Photoinduced second-harmonic generation in fibers doped with rare-earth ions

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Photoinduced second-harmonic generation in silica fibers doped with Er$^{3+}$, Sm$^{3+}$, and Tb$^{3+}$ has been investigated. Er$^{3+}$-doped fibers have been found to tune a $\chi^{(2)}$ grating easily to the mode structure of the laser radiation, whereas Sm$^{3+}$-doped fibers have been found to possess the greatest resistance to $\chi^{(2)}$ grating erasure by radiation at 532 nm. From the beginning of the preparation processes, the third harmonic at 355 nm is registered in all the fibers.

Along with Ge-doped silica fibers, the most efficient second-harmonic (SH) generation in media with inversion symmetry has been found in fibers doped with rare-earth ions: Ce$^{3+}$, Nd$^{3+}$, and Eu$^{3+}$. The extension of this list to the other rare-earth ions with well-studied electron structures would promote a better understanding of the general laws underlying this phenomenon and determining the SH efficiency.

In this Letter we deal with the formation of a $\chi^{(2)}$ grating in the case of three other rare-earth ions. We have investigated three aluminosilicate fibers ($7 Al_2O_3/93 SiO_2$) doped with Er$^{3+}$ [1000 parts in 10$^6$ (ppm)], Sm$^{3+}$ (500 ppm), and Tb$^{3+}$ (1000 ppm) and one germanosilicate fiber ($7 GeO_2/93 SiO_2$) doped with Er$^{3+}$ (1000 ppm). The results are compared with those obtained with an undoped germanosilicate fiber ($10 GeO_2/90 SiO_2$). The core diameter varied only slightly from fiber to fiber and was ~8 μm, allowing us to launch ~30% of the laser IR radiation and its SH into the fundamental mode of all the fibers with a 20× objective. A Q-switched and mode-locked Nd:YAG laser (Coherent Antares) was used in the experiments. The in-fiber power density of the IR radiation was 30–50 GW/cm$^2$. The power density of the seed SH obtained with the aid of a nonlinear KTP crystal could be varied over a wide range, depending on the specific experimental conditions.

The experiments showed that all the fibers tested were able to generate photoinduced SH. However, the aluminosilicate fibers doped with Er$^{3+}$, Tb$^{3+}$, and Sm$^{3+}$ required a rather intense seed SH ($I_2 \sim 10^{-2}I_1$) for the saturation level of photoinduced SH to be reached during a preparation time not exceeding 1 h. The SH efficiency achieved with rare-earth-doped aluminosilicate fibers was approximately the same (10$^{-3}$). Under the same preparation conditions the undoped germanosilicate fiber and the one doped with Er$^{3+}$ yielded a 6–8 times greater conversion efficiency at a much lower SH ($I_2 < 10^{-1}I_1$).

Figure 1 shows the distinctions in the saturation rate of four types of fiber measured under the same conditions. In the Er$^{3+}$-doped aluminosilicate fiber the conversion efficiency reaches saturation rather quickly, the saturation rate being only a little lower than that in germanosilicate fibers. In the Tb$^{3+}$-doped fiber the attainment of saturation is much slower. Under the same conditions, one needs a much more powerful (by at least an order of magnitude) seed SH to achieve the maximal efficiency. With the fibers used in the experiments having practically equal concentrations of the rare-earth dopants, these differences in the saturation rate are of a general nature and reflect the differences in the rates of photoionization and phototransfer of charge carriers—the processes responsible for the $\chi^{(2)}$ grating formation.

The experiments also showed that the higher rate of the $\chi^{(2)}$ grating formation in the Er$^{3+}$-doped fiber was accompanied by a higher rate of reformation in

![Figure 1](image_url)
ing growth. A typical growth of photoinduced SH in the fundamental mode occurred during the $y^{(2)}$ grating had, been prepared, an efficient transfer into interesting, in whatever higher-order mode a seed $X^{(2)}$ grating with an efficiency of approximately $10^{-6}$. In-}


gination of the preparation to produce a very weak $x^{(2)}$ grating to the mode structure of the interfering radiation. Curves 1 and 3, initial photoinduced SH in a higher-order mode; curve 2, initial photoinduced SH in the fundamental mode.

\begin{figure}
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\caption{Temporal evolution of photoinduced SH for an Er$^{3+}$-doped germanosilicate fiber (curve 1) and for a germanosilicate fiber without rare-earth-ion doping (curves 2 and 3) when the fibers were irradiated only by the IR radiation $I_1$. A weak photoinduced SH with an efficiency $I_2/I_1 = 4 \times 10^{-6}$ had been previously prepared with a seeding SH radiation. Curves 1 and 3, initial photoinduced SH in a higher-order mode; curve 2, initial photoinduced SH in the fundamental mode.}
\end{figure}

comparison with the other types of fiber. So, for the Er$^{3+}$-doped fiber, with a change in the modal structure of the radiation inside the fiber core at the preparation stage, the $\chi^{(2)}$ grating quickly changed to match the new phase relationships with a minimal loss in the rate of preparation. This effect was most pronounced in the Er$^{3+}$-doped germanosilicate fiber. Its SH efficiency being close to that of the undoped germanosilicate fiber, this fiber allowed us to carry out the preparation without a seed SH, by IR radiation only. A seed SH was used only at the beginning of the preparation to produce a very weak $\chi^{(2)}$ grating with an efficiency of approximately $10^{-6}$. Interestingly, in whatever higher-order mode a seed $\chi^{(2)}$ grating had been prepared, an efficient transfer into the fundamental mode occurred during the $\chi^{(2)}$ grating growth. A typical growth of photoinduced SH in this fiber, when the phase relationships were broken at the preparation stage, is shown in Fig. 2. It is also seen that under the same conditions such preparation of the undoped germanosilicate fiber was practically impossible, even though a weak $\chi^{(2)}$ grating was in the fundamental mode.

