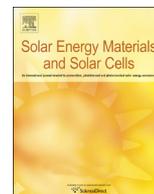




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Experimental enhancement of the photocurrent in a solar cell using upconversion process in fluoroindate glasses exciting at 1480 nm

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ABSTRACT

In this work, the generation of photocurrent in a commercial solar cell has been achieved under excitation at 1480 nm in fluoroindate glass samples codoped with Yb³⁺ and Er³⁺ ions. These samples were placed over a solar cell and under excitation at 1.48 μm, upconversion emissions coming from the Yb³⁺ and Er³⁺ ions were obtained. In the upconversion emission spectra, several emission bands at 545 nm, 660 nm and 975 nm have been detected for all the analyzed samples (doped with different Yb³⁺ and Er³⁺ concentrations). The dependence of the external quantum efficiency to produce photocurrent with the Er³⁺ and Yb³⁺ concentration and with the pump power excitation at 1480 nm has been analyzed. As conclusion, the maximum generation of photocurrent was obtained with the highest and lowest concentrations of Er³⁺ and Yb³⁺, respectively.

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1. Introduction

The renewable energy sources are being strongly studied and supported in the recent years in many developed countries. Among them, photovoltaic's one is the most broadly spread technologies. Nowadays, there are new kinds of solar cells constituted with new materials as carbon nanotubes and/or organic semiconducting polymers. These ones are on research and show good perspectives for this technology in the future [1,2]. However, more than 90% of the current solar cells in use are made of crystalline-silicon and, as a consequence, the improvement of the efficiency of conventional Si solar cells remains as a very appealing target for a near future. These solar cells only take advantage in the visible to NIR range up to 1100 nm, because these high energy photons exceeds the Si bandgap followed by the generation of the electron–hole pair, and therefore the generation of electrical energy. However, the mid-infrared range does not contribute, because this infrared radiation energy does not exceed the Si bandgap. As about 20% of the solar irradiation has a wavelength above 1100 nm, a potential improvement of the solar cell efficiency might be achieved if part of this energy could be used by the Si solar cells.

There are many reviews that suggest many ways to improve the solar cells efficiency, consisting to incorporate lanthanide doped materials (Er³⁺, Yb³⁺, Ho³⁺ or combination of them) on to a solar cell, taking advantage of their optical properties to convert the infrared radiation to visible light [3–8]. This kind of process, called upconversion, is a luminescence process where two low-energy infrared photons are absorbed by rare-earth ions to generate a high-energy visible photon. As example, the energy transfer upconversion process (ETU), consists in successive transfer processes between excited ions under excitation in the near infrared. Therefore, an excited ion can reach a high energetic level from which de-excites emitting a visible photon.

By this way, an upconverter material could be placed below the solar cell in order to absorb the infrared light (not absorbed by the Si) and produce visible photons to excite the solar cell, enhancing its response to infrared radiation. There are many upconversion materials, but usually they are doped with lanthanide ions because they are efficient in these upconversion processes. These ions are most commonly found in the trivalent state and their rich energy level structure over a broad spectral range explains their numerous applications [9]. Among them, the Er³⁺ ions have been the most used. The Er³⁺ ions absorb the solar radiation around 1.54 μm. Two Er³⁺ ions that have absorbed this radiation can interact with each other through an upconversion process. The excited ion emits light above the Si bandgap that is absorbed by the solar cell and creates an additional electron–hole pair that can contribute to generate current [10]. However, the obtained

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efficiency of the solar cell was small. In addition, the fluorindate glasses have shown to have very low phonon energy and they have been also proposed as suitable matrix doped with Ho^{3+} ions in order to improve the efficiency of silicon solar cells, using upconversion processes [8].

One of the most studied energy transfer processes between rare-earth ions is the one found between Yb^{3+} and Er^{3+} ions that has been used to obtain laser radiation and optical amplification in the 1.54 μm range. This spectral region corresponds to the third low loss telecom window in silica optical fiber [11]. In this kind of system the 975 nm radiation is strongly absorbed by Yb^{3+} ions, which acts as the sensitizer due to its large absorption cross-section. These ions transfer their energy to Er^{3+} ions that relax emitting light at 1.54 μm .

