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Frequency upconversion in Tm- and Yb:Tm-doped silica fibers

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Frequency upconversion has been observed and studied in Tm^{3+}-doped and Yb^{3+}-sensitized Tm^{3+}-doped silica fibers. In the singly singly doped fiber upconversion to the blue and UV has been observed under excitation in the red (660 nm) and infra-red (1.064 μm). In the co-doped fiber upconversion has also been observed under excitation at around 800–900 nm.

1. Introduction

There has recently been great interest in the study of materials capable of converting infrared radiation to shorter wavelengths by means of sequential multiphoton excitation. This process, termed frequency upconversion and first studied by Auzel [1], is of practical interest as it provides an alternative method to intracavity frequency doubling for short wavelength generation from infrared sources such as AlGaAs laser diodes. The first diode pumped upconversion laser has recently been demonstrated by Tong et al. [2] and operated in the green at 551 nm when pumped with a diode array at 791 nm. The laser medium in that case was an Er^{3+}:YLF crystal which was cooled to 40 K.

Owing to the multiphoton nature of the processes involved in upconversion, high pump intensities are required for efficient excitation. Optical fibers having the core doped with the ion of interest offer a particularly attractive approach as even for quite modest powers, high pump intensities can be achieved and maintained over long lengths. Fused silica fibers have the advantage of well developed fabrication procedures for low-loss monomode fibers, although they have the disadvantage compared to fluorozirconate fibers that many of the excited levels of the impurity ions have lifetimes which are considerably shorter than their radiative lifetimes.

In this work we describe observations of frequency upconversion in Tm^{3+}-doped and Yb^{3+}-sensitized Tm^{3+}-doped silica fibers. The Tm^{3+} ions was chosen for this investigation as it is the only rare-earth ion which has a long lived blue-emitting level when incorporated into a silica host. A blue laser is potentially very attractive as it may find uses in fields as diverse as submarine communications and optical data storage. A Tm^{3+}:YLF upconversion laser operating at 450 nm has been demonstrated by Nguyen et al. [3]. Further confirmation of the potential importance of upconversion schemes in Tm^{3+} is provided by the very recent observation of upconversion lasing in the blue in Tm^{3+}-doped fluorozirconate fiber [4].

2. Experimental methods

The Tm^{3+}-doped fiber used in this experiment was fabricated by the solution doping technique [5] to give a Tm^{3+} concentration of ≈800 ppm. The fiber was further characterized by a numerical aperture of 0.15, cut-off wavelength for monomode operation of 1.7 μm and core diameter 9 μm. The behaviour of this fiber operating as a laser around 1.9 μm has been described elsewhere [6,7]. The Yb^{3+}-sensitized fiber (also fabricated by the solution doping method)

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had an Yb^{3+} concentration of 15000 ppm and a Tm^{3+} concentration of 1500 ppm. For this fiber the numerical aperture was 0.12, the cut-off wavelength 1.0 μm and core diameter 6.4 μm.

Absorption spectra for both fibers were obtained by a multiple cut-back technique. Emission spectra were obtained by exciting the fibers with light from either an argon ion laser, a DCM dye laser operating at around 660 nm, a Styryl 9M dye laser operating between ≈800 nm and ≈900 nm or a Nd:YAG laser operating at 1.064 μm. Light was launched into the fiber by means of a 10× microscope objective to give a launch efficiency of typically ≈50%. The experimental arrangement used to measure the "side-light" spectra was similar to that described by Percival et al. [8]. Unguided fluorescence escaping radially from the core of the fiber is collected by a slit-to-slit fiber optic bundle and dispersed by a 0.25 m grating monochromator. The light was detected by a Thorn EMI model 9658A photomultiplier with extended IR response. The signal from the PMT was passed to a lock-in amplifier, the reference being taken from a variable speed mechanical chopper which was placed in the excitation beam. It should be noted that no correction has been made to the emission spectra for the response of the bundle, monochromator or detector. The advantage of studying the sidelight spectrum as opposed to the spectrum of light which has been guided down the core of the fiber is that the spectrum remains undistorted by ground state reabsorption as the sample is optically thin even at the peak of absorption bands. Measurements of fluorescence lifetimes were made by observing the signal decay on a digital storage oscilloscope after the interruption of the pump beam by a mechanical chopper. The response of the system, found by observing the fall time of the pump signal, was measured to be approximately 10–20 μs.