It should be noted that such mobility of the $\chi^{(2)}$ grating in the erbium fibers was not accompanied by high erasability under the action of an intense SH radiation.

Figure 3 shows the rate of the photoinduced SH decrease in different types of fiber after the fibers were irradiated with an intense SH at 532 nm. All the rare-earth-doped fibers showed a greater resistance of the $\chi^{(2)}$ grating to erasure by intense SH radiation than the undoped germanosilicate fiber. Note that in the case of erbium the rates of erasure are practically the same for both the germanosilicate and the aluminosilicate fibers. This means that the Er$^{3+}$ doping of a germanosilicate fiber makes the erasure effect even weaker. The highest resistance to erasure was demonstrated by the Sm$^{3+}$-doped aluminosilicate fiber: Four times more intense SH was needed to erase a $\chi^{(2)}$ grating in this fiber at approximately the same rate as that for the undoped germanosilicate fiber.

The behavior of the $\chi^{(2)}$ grating formation in rare-earth-doped fibers can be explained, if one assumes that it is rare-earth ions that play the key part in the photoinization and phototransfer of charges, creating an ordered charge structure under the action of interfering light fields. As described for $\gamma$-irradiated fluorite crystal doped with rare-earth ions, three-valent rare-earth ions can act as ionized centers and as centers capturing photoelectrons, depending on the local environment. Thus the high rate of the efficiency saturation in Er$^{3+}$-doped fibers should first be attributed to the high rate of photoionization of three-valent erbium ions acting as donor centers. This in turn is associated with the presence of resonance energy levels for all the radiations participating in the $\chi^{(2)}$ grating writing, in particular, Nd:YAG laser radiation and its SH. Tb$^{3+}$-doped fibers that do not have such resonance levels demonstrate the lowest rate of $\chi^{(2)}$ grating preparation.

The great ability of the Er$^{3+}$-doped fibers to tune a $\chi^{(2)}$ grating to the mode structure of the interfering fields testifies to an efficient phototransfer of charge carriers in this type of fiber. If we assume that in the periodic system of charges produced as the result of the preparation process the less-stable ions are centers capturing photoelectrons (the role of such centers can be played by two-valent rare-earth ions), then such photomobility of erbium centers correlates with the known data on temperature stability of these two-valent ions: two-valent erbium ions were found to be among the less stable of all rare-earth ions.

Note that such mobility of erbium radiation centers does not lead to an increased rate of erasability by SH radiation. We think that this may be explained by the presence of a strong absorption band at 532 nm (100 dB/m) that is due to three-valent erbium ions. This absorption is too strong to promote multiphoton ionization with the participation of reso-
nance SH quanta, because it suppresses this process through reabsorption of the SH radiation into the phonon spectral region.

At the same time, for the Tb$^{3+}$-doped fiber, which is similar to the Er$^{2+}$-doped fiber in the properties of its two-valent ions but does not have strong resonance levels at the SH frequency; a higher rate of erasure is observed in comparison with the erbium fiber. The higher erasure resistance of the radiation color centers in the Sm$^{3+}$-doped fiber may testify to the fact that phototransfer with the participation of Sm$^{3+}$ ions in this case proceeds inefficiently. This fact correlates with the data on temperature stability of two-valent rare-earth ions: Sm$^{2+}$ ions are among the most stable of all rare-earth ions.

To gain more penetrating insight into the processes of photoionization and charge formation in rare-earth-doped fibers, we must take into account all the radiation involved: external radiation and that arising inside a fiber as a result of nonlinear phenomena. It should be stressed that in all the fibers tested radiation of the third harmonic of the Nd:YAG laser at 355 nm was observed at the fiber output from the beginning of the preparation. Its intensity was proportional to the cubed intensity of the IR radiation, and under the same conditions it depended weakly on the fiber type. Normally it did not exceed $10^{-7}$ of the IR radiation intensity. It is worth mentioning a study in which the third harmonic was also revealed during the preparation of a $\chi^{(3)}$ grating in germanosilicate fibers. However, a detailed analysis of the influence of the third-harmonic UV radiation arising inside a fiber on the mechanism of the $\chi^{(3)}$ grating formation was not made. Clearly this UV radiation of even relatively small intensity can make a major contribution to photoionization of the dopant atoms and to phototransfer of charge carriers.

We are planning further investigations of the effect of the UV radiation arising inside a fiber as a result of cubic nonlinear process on the $\chi^{(3)}$ grating formation.

In summary, we give our major findings: SH generation with an efficiency of $10^{-3}$ was obtained in aluminosilicate fibers doped with Er$^{3+}$, Sm$^{3+}$, and Tb$^{3+}$ ions. In germanosilicate fibers doped with Er$^{3+}$ ions the efficiency was $8 \times 10^{-3}$. In Er$^{3+}$-doped fibers we observed a high rate of photoionization and phototransfer of charge carriers forming a $\chi^{(3)}$ grating. This allowed us to reduce the level of the seed SH and to tune a $\chi^{(3)}$ grating easily to changes of the mode structure of the laser radiation. Sm$^{3+}$-doped aluminosilicate fibers featured a high resistance of the $\chi^{(3)}$ grating to erasure by 532-nm radiation. In all the fibers, UV radiation of the third harmonic existed from the beginning of the SH preparation.

References