Infrared-to-visible upconversion in silica and fluorozirconate (ZBLAN) based erbium doped optical fibers. Er³⁺:ZBLAN optical fiber upconversion laser

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Infrared-to-visible upconversion has been observed by many authors in a variety of rare-earth doped crystals, glasses and optical fibers (see, *e.g.*, review paper of JOUBERT [1]). We report on the studies of this process in erbium doped silica and fluoroziconate (ZBLAN) optical fibers. Fluorescence quenching in silica fibers appeared to be so strong that the efficiency for upconversion is very low. On the other hand, erbium doped fluorozirconate fiber exhibits extraordinary strong fluorescence in the "green". We report on a novel optical resonator configuration for an upconversion laser, pumped at 805 nm by a laser diode. The upconversion laser operated at 544 nm in a single mode. We came to the conclusion that the excited state absorption process is responsible for the upconversion observed.

1. Introduction

As early as in 1969 JOHNSON and GUGENHEIM [2] and VAN UITERT et al. [3] observed the infrared-to-visible upconversion in rare-earth compounds. Infrared-pumped visible lasers operating at liquid nitrogen temperatures in Yb³⁺ or Er³⁺ doped crystals were demonstrated by JOHNSON and GUGENHEIM [4] and SILVERS-MITH et al. [5]. A lot of fundamental as well as technical work on optical fibers and upconversion has been performed by the Payne's and Hanna's groups at the Optoelectronics Research Centre, University of Southampton. Infrared-to-visible upconversion is especially very effective in erbium-doped optical fibers, where the pumping power density easily reaches a value of about 1 MW/cm^2 , and the pumping length in the active medium can be of a meter or longer. We are going to restrict our paper to optical fibers operating only at room temperatures. Of our special interest will be the CW room temperature green upconversion fiber laser pumped by a laser diode at 805 nm. WHITLEY et al. [6], ALLAIN et al. [7], and PIEHLER and CRAVEN [8] were the first to build upconversion lasers operating in erbium-doped optical ZBLAN fibers. We have studied the infrared-to-visible upconversion in silica and fluorozirconate based erbium doped optical fibers. A novel configuration of the optical resonator has been proposed. Fluorescence studies and the spectral charac-



Fig. 1. Absorption band below 1 µm of an erbium doped silica optical fiber [9].

teristic of the emitted laser beam were measured. The upconversion fluorescence observed is due to the mechanism of excited state absorption (ESA), sometimes denoted as excited state upconversion (ESU).

2. Pumping of the fibers

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We have pumped the optical fibers using a diode laser emitting at 805 nm, and in some experiments, also at 970 nm. The core diameters of the ZBLAN fibers were: fiber No. 1, $\Phi_{core} = 1.7 \ \mu m$, $\Phi_{cladding} = 125 \ \mu m$, doping 1000 ppm of Er^{3+} , numerical aperture NA = 0.39; Fiber No. 2, $\Phi_{core} = 1.9 \ \mu m$, $\Phi_{cladding} = 125 \ \mu m$, doping 580 ppm of Er^{3+} , NA = 0.4. A very small entrance face of the core causes a lot of difficulties in the pumping experiments. For this reason, we have used a single spatial mode diode laser that emits up to 150 mW at 850 nm from an active area of

1 μ m × 3 μ m. There is also a severe problem arising from the fact that the diode emission is very divergent (9° – parallel to junction, and 30° – perpendicular to junction). In this way, one cannot focus the whole emitted power. The main absorption bands (Fig. 1) below 1 μ m for the erbium doped fibers are: 800 nm and 970 nm (center wavelengths). Since heavy duty diodes emitting at 800 nm are not available, we have applied a diode emitting at 811 nm (at 25 °C), and then, by cooling the diode to 5 °C, the wavelength decreased to 805 nm, but not to the center of the absorption band. Nevertheless, it was sufficient to reach the threshold for upconversion laser to operate. In the case of silica based erbium fibers, the diameter of the core was of the order of several tens of micrometers, thus, there was no problem to pump these fibers with laser diodes having larger emitting areas (3 μ m × 100 μ m, $\lambda \simeq 800$ nm or 970 nm).

3. Erbium doped silica optical fibers

The fibers were produced at the Laboratory for Optical Fibers Technology, University of Maria Curie-Skłodowska, Lublin [9]. Erbium doping was in the range from about 500 ppm to several thousand ppm. We have found that the optimum doping for upconversion process (in the green) to occur is the value of 1300 ppm. However, the green fluorescence signal observed was approximately 100 times weaker as compared to an equivalent ZBLAN: Er^{3+} fiber. The reason for this is the extremely strong quenching of the fluorescent level, ${}^{4}S_{3/2}$. We have measured the lifetime of this level, and it appeared to be as short as about 0.5 µs [10], contrary to ZBLAN glass, for which this lifetime is in the range of several hundred µs, depending on the erbium concentration. The conclusion is: no upconversion laser in the green can be built using silica based erbium doped fibers, pumped by infrared laser sources. On the other hand, erbium doped silica fibers are very suitable as active media to build lasers and amplifiers operating in all three optical windows (~1 µm, ~1.3 µm and 1.5 µm).