3. Experimental results

3.1. Tm^{3+}-doped fiber

(i) *Ar^+-laser pumping.* The absorption spectrum of the Tm^{3+}-doped fiber in the 8000–23000 cm^{-1} range is shown in fig. 1 matched to the Tm^{3+} energy level diagram. Fig. 2 shows the spectrum of emission

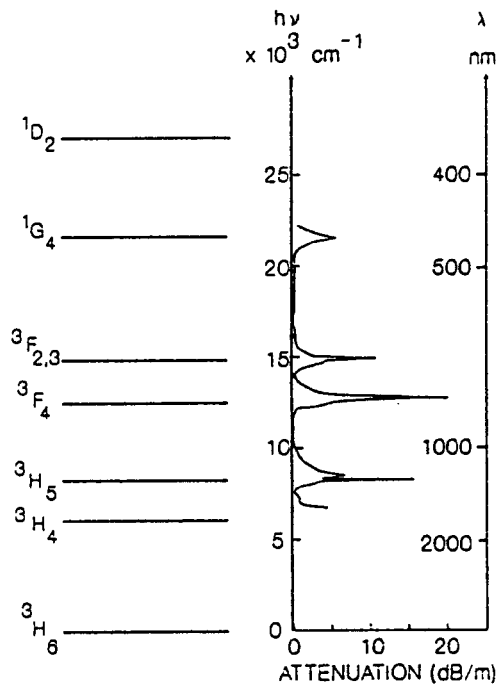

Fig. 1. Energy Levels and absorption spectrum for a silica fiber doped with 800 ppm Tm^{3+} ions.

from this fiber in the 300–850 nm range under excitation directly into the $^1G_4$ multiplet using an Ar^+ ion laser operating at 457.9 nm. The dominant blue emission band peaked at 467 nm corresponds to decay from the $^1G_4$ level to upper Stark levels of the ground multiplet. The red emission centered at 650 nm and the emission at ≈750 nm are also thought to originate from the $^1G_4$ level, terminating on the $^3H_4$ and $^3H_5$ multiplets respectively. All three emission bands exhibit an approximately exponential time decay with the same 1/e decay time, measured to be ≈220 μs. The absorption data of fig. 1 has been used to calculate a value of ≈860 μs for the $^1G_4$ to $^3H_6$ radiative lifetime, suggesting that the quantum efficiency for blue emission is around 25%.

The weak upconverted fluorescence at ≈370 nm is believed to correspond to decay from the $^1D_2$ level to the $^3H_6$ ground multiplet. This fluorescence was found to have a non-exponential time decay with a 90%–10% fall time of ≈80 μs. This level is probably populated by excited state absorption of blue photons from the $^3H_4$ level. The $^3H_4$ multiplet is the metastable level responsible for 1.8 μm laser emis-