Another characteristic of the Yb^{3+} ions is that they have a simple energy level scheme which one consists in two levels with a gap about $10,000\text{ cm}^{-1}$. Moreover, the maximum energy phonons in these glasses is about 500 cm^{-1} [12] which indicates that there are necessary about 20 phonons to produce nonradiative relaxation. Therefore, no temperature dependence has been found for the intrinsic lifetime of Yb^{3+} ions [12] and a radiative quantum efficiency about 100% is expected. Consequently, the incorporation of Yb^{3+} ions could be used in order to improve the upconversion processes from Er^{3+} ions when they are excited about 1.5 μm . Therefore, in the present work, fluorindate glass materials doped with Er^{3+} and Yb^{3+} ions have been used to improve the solar cell efficiency. The external quantum efficiency and its dependence with the concentration of doping ions were analyzed.

2. Experimental

The samples used in this study were prepared with the following composition in mol %: $(40-x-y)\text{InF}_3$, 20ZnF_2 , 20SrF_2 , 20BaF_2 , $x\text{YbF}_3$ and $y\text{ErF}_3$, with x and y in the range 0–2.25. The details about the preparation of this glass matrix have been reported in a previous work [12]. The samples were excited with a commercial 1480 nm laser (Alcatel 1933 SMG) with a maximum power of 50 mW. The laser was focused on the samples using a lens with a focal length of 30 mm. The waist spot size on the sample (defined as the $1/e^2$ radius of the intensity) was shown to be $235 \times 235\ \mu\text{m}^2$. The emitted light from the top of the samples was recorded using a lens of 50 mm of focal length and focused on the entrance slit of a CCD spectrograph. The sample was located above a commercial solar cell and the photocurrent obtained was recorded by a Keithley picoamperemeter.

The temporal evolution of the emissions was measured by modulating the exciting light with a mechanical chopper and using a computer controlled Tektronix 2432 digital storage oscilloscope.

3. Results and discussion

The upconversion emission spectra of the glass samples codoped with different Er^{3+} concentrations and 0.1 mol% of Yb^{3+} as well as the upconversion emission spectra of the glasses samples codoped with different Yb^{3+} concentrations and 2.25 mol% of Er^{3+} are shown in Fig. 1. All these spectra were obtained under excitation at 1480 nm. The main emission bands due to Er^{3+} electronic transitions are centered at about 545 nm ($^4\text{S}_{3/2} (^2\text{H}_{11/2}) \rightarrow ^4\text{I}_{15/2}$), 660 nm ($^4\text{F}_{9/2} \rightarrow ^4\text{I}_{15/2}$) and 975 nm ($^4\text{I}_{11/2} \rightarrow ^4\text{I}_{15/2}$). Moreover, the emission of the Yb^{3+} ions ($^2\text{F}_{5/2} \rightarrow ^2\text{F}_{7/2}$) is centered at this wavelength as well. As it can be in Fig. 1, decreasing the Yb^{3+} concentration or increasing the Er^{3+} concentration, lead to enhance the upconversion emission intensity.

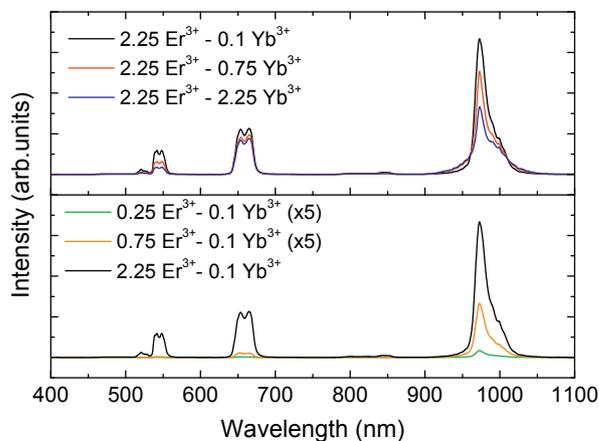


Fig. 1. Upconversion emission spectra of the glass samples codoped with different Er^{3+} and Yb^{3+} concentrations. Note that the upconversion spectra of the glass samples codoped with 0.25 and 0.75 mol% of Er^{3+} and 0.1 mol% of Yb^{3+} were multiplied by a factor 5.