4. Origin of the upconverted fluorescence

Three different processes are generally responsible for the infrared-to-visible upconversion to occur. Photon avalanche observed in rather heavily doped erbium materials is charecterized by an abrupt increase of the fluorescent signal, as the pumping overcomes a characteristic threshold. We did not observe photon avalanche. The other two processes are: ion-pair cooperation (or energy transfer upconversion, ETU) and excited state absorption (ESA). In the case of ion-pair cooperative excitation, absorption and emission do not take place with the same center. Energy transfer in such systems occurs without charge transport through Coulomb interaction, *e.g.*, of dipole-dipole type. As a result, the excitation energy migrates from one ion to its neighbour before it is able to radiate a quantum of energy [11]. Figure 2 explains these processes. In the case of ion-pair cooperative excitation, a characteristic delay between the excitation pulse and the peak intensity



Fig. 2. Ion-pair cooperative excitation (or ETU) -a, and ESA -b.

of the upconverted fluorescence is observed. The reason for this is the relatively large lifetime of the intermediate level (of the order of about 10 ms). In this way, exchange of energy between the neighbouring ions can take place with no excitation pulse still present. We have observed this mechanism in erbium doped ZBLAN glass bulk samples that were heavily (a few percent) doped with erbium (Fig. 3). The delay between the nanosecond pumping pulse and the fluorescence peak amounts to about 6 μ s. On the contrary, excited state excitation, *i.e.*, transition from (2) to (3) is caused by the absorption of infrared photons from the excitation pulse, thus, disappears when the excitation pulse is being cut off. Studying the time display of the upconverted fluorescence with respect to the pumping pulse, one can determine the dominant mechanism of excitation.







Fig. 3. Time display of the upconversion fluorescence in the green in an erbium doped ZBLAN glass bulk sample when pumped with a nanosecond pulse (at 800 nm) recorded at millisecond (a) and microsecond time base (b).

5. Unconverted fluorescence

When pumping the ZBLAN: Er^{3+} optical fibers, several fluorescence bands have been observed, namely in the violet, in the blue, in the green (the most intense one), in the orange and in the red. We have used a grating spectrograph, a CCD camera and an optical multichannel analyzer (Fig. 4) to analyze the spectrum. Laser operation



Fig. 4. Setup for measuring the fluorescence spectrum or laser emission.



Fig. 5. Upconversion fluorescence in the green as a function of power of the pumping beam, measured for an erbium doped (580 ppm) ZBLAN fiber.

can be achieved only in the strongest "green" band. The upconverted fluorescence in the green as a function of power of the pumping beam is illustrated in Fig. 5. The total width of the strongest band in the green is about 25 nm, which is very characteristic of erbium ions in a glass matrix. This is the main reason why no laser operation has been observed in bulk glasses (even in ZBLAN bulk media) doped with erbium.

6. Optical resonator

When constructing an optical resonator for an upconversion laser, several precautions have to be considered. First of all, the fiber terminals have to be optically flat and strictly perpendicular to the fiber axis. To this end, we have used diamond



Fig. 6. Illustration of the case where there is almost no divergence of the radiation within an optical resonator with bulk active medium (a), and, on the contrary, outside mirrors couple back into the fiber only a small fraction of the radiation (b).

cutting equipment or, occasionally, both fiber terminals were optically polished with the help of a glass holder. In the case of a bulk laser active medium, the mirrors can be placed apart from the medium faces, because divergence of the beam is negligible (Fig. 6a). With a fiber active medium, the outgoing emission cone is highly divergent (Fig. 6b), thus, the fiber terminals must be butt coupled to the mirrors or multilayer dielectric mirrors have to be evaporated directly onto the fiber terminals. Figure 7



Fig. 7. Graphical illustration of the divergence for a core diameter of 2 μ m, and a distance between the fiber and the mirror equal to 100 μ m.