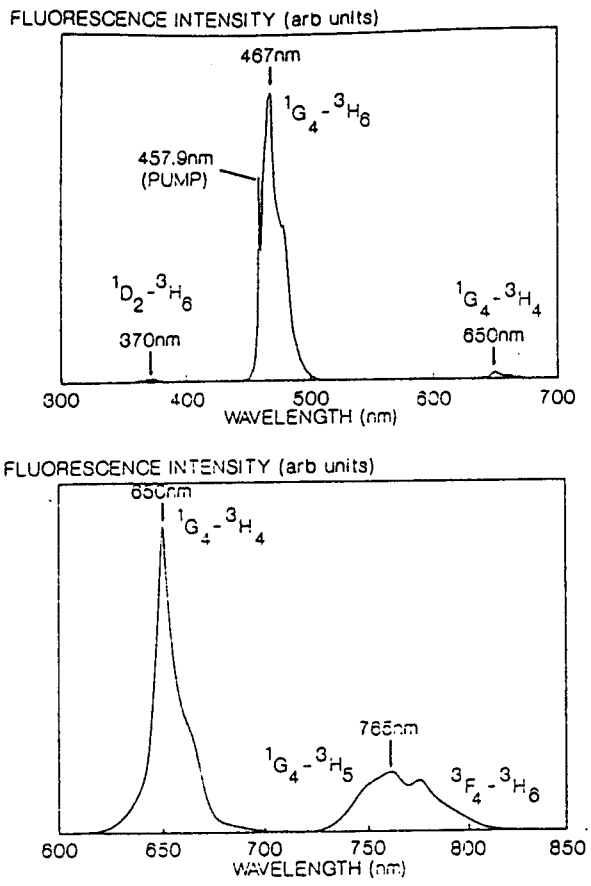

Fig. 2. Emission spectrum for Tm^{3+}-doped silica fiber under excitation at 457.9 nm: (a) from 300–700 nm. (b) from 600–850 nm (note change in scale between the two spectra).

sion [6] with a lifetime of ≈220 μs. Finally, the spectrum in fig. 2b also contains a contribution around 810 nm from the $^3F_4$–$^3H_6$ transition. The lifetime of the $^3F_4$ level has been measured in this same fiber under direct pumping with a Styryl 9M dye laser at 797 nm and found to be ≈10 μs [6].

(ii) *DCM laser pumping.* Upconversion to the blues and UV has been studied using a DCM dye laser operating in the red at 660 nm as the pump source. This pump wavelength corresponds to absorption from the ground state to the $^3F_2$ and $^3F_3$ excited levels (which overlap in a silica host). Upconverted fluorescence is observed in both the blue and UV. The spectrum is shown in fig. 3. Both the blue emission centered at 460 nm and the UV emission at 370 nm originates from the $^1D_2$ level with the lower levels being $^3H_4$ and $^3H_6$ respectively. It is thought that

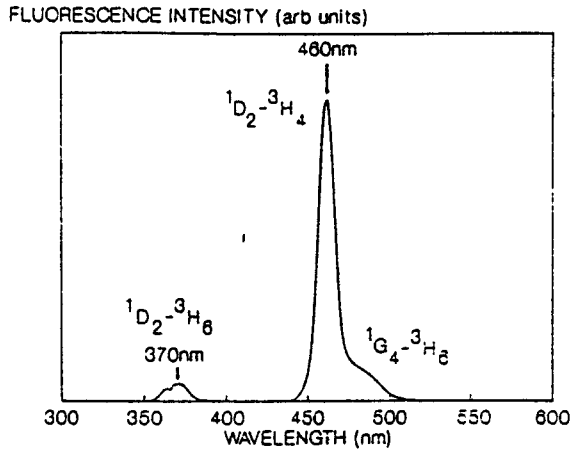

Fig. 3. Emission spectrum for Tm^{3+}-doped silica fiber under excitation at 660 nm.

the shoulder on the long wavelength side of the blue emission is due to decay from the $^1G_4$ level to the $^3H_6$ ground state.

The number n of pump photons required per upconverted photon emitted can easily be determined from the dependence of upconverted emission intensity on the excitation intensity. From a simple rate equation model it can be deduced that the upconverted fluorescence intensity I_u will be proportional to the power n of the excitation intensity I_e; thus, $I_u \propto I_e^n$. It should be noted, however, that such a model takes no account of phonon-assisted processes, or of depletion of the ground and intermediate state populations, so that in general a non-integral value of n can be expected and the curve may also saturate. The dependence of the peak blue fluorescence intensity (as measured by a photomultiplier) on excitation intensity is shown in fig. 4. The quadratic dependence observed over the available pump power range is consistent with the emitting level being populated via a two-photon process. The proposed mechanism for this scheme is shown in fig. 5 and is the same as that observed by Esterowitz et al. for Tm^{3+}-doped fluoride crystals [9]. A red photon causes excitation to the $^3F_2$ and $^3F_3$ levels. Rapid non-radiative decay then takes place to the $^3F_4$ level because of the small energy gap and high phonon energies in silica. A further red photon is then absorbed resulting in population of the $^1D_2$ level. A problem with a silica host is that significant non-ra-