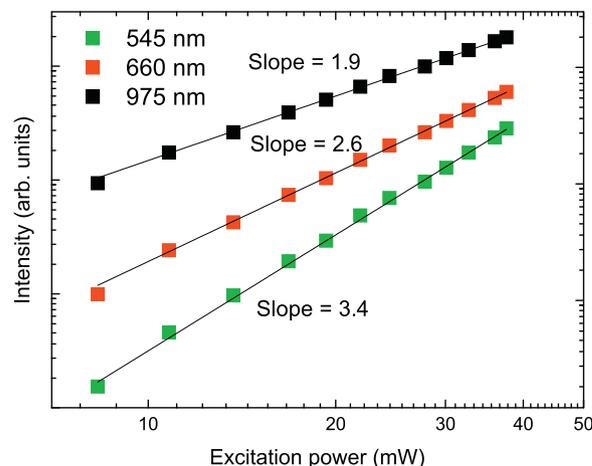


Fig. 2. Upconversion intensities as a function of the pump power at 1480 nm in a double-log scale obtained in a glass sample codoped with 2.25 mol% of Er^{3+} and 0.1 mol% of Yb^{3+} . The best fits to linear dependences give slopes of 3.4, 2.6 and 1.9 for the 545 nm, 660 nm and 975 nm bands, respectively.

Comparing the upconversion emission spectra shown in Fig. 1, it can be seen that the glass sample codoped with 2.25 mol% of Er^{3+} and 0.1 mol% of Yb^{3+} gives the largest upconversion intensity. The 975 nm band in this sample is the most intense in comparison with the other glass samples. This emission is the most interesting for the solar cell applications because corresponds to the Si bandgap, so it is possible to create an electron–hole pair and therefore to generate photocurrent. It is interesting to improve this emission in order to increase the photocurrent compared to the other upconversion bands whose excess of energy over the bandgap is lost as heat in the solar cell. This is the reason why the 975 nm upconverted band is very interesting for the solar cell applications with these doping ions.

3.1. Upconversion mechanism

In order to understand the upconversion process in Er^{3+} – Yb^{3+} codoped glasses, the dependence of the upconversion intensity bands with the pump power excitation has been measured. As it is shown in Fig. 2, the slopes of log–log power dependence of the 545 nm, 660 nm and 975 nm are 3.4, 2.6 and 1.9, respectively. These results indicate that in order to produce these upconversion emissions four, three and two photons processes are required,

respectively. This upconversion mechanism has been studied in other glasses [5,13].

In Fig. 3 the upconversion intensity associated with all the main emission bands has been plotted for the glass samples codoped with different Er³⁺ concentrations and 0.1 mol% of Yb³⁺ as a function of the Er³⁺ concentration. In general, the upconversion processes under stationary excitation depend on the number of involved ions. Therefore, a dependence of the intensity of each band with the concentration is expected as follows [14],

$$I = AC^P \quad (1)$$

where *I* is the intensity recorded by the upconversion emission, *A* is a constant, *C* is the concentration of the active ions and *P* is the power index that gives information about the number of Er³⁺ ions necessary to generate the upconverted emission. The values of *P*, obtained from the fits of the upconverted intensity bands, were 4.4, 3.7 and 2.3 for the 545 nm, 660 nm and 975 nm bands, respectively. Attending to these results, the 975 nm upconversion emission band is based on a process of two absorptions from the Er³⁺ ground state (⁴I_{15/2} → ⁴I_{13/2}) while the 650 and 545 nm emission bands are originated in three Er³⁺ and four Er³⁺ GSA processes, respectively.

The temporal dependence of the upconversion intensities at 545 nm, 660 nm and 975 nm are shown in Fig. 4. The rise time that can be observed in all the curves indicates that the mechanism necessary to produce the upconversion is based in energy transfer processes (ETU) in which an excited ion transfer its excitation energy to a neighbor excited ion, which is promoted to a higher excited state. In other way, when the intensity curve follows to the excitation pump, i.e., shows a negligible rise time, then the upconversion processes is due to an excited state absorption mechanism (ESA) that involves the absorption of a photon by a previous excited ion and promotes to a higher excited state.