Fig. 8. Ratio D/D' as a function of the distance between the fiber terminal and the mirror.

illustrates this situation for a fiber of diameter D (2 µm), and for the distance between the mirror and the terminal of the fiber equal to 100 µm (typical divergence, $\theta = 23$ °C). The ratio D/D' describes the coupling factor (Fig. 8). We have proposed



Fig. 9. Novel configuration of the optical cavity for an upconversion erbium doped fiber laser.

a novel configuration of the optical cavity (Fig. 9). The ZBLAN fiber is extremely fragile, so any mechanical pressing to make the contact between the fiber and the mirror must be avoided. On the other hand, this contact is very critical, and no laser operation can be obtained when there is no optical adjacency. In our vertical configuration, the fibers stick to the mirrors due to gravity. To ensure very stable clamping of the fiber, special glass funnels were drawn out from a capillary tube, with entrance holes of about 2 mm, and exit holes of 120 μ m, *i.e.*, exactly equal to the diameter of the fiber cladding.

The optical resonator has been formed using two dichroic dielectric mirrors. The first one transmits totally the infrared pumping beam and reflects the green upconverted fluorescence. The output mirror (with transmission of the order of a few up to about 30% for the green emission) totally reflects the pumping power that was not absorbed along the fiber length.

7. Laser threshold

In order to reach the laser threshold one has to increase the pumping power density up to about 1 MW/cm². In the case of CW operation, the dichroic resonator mirrors must have a damage threshold of several MW/cm². During our experiments, several tens of dielectric mirrors were burnt because of these power levels. The mirrors we have finally used were prepared at the Institute of Optoelectronics, Technical Military Academy in Warsaw. With our focusing lenses placed directly at the laser diode, the effective power launched into the fiber did not exceed 65 mW, i.e., it was almost half of the total output power of the laser diode. One has to remember that the pumping beam entering the fiber has a diameter of about 2 µm. The effective power launched into the fiber was measured at the end of a short piece of the fiber cut off from the main fiber used. For a typical ZBLAN erbium doped fiber (1000 ppm) of 1 meter length, core diameter of 1.7 µm, and output mirror transmission of about 10%, the threshold power density measured (pumping beam wavelength 805 nm) was 1.3 MW/cm². It is interesting to note that with available pumping power density of about 3 MW/cm², no laser operation could be observed in ZBLAN erbium doped fibers of core diameters of 3.5 µm. Upconverted fluorescence below the threshold and above the threshold for laser operation is shown in Fig. 10. Once the laser starts, the emission abruptly narrows to almost a single line. To determine the full width of the laser emission, a Fabry-Perot interferometer with a free spectral range of 0.5 cm^{-1} was used. A single mode operation with a spectral width of less than 0.25 cm^{-1} was usually observed, depending upon the power launched into the fiber. The output beam has to be collimated with a microscopic objective (see Fig. 9). Any even micrometer motion of the fiber (or mirror holder)



Fig. 10. Fluorescence in the green and laser emission above the threshold.

causes the laser emission to cease. An upconversion laser of practical usefulness must be built in a special way, to ensure extraordinary stability of all its elements. We did not optimize the output emission of the upconversion laser. Two photographs (Figs. 11 and 12) present the laboratory model built, and the upconversion laser at work, respectively.

8. Conclusions

We have shown that silica based erbium doped fibers, suitable to generate or amplify radiation in all three optical windows, cannot be used for upconversion. On the other hand, erbium doped ZBLAN optical fiber, although its upconverted emission band width is very broad, does amplify this emission to a level required for an upconversion laser. However, the pumping power density must be of the order of a few MW/cm^2 , and the length of the fiber not less than a few tens of centimeter. The critical points of operation are extremely high stability of the whole device, and perfect butt coupling of the fiber faces to the mirrors. The mirrors have to keep their parameters when irradiated by power densities (in CW regime) up to several MW/cm^2 .

Within erbium concentration in the ZBLAN fiber from 580 to 1000 ppm we did not find any delay between the pumping pulse and the upconverted fluorescence. This may indicate that the excited state absorption is the dominant excitation process responsible for the upconversion fluorescence observed. However, a contribution from the ion-pair cooperative excitation cannot be excluded. Ion-pair cooperative excitation strongly depends upon the ions separation within the fiber. Formation of ion clusters [12] may reduce the interaction time between them as compared to the interaction time for homogeneously distributed ions. As a result, the time display of the upconverted fluorescence may be similar to that characteristic of excited state absorption. A parasitic process that may quench the upconverted fluorescence, namely the generation of colour centres, must also be taken into account [13]. In conclusion, a series of various further experiments have to be performed to clarify all the mechanisms responsible for infrared-to-visible upconversion, and, of course, to understand the operation of an upconversion fiber laser.

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Fig. 11. Upconversion fiber laser setup.



Fig. 12. Upconversion erbium doped fiber laser. The laser emission, collimated by a microscopic objective $(10 \times)$ is sent horizontally by a 45° mirror located in the ouput mirror tube.

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