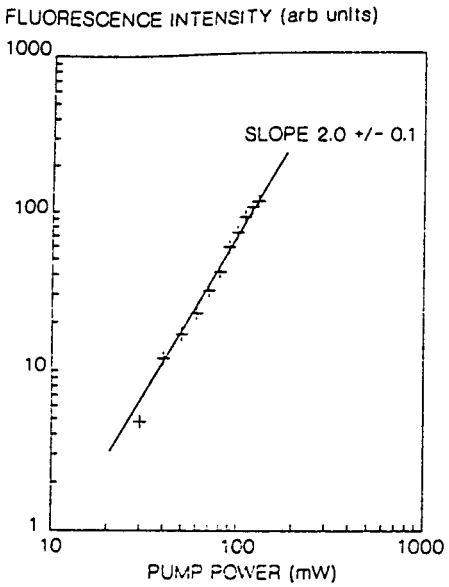

Fig. 4. Dependence of 460 nm emission intensity on 660 nm excitation power.

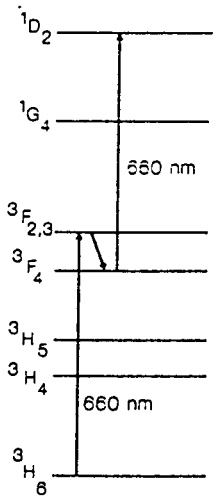

Fig. 5. Mechanism for population of $^1D_2$ level under excitation at 660 nm.

diative decay occurs from the $^3F_4$ level (a consequence of the small energy gap between this level and the $^3H_5$ level and high phonon energy of silica) which means that the lifetime of this level is significantly shorter than that observed in, for example, a fluorozirconate host with a corresponding reduction in the upconversion efficiency.

(iii) *Nd:YAG laser pumping.* Upconversion has also been observed under excitation from a continuous-wave Nd:YAG laser at 1.064 μm. The photons from this pump source fall in the high energy tail of the $^3H_5$ absorption band where the absorption is relatively weak. Despite this relatively weak absorption, upconversion to the blues is readily observed as the pump source has a high power and there exist absorbing transitions form excited states which are near resonant for the 1.064 μm light. The resulting upconversion emission spectrum under excitation with 1.6 W of pump power is shown in fig. 6. The relative intensities of these spectral peaks are pump power dependent; at lower pump powers the 790 nm emission is enhanced and the 370 nm emission reduced.

The power dependence of the blue emission at 467 nm and the UV at 365 nm was then investigated and is shown in fig. 7. It can be seen that the blue emission has an approximately cubic dependence on excitation intensity for pump powers up to 200 mW consistent with population of the emitting level via a three-photon process. For higher pump powers the blue emission begins to saturate. The lifetime of the emission was measured to be 250 μs identifying the initial level as $^1G_4$. Thus the blue emission is predominantly due to decay from the $^1G_4$ level to the ground state, although a contribution at the short wavelength side may be due to decay from the $^1D_2$ level to the $^3H_4$ level. The proposed mechanism for the population of the $^1G_4$ levels is shown in fig. 8.

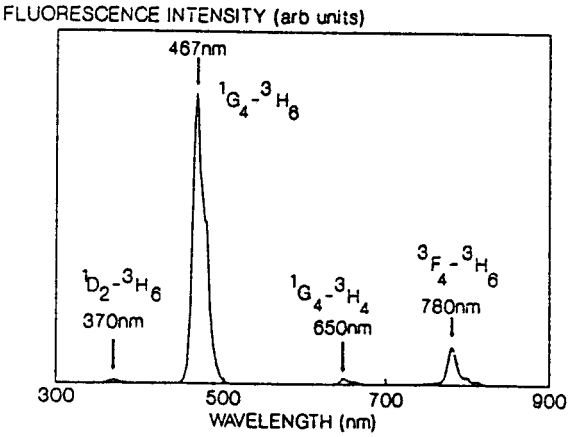

Fig. 6. Emission spectrum for Tm^{3+}-doped silica fiber under excitation at 1.064 μm.