Based on the results shown in Figs. 2–4, it can be deduced that the mechanism to produce the upconversion emission is an energy transfer process with different steps. Under direct excitation around 1480 nm, the ⁴I_{13/2} level of Er³⁺ (see Fig. 4) is populated due to ground state absorption, Eq. (2). One of the Er³⁺ ions that has been promoted to the ⁴I_{13/2} metastable level can transfer its energy to a nearby Er³⁺ ion on the same level, Eq. (3), which promotes to the ⁴I_{9/2} level. The Er³⁺ ions that are in the ⁴I_{9/2} excited level can relax non-radiatively to the ⁴I_{11/2} excited level, and eventually decay radiatively to the ⁴I_{15/2} ground state emitting a photon. This explains the emission band found at around 975 nm, mainly due to the Er³⁺ as is shown in Fig. 1. On the other

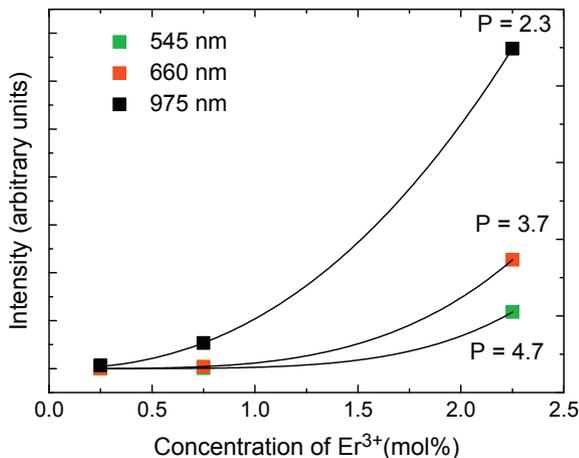


Fig. 3. Intensities of the upconversion emission bands as a function of the Er³⁺ concentration obtained in samples codoped with 0.1 mol% of Yb³⁺ and excited with 37 mW at 1480 nm.

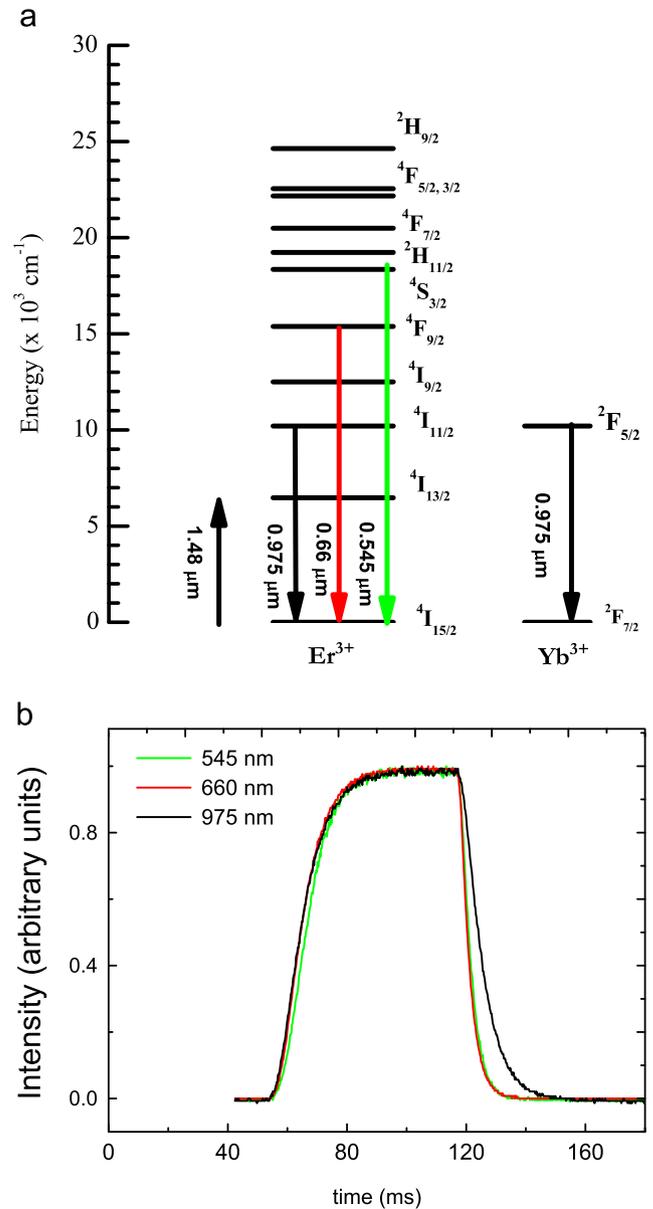
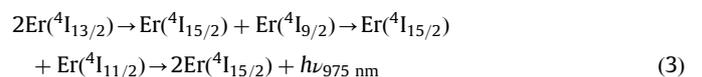
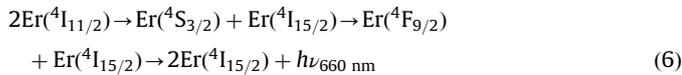
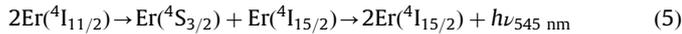
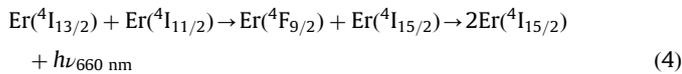


