3% at $I$ = 24000 $Wm^{-2}$), which is in agreement with the value from Richards [6]. The fitting parameters for this calculation have been c1 = 0.62 and c2 = -3.21. Overall, the achieved high efficiencies make $\beta$-NaYF$_4$:20% Er$^{3+}$ a promising material for UC of sub-band-gap photons.

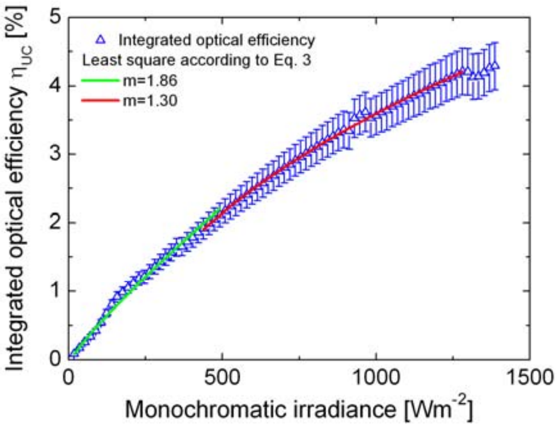

**Figure 5:** Due to the non-linear UC process, the integrated optical UC efficiency $\eta_{UC}(\lambda_{inc}, I)$ increases with the irradiance $I$. At high irradiance values the increase slowly saturates. Therefore, the characteristic exponent $m$ decreases from 1.86 to 1.35.

## 4 SUMMARY AND OUTLOOK

In this paper, we explored the potential of $\beta$-NaYF$_4$:20% Er$^{3+}$ to significantly reduce the sub-band-gap losses of silicon solar cells by UC. UC efficiencies have been determined directly from calibrated photoluminescence measurements, both depending on of the excitation wavelength and the excitation irradiance.

An UC efficiency $\eta_{UC}(\lambda_{inc}, I)$ of 4.3% was determined at a wavelength of 1523 nm for a moderate irradiance of 1390 $Wm^{-2}$. Following the definition of optical UC efficiency, the value of 4.3% means that the energy of 8.6% of the incident photons has been utilized.

A normalized UC efficiency $\eta_{UC,norm}(\lambda_{inc}, I)$ of 0.31 $cm^2W^{-1}$ was calculated from the UC efficiency $\eta_{UC}(\lambda_{inc}, I)$. To our best knowledge this is the highest reported value to date. We have discussed in detail that the normalized values are of limited relevance for the comparison of efficiency values that were determined at different irradiances. Instead we propose to extrapolate the irradiance dependent measurements to a comparable irradiance. Using extrapolation, our data are in good agreement with data from literature [6].

The absorption width (FWHM) of NaYF$_4$:20% Er$^{3+}$, where photons can be efficiently converted, is around 55 nm. This absorption is narrow compared to the whole spectral range of subband-gap photons. There are two possibilities to overcome this limitation: First, a material with different rare earth ions and therefore broader absorption range might be used. Second, the upconverter could be combined with luminescent materials absorbing in a broad spectral range and emitting in the absorption range of the upconverter [5, 24]. The second concept has the additional advantage that the photon flux in the absorption range of the upconverter is increased. Because of the non-linear characteristics of the upconverter an additional efficiency increase is expected. Consequently, such combined systems are an attractive field for further research.

## 5 AKNOWLEDGEMENT

The presented work is partly funded by the German Federal Ministry of Education and Research in the project "Nanovolt - Optische Nanostrukturen für die Photovoltaik" (BMBF, project number 03SF0322H). The research leading to these results has also received funding from the European Community's Seventh Framework Programme (FP7/2007-2013) under grant agreement n° [246200]. S. Fischer gratefully acknowledges the scholarship support from the Deutsche Bundesstiftung Umwelt DBU and P. Löper the scholarship support from the Reiner Lemoine Stiftung.

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