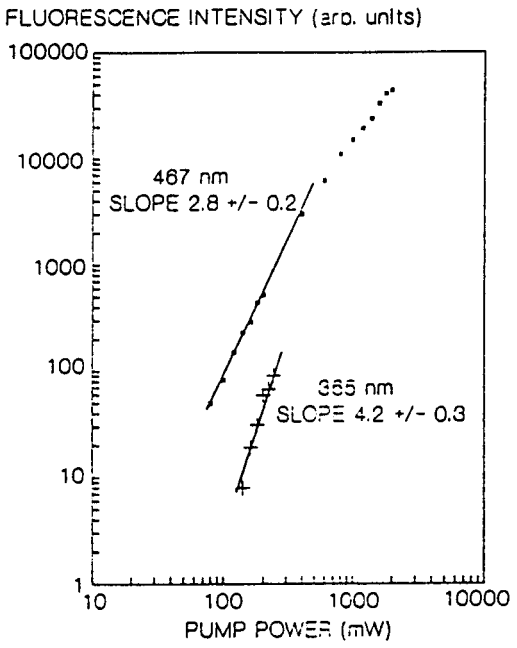

Fig. 7. Dependence of 467 nm and 365 nm emission intensity on 1.064 μm excitation power.

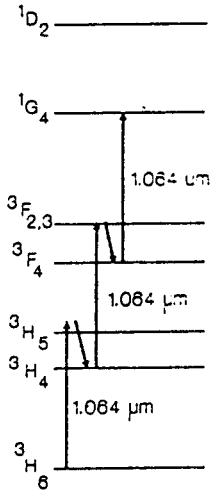

Fig. 8. Mechanism for population of $^1G_4$ level under excitation at 1.064 μm.

Absorption of a pump photon causes excitation to the $^3H_5$ level. Rapid non-radiative decay then causes population to build up in the $^3H_4$ level. Another pump photon is then absorbed causing excitation to the $^3F_2$ and $^3F_3$ levels. Non-radiative decay then takes place to the $^3F_4$ level with the further absorption of a pump photon giving rise to population of the $^1G_4$ level.

For pump powers below 200 mW the UV emission (from $^1D_2$) has an approximately quartic dependence on pump power which implies population of the emitting level through a four-photon process. For higher excitation powers a cubic dependence is observed presumably resulting form the depletion of the ground state. The mechanism for population of the $^1D_2$ level is not certain although population of this level may take place through the absorption of a fourth pump photon from the $^1G_4$ level. This may be inefficient because the energy difference between the $^1G_4$ and $^1D_2$ level is considerably less than that provided by a 1.064 μm photon. A different route to population of the $^1D_2$ level is by energy transfer from one Tm^{3+} ion in the $^1G_4$ level, to another in the $^3H_4$ level. Alternatively absorption of a blue photon emitted from $^1G_4$ by an ion in the $^3H_4$ level could also give rise to population of the $^1D_2$ level in a scheme similar to that suggested earlier for population of this level under excitation at 457.9 nm. Such a hypothesis is suggested by comparing measurements of the transmission of blue light down the fiber when the fiber is pumped at 1.064 μm and when it is not. Using the arrangement described by Morkel et al. [10] involving probing the excited fiber with counter-propagating white light we have observed a significant decrease (approximately 80% at 455 nm for a 15 cm length of fiber) in the transmission of light from 450 nm to 465 nm when the fiber is pumped at 1.064 μm. This result is consistent with photons being absorbed from the $^3H_4$ level to the $^1D_2$ level.

Emission at around 800 nm is mainly due to decay from the $^3F_4$ level to the ground state although a contribution at short wavelengths may be due to decay from $^1G_4$ to $^3H_5$.