Fig. 4. (a) Energy level diagrams of Er³⁺ and Yb³⁺ ions. (b) temporal evolutions of the upconversion emissions at 545 nm, 660 nm and 975 nm obtained under excitation at 1480 nm in a glass sample codoped with 2.25 mol% of Er³⁺ and 0.1 mol% of Yb³⁺.

hand, one of the Er³⁺ ions that has been promoted to the ⁴I_{11/2} could transfer its energy to a nearby Er³⁺ ion that is in the ⁴I_{13/2} excited metastable level, so that it promotes to ⁴F_{9/2} excited level, Eq. (4), from which it can undergo a radiative relaxation to the ground state obtaining the red emission at 660 nm. One of the Er³⁺ ions in the ⁴I_{11/2} excited level can interact transferring its energy to a nearby Er³⁺ on the same level, so that the Er³⁺ ion promotes to the ⁴S_{3/2} excited level and relax to the ⁴I_{15/2} ground state, giving place to the green emission around at 545 nm, Eq. (5). Moreover, from the ⁴S_{3/2} excited level the ions can relax radiatively or non-radiatively to the ⁴F_{9/2} level, giving place to the red emission around at 660 nm, Eq. (6). Therefore, these processes are given by





These energy transfer processes explain the results obtained in Figs. 2 and 3. Moreover, from the ${}^4\text{I}_{11/2}$ level of the Er^{3+} ions can be excited the Yb^{3+} ions by resonant transfer processes. The emission of these ions is also centered at 975 nm, but as can be seen in Fig. 1, the increase of the concentration of Yb^{3+} ions produce a decrease in the upconversion processes. A possible explanation of this result is based in the migration of the excitation between the Yb^{3+} ions and transfer to traps in the matrix. These migration processes between Yb^{3+} ions have been analyzed in the same matrix using time resolved fluorescence line narrowing experiments [15].

3.2. Application to a solar cell device

The second part of the experiment was to measure the efficiency variation of a photovoltaic device by placing it under different codoped samples. For commercial purposes the samples would be placed under transparent solar cells facing to the infrared irradiation [5]. Therefore, it is expected that under 1480 nm excitation a fraction of the upconverted emission is absorbed by the solar cell. In addition, as was mentioned before, and attending to the upconversion emission spectra (see Fig. 1), the most important emission that could produce photocurrent is the 975 nm band, the energy of which is just above the Si band-gap.

In order to measure the photocurrent, the glass samples were placed on top of the cell, the incident light is perpendicular to the solar cell and the glass sample (see Fig. 5). Note that the samples were not optically coupled with the solar cell, i.e., index matching oil between the glasses and the cell was not used in this study.

