ANOMALOUSLY HIGH UNIFORM UPCONVERSION IN AN ERBIUM-DOPED WAVEGUIDE AMPLIFIER

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The performance of a planar Er³⁺-doped ion-exchanged waveguide is compared to a detailed model, including uniform upconversion estimated from spectral measurements. A discrepancy between experiment and theory requires a much higher level of uniform upconversion than predicted. We consider possible explanations for this anomaly.

Introduction

In the course of developing a planar ion-exchanged glass waveguide Er^{3+} -doped amplifier, we have considered methods for rapid selection of suitable host glasses¹. The parameters directly related to amplifier operation are the pump and signal cross-sections, which are readily established. However, at the high dopant levels used in short planar amplifiers, cooperative ion-ion interactions impose practical limits on the performance. A distinction can be drawn between "uniform upconversion" [UU] between randomly distributed "isolated" ions and upconversion within clusters of ions. Although clustering, which arises from imperfect rare-earth solubility in the host, has been the main problem in fibres even at relatively low dopant levels, it has recently been suggested that UU will set a fundamental limit in planar amplifiers², even if clustering can be avoided.

We have made spectral measurements on several host glasses to permit first-principles prediction of upconversion strength, and we have estimated the clustering levels in these glasses. Using these figures and the standard spectroscopic quantities required for amplifier characterisation, we have modelled the gain and fluorescence time-dependence in doped glasses. We compare here the predicted and measured performance for one such glass, and find a substantial discrepancy, for which we consider possible explanations.

The Numerical Amplifier Model

Pump and signal evolution along a straight waveguide in a bulk-doped host are calculated using the modal intensity profiles and allowing for saturation effects and bidirectional amplified spontaneous emission. The model can also compute the decay of guided fluorescence when steady-state pumping is abruptly turned off. Ion-ion interactions are incorporated as two independent components, involving the "isolated" and the clustered ions respectively. The rate equation for the metastable population density N_2 of "isolated" ions includes a term of the form $-C_{UC}N_2^2$ to account for UU, where C_{UC} is the upconversion coefficient. A standard rate equation describes the clustered ions³.

Estimation of Ion-Ion Interaction Strengths

We estimate interaction strengths using the Förster-Dexter theory^{4,5}, in which the fundamental quantity is an overlap integral between the spectral distributions of the two transitions involved in the interaction. We neglect phonon-assisted interactions since the relevant interactions involve direct spectral overlaps⁶. The distance of closest approach of Er³⁺ ions we take to be 0.35 nm, as in

crystalline⁷ Er₂O₃. An integration over space with a random distribution of ions yields the average interaction strength per excited ion, which is proportional to the excited state population density. The ion-ion interactions then contribute a quadratic term to the rate equation for the excited state population, as indicated above. The issue of representation of the inversion level by a rate equation is complicated - it is frequently asserted but not true, for example, that dipole-dipole interactions give rise to 3rd order terms⁸. A rate equation approach with this quadratic upconversion term is appropriate provided the excitations are "shuffled" rapidly by migration.

The host glass was an aluminoborosilicate, with 3.6x10¹⁹ Er³⁺ ions cm⁻³. Waveguides 3.8 cm long were made by Tl⁺ ion-exchange from a molten salt bath, and buried in a second step. They were single-moded at signal wavelengths, and modal profiles were determined by the near-field imaging of a polished endface. The required spectra were measured by techniques described elsewhere⁹. We have attempted to measure the clustering level through detection of an unbleachable absorption³, but the method is not very sensitive in short guides and we could specify only an upper limit.

Fluorescence lifetime	5.6 ms
Radiative lifetime	8.6 ms
Dopant concentration	3.6x10 ¹⁹ cm ⁻³
Clustering fraction	<30% of ions in clusters
Q_{abs}	1.5x10 ⁻²² eV cm ²
Q_{em}	2.1x10 ⁻²² eV cm ²

Q _{ESA}	0.4x10 ⁻²² eV cm ²
Overlap int. (migration)	6x10 ⁻²⁴ m ⁵ J ⁻¹
Overlap int. (upconversion)	1x10 ⁻²⁵ m ⁵ J ⁻¹
Min. lifetime by upconversion	25 μs
C _{UC} (see text)	7x10 ⁻¹⁸ cm ³ s ⁻¹
C _Q (see text)	<60x10 ⁻¹⁸ cm ³ s ⁻¹

Table I: Measured properties of doped glass

Table I lists the properties of the doped glass, including integrated transition strengths for the ${}^4I_{15/2} \rightarrow {}^4I_{13/2}$ absorption (Q_{abs}) , the ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ emission (Q_{em}) and the ${}^4I_{13/2} \rightarrow {}^4I_{9/2}$ absorption (Q_{ESA}) . The overlap integral is defined as:

$$\frac{1}{Q_{em}} \int \lambda^2 \sigma_{em}(\lambda) \sigma_x(\lambda) d\lambda$$

where λ is the wavelength and σ the cross-section, with σ_x representing the absorption or ESA cross-section respectively. The overlap between the emission and ESA cross-sections leads to upconversion, whereas excitation migration or hopping involves the overlap of the emission and absorption cross-sections. Migration is much faster than upconversion, so it is plausible that the "shuffling" condition holds. The uncertainty in the calculation of C_{UC} which arises from the noise on the ESA spectrum is considerable, perhaps as high as 50%, even neglecting the theoretical uncertainties. These latter include the contribution of phonon-assisted processes, other interaction modes (eg. exchange interactions) possible correlations in the orientation of close-lying ions, the influence of the magnetic dipole component of the ${}^4I_{13/2}$ - ${}^4I_{15/2}$ transition, and local field corrections.