The transmission of the chopped pump beam as a function of time was also examined and is shown in fig. 9 for an excitation power of 2 W. The time evolution of the fluorescence at 467 nm is also shown. As can be seen the transmitted pump decays on approximately the same time-scale as the fluorescence evolves. The reason for this is the sequential nature of the upconversion process, with absorption of pump

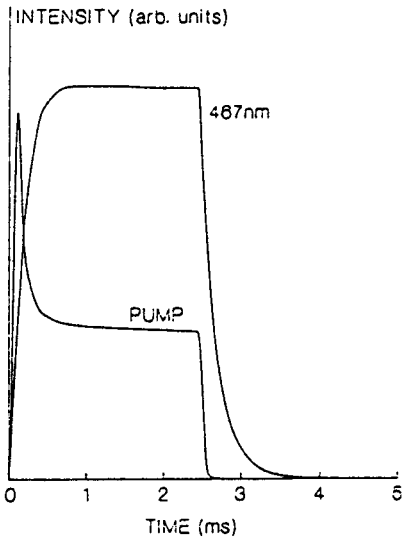

Fig. 9. Temporal evolution and decay of the 467 nm fluorescence and 1.064 μm pump.

photons taking place from successively excited states.

3.2. Yb^{3+}-sensitized Tm^{3+}-doped fiber

The use of Yb^{3+} as a sensitizer for efficient upconversion in Tm^{3+}-doped crystals and glasses is well known [1,11]. The present work describes upconversion in an Yb^{3+}:Tm^{3+} silica fiber under excitation at around 800–900 nm and at 1.064 μm. The spectroscopy of Yb^{3+}-doped silica has been reported elsewhere [12], the most important result for this work being the very broad absorption band for the $^2F_{7/2}$ to $^2F_{5/2}$ transition extending from around 800 nm to well past 1 μm.

(i) *Styryl 9M pumping.* Initial measurements of upconversion in the Yb^{3+}:Tm^{3+} fiber were made under excitation from a Styryl 9M dye laser which operated form ≈795 nm to ≈880 nm. The resulting emission spectrum under excitation t 840 nm is shown in fig. 10. The blue emission at around 475 nm is due to decay from the $^1G_4$ level in Tm^{3+} to the $^3H_6$ ground state with a lifetime of ≈250 μs. The red emission at 650 nm also originates from the $^1G_4$ level with the lower level being the $^3H_4$ level. Weak emission at ≈370 nm (≈1/250 of the blue intensity) was also observed from the $^1D_2$ level.

The intensity of the 475 nm emission as a function

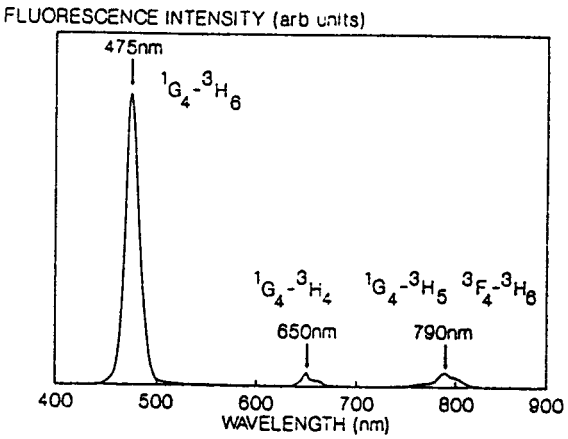

Fig. 10. Emission spectrum for Yb^{3+}:Tm^{3+} silica fiber under excitation at 1.064 μm.

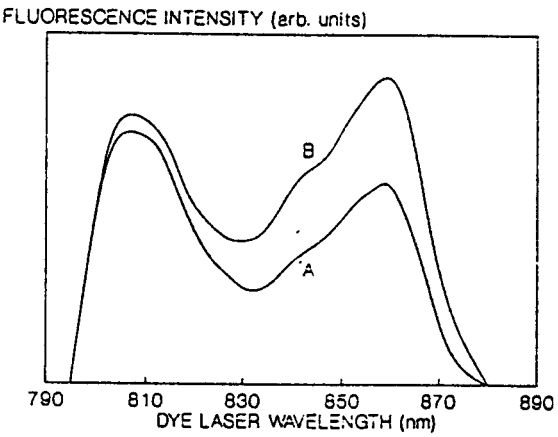

Fig. 11. Excitation spectrum of 475 nm fluorescence from Yb:Tm fiber. Curve A is for dye laser pumping alone, curve B is for additional pumping with a 705 nm diode.

of pump wavelength was examined and is shown as curve A in fig. 11. It should be noted that this excitation spectrum is not normalized with respect to the dye laser power. However, it is clearly evident from the two peaks that two processes occur; one which takes place at short wavelengths around 810 nm, and one which takes place at wavelengths longer than 825 nm. The effect of additionally pumping with a 5 mW laser diode at 795 nm (≈0.5 mW launched into the fiber) while scanning the dye laser was also investigated and the resulting excitation spectrum shown as curve B in fig. 11. It can be seen that the addition of this very modest amount of power at 795

nm significantly increases the yield of blue fluorescence when pumping at wavelengths greater than 830 nm. The effective number of pump photons required to populate the $^1G_4$ level was deduced from the power dependence of the blue fluorescence yield and the results are summarized in table 1. It appears that a two-photon process is responsible at short wavelength excitation and that a three-photon process becomes progressively more important as the pump wavelength gets longer.

The mechanism of the three-photon process is shown in fig. 12 and is similar to that observed in other Yb^{3+}:Tm^{3+}-doped systems [1]. Absorption of pump photons populates the long lived $^2F_{5/2}$ level in Yb^{3+}. Non-resonant energy transfer from the Yb^{3+} ion to the Tm^{3+} ion then takes place causing excitation to the $^3H_5$ level in Tm^{3+} along with the emission of phonons. This population relaxes rapidly to the $^3H_4$ level by non-radiative multiphonon decay. A second non-resonant energy transfer form Yb^{3+} to Tm^{3+}, accompanied again by phonon emission, then populates the $^3F_2$ and $^3F_3$ levels. Again multiphonon decay occurs leading to population of the $^3F_4$ level. A third energy transfer and phonon emission finally populates the $^1G_4$ level from which the blue and red emission emanates. From the shortening of the fluorescence lifetime of Yb^{3+} in the co-doped fiber (550 μs) compared to a fiber containing only Yb^{3+} (770 μs) it is possible to estimate a transfer efficiency of 25% form Yb^{3+} to Tm^{3+}.

Table 1
Number of infra-red pump photons involved in population of $^1G_4$ blue emitting level in Yb^{3+}:Tm^{3+} fiber as a function of pump wavelength.

Wavelength	No. of photons
800 nm	1.7
820 nm	2.1
840 nm	2.5
860 nm	2.8

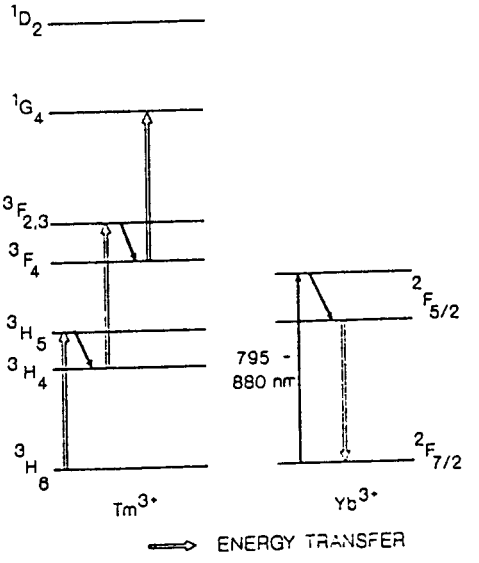

Fig. 12. Mechanism for three-photon upconversion leading to population of the $^1G_4$ level in Yb:Tm silica fiber.

The two-photon process giving rise to population of the $^1G_4$ level also involves energy transfer from the Yb^{3+}. In this case however, the $^3F_4$ level in Tm^{3+} is populated directly by absorption of pump photons below ≈820 nm. The pump photons are only weakly absorbed in the Yb^{3+} and this means that the final stage involving energy transfer is relatively inefficient. In the case of simultaneous pumping with both the diode and dye laser it can be seen that energy transfer at longer dye laser wavelengths is more efficient at raising population from the $^3F_4$ level to $^1G_4$ than shorter dye laser wavelengths.

(ii) *Nd:YAG pumping.* Upconversion under excitation at 1.064 μm was also examined in this fiber. In this case there is ground state absorption in both the Yb^{3+} and the Tm^{3+}. The results for this fiber were similar to those obtained for the singly-doped fiber described earlier. One slight difference, however, is the shape and peak wavelength of the blue fluorescence as shown in fig. 13. It can be clearly seen that the emission peaks in a similar place to that when under excitation from the Styryl 9M dye laser. It is possible that the concentration of Yb^{3+} ions in this fiber is such that clustering may occur. This may significantly affect the local crystal field experienced by the Tm^{3+} ions compared to that in the singly doped fiber. Unfortunately it was ot possible to record a sidelight emission spectrum for this fiber under direct excitation at 457.9 nm owing to significant scattering of pump light out of the side of the fibre and so the peak emission wavelength of the $^1G_4$ to $^3H_6$ transition could not be measured.

It should be noted that there was no significant difference in upconversion efficiency for the co-doped fiber compared to the singly doped fiber. In

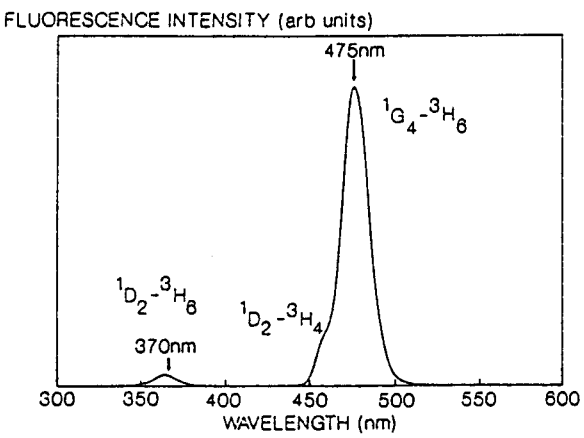

Fig. 13. Emission spectrum for Yb:Tm silica fiber under excitation at 1.064 μm.

both cases it is estimated that for 1W of launched pump power at 1.064 μm about 10^{-5} of the ground state population gets excited to the blue emitting levels. This estimate of the excited population was made by measuring the intensity of the blue emission with the photomultiplier and using the manufacturer's data on responsivity in order ot deduce the fluorescence power. Given this power and the measured lifetime of the emitting level, the population of that level could be deduced, although with a significant uncertainty.

4. Conclusions

The mechanisms involved in upconversion in Tm^{3+}- and Yb^{3+}:Tm^{3+}-doped silica fibers have been examined for several pumping schemes. Under the conditions of our measurements the efficiencies of the upconversion processes have been found to be rather low with typically 10^{-5} of the ground state population being excited to the blue emitting level. The main reason for the low efficiency in silica is considered to be significant shortening of intermediate level lifetimes as a result of non-radiative decay. The rapid non-radiative decay in silica is believed to be the result of the high phonon energies, whereas in heavy metal fluoride glasses where phonon energies are much lower, much longer lifetimes are observed [4], which makes fluoride fibers more promising for upconversion.

While the results of our measurements suggest that prospects for upconversion in silica are not encouraging it should be noted that significant variations in the lifetimes of the $^3H_4$ and $^3F_4$ levels have been observed in silica fibres when the fabrication conditions are varied [13]. A better understanding of the causes of this lifetime variation is needed, and while the possibility exists for further improvements in lifetimes it keeps alive the prospect for upconversion lasing in silica fibers or other oxide glass fibers.

Acknowledgements

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