The solar cell's photocurrent and the external quantum efficiency for a non-bifacial monocrystalline silicon solar cell with a typical bandgap of 1.1 eV as function of the source excitation

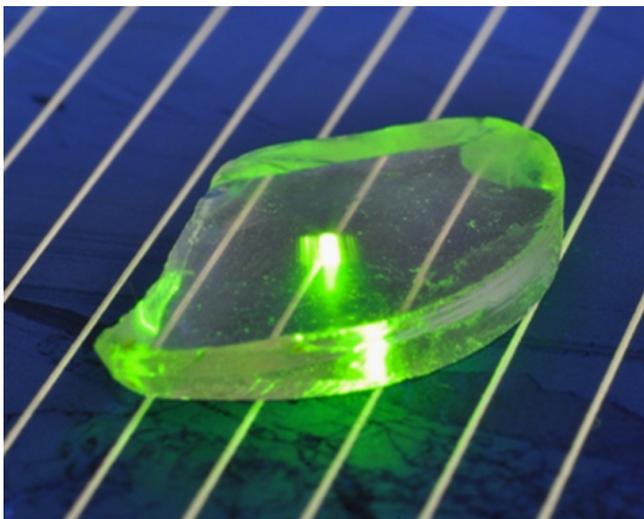


Fig. 5. Photo of a non-bifacial monocrystalline silicon solar cell with a glass sample codoped with Er^{3+} - Yb^{3+} on the top. The excitation is carried out from the top with a laser diode operating at 1480 nm (not visible in the photo). The incident light is perpendicular to the solar cell and the glass.

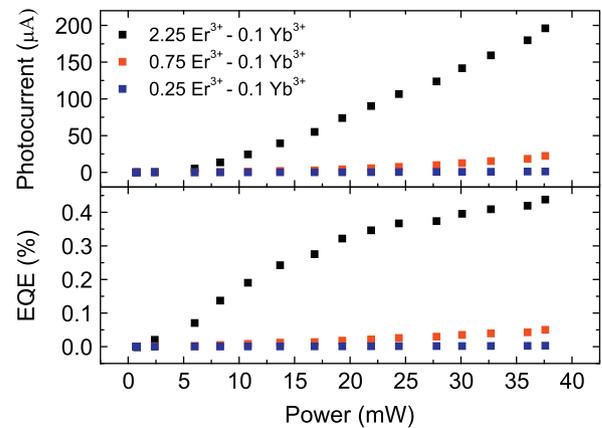


Fig. 6. Solar cell's photocurrent and external quantum efficiency (EQE) as function of the power laser at 1480 nm. The data have been obtained for the glass samples codoped with different Er^{3+} concentrations and 0.1 mol% of Yb^{3+} .

power are shown in Fig. 6. In this figure are only included the results obtained for the sample with lowest Yb^{3+} concentration. As it was shown previously in these samples, the Yb^{3+} concentration decreases the upconversion processes under excitation at 1480 nm. These measurements were made by recording the short-circuit current of the solar cell (with an upconverting glass on the top). As it can be seen in Fig. 6, a photocurrent about 200 μA could be obtained at 37 mW of pump power for the sample codoped with 2.25 mol% of Er^{3+} and 0.1 mol% of Yb^{3+} . Moreover, the results obtained for the other samples are in concordance with the upconversion intensities shown in Fig. 1. Therefore, there is clear evidence between the upconversion intensity and the photocurrent generation. It is interesting to note that when the samples are removed from the top of the solar cell the registered photocurrent was negligible.

The external quantum efficiency (EQE) of a solar cell is defined as the ratio between the number of generated electron-hole pairs due to the upconversion processes and the number of incident infrared photons. The EQE increases with the power of the source and depends strongly with the concentration of optically active ions in the host material. With 37 mW of excitation power at 1480 nm an external quantum efficiency about 0.4% was found for the codoped sample with 2.25 mol% of Er^{3+} and 0.1 mol% of Yb^{3+} . Moreover, the response of the solar cell depends of the Er^{3+} concentration (as can be seen in Fig. 6), but decreases with the Yb^{3+} concentration, being the codoped sample with 2.25% mol of Er^{3+} and 0.1% mol of Yb^{3+} the most efficient.

It can be seen in the literature [5] that the most efficient infrared to visible upconverting material reported to date is a NaYF_4 crystal doped with Er^{3+} ions. The optimal Er^{3+} concentration for upconversion processes under 1520 nm excitation was found to be around 20%. Therefore, an EQE about 0.1% is obtained for a pump excitation equivalent to a solar concentrator of around 1000 suns.

For the most efficient sample of fluoroindate glasses doped with 2.25 mol% of Er^{3+} and 0.1 mol% de Yb^{3+} , is necessary a solar concentrator of 864 suns in order to obtain an EQE about 0.1%. However, this value could be improved increasing the concentration of Er^{3+} in the fluoroindate glasses. As can be seen in Fig. 3, with the maximum power of excitation about 37 mW, the dependence with the Er^{3+} concentration shows a quadratic dependence for the upconversion emission at 975 nm and does not show saturation effect with the Er^{3+} concentration. This quadratic dependence with the Er^{3+} concentration is also observed in the photocurrent and in the EQE values (see Fig. 6). Therefore, this concentration could be increased in order to optimize the EQE. As

example, for an Er^{3+} concentration about 4.5 mol% of Er^{3+} it is expected an increase of a factor 4 for the emission intensity at 975 nm and a solar concentrator under 220 suns is required in order to obtain an EQE enhancement about 0.1%.

The luminescence quantum efficiency (LQE) of the upconversion process is defined as the ratio between the number of upconverted photons and the number of absorbed infrared photons. Therefore, the relation between EQE and LQE is given by [3]:

$$\text{EQE} = (1-L) (1-\exp(-\alpha l)) \text{LQE} \quad (7)$$

where L gives the optical losses, α the absorption coefficient at the excitation wavelength and l is the thickness of the upconversion material. A value of $L=0.75$ has been assumed for the optical losses of the system in similar way to the Refs. [3,5]. The absorption coefficient for the fluoroindate glasses at 1480 nm is 1.14 cm^{-1} and considering a sample with a thickness of 1 cm, from Eq. (7) a relationship of $\text{EQE}=0.17 \times \text{LQE}$ is obtained. Therefore, a value of 2.6% is obtained for LQE under excitation with 37 mW at 1480 nm for a sample doped with 2.25 mol% of Er^{3+} and 0.1 mol% of Yb^{3+} . This value is lower than the value of 12.1% for LQE obtained for an optimized sample of NaYF_4 doped with 20% of Er^{3+} in Ref. [5]. However, as is shown in this reference this value depends so much with the Er^{3+} concentration and for a concentration about 2% (similar to the fluoroindate glasses) the LQE value for NaYF_4 is nearly negligible. This result indicates that increasing the Er^{3+} concentration in the fluoroindate glasses could be improved the value for LQE.

4. Conclusions

The upconversion emission spectra of Er^{3+} - Yb^{3+} codoped fluoroindate glasses have been obtained under infrared excitation at 1480 nm below the Si's bandgap. The upconversion emission spectra are given by visible emission bands around 545 nm and 660 nm (Er^{3+} : $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$, Er^{3+} : $^4\text{F}_{9/2} \rightarrow ^4\text{I}_{15/2}$, respectively) and a near infrared band at 975 nm (Er^{3+} : $^4\text{I}_{11/2} \rightarrow ^4\text{I}_{15/2}$ and Yb^{3+} : $^2\text{F}_{5/2} \rightarrow ^2\text{F}_{7/2}$). This radiation can be absorbed by a Si solar cell and transformed into electron-hole pairs, resulting in an enhancement of the solar cell efficiency. The upconverted intensity at 975 nm decreases when the Yb^{3+} concentration is increased. Measurements of the EQE of a monocrystalline silicon solar cell with a glass sample codoped with 2.25% mol of Er^{3+} and 0.1% mol of Yb^{3+} has an EQE of 0.4% under excitation with 37 mW. An estimation of a concentrator solar with a value about 864 is required in order to reach an EQE of 0.1%. Moreover, the values obtained have a quadratic dependence with the Er^{3+} concentration, therefore it is expected that increasing this concentration the EQE could be improved for this efficient doped matrix. In comparison with NaYF_4 crystals it is expected similar or better results when the

Er^{3+} concentration in fluoroindate glasses is optimized. Moreover, these glass samples have the advantage of their easy preparation respect to the crystals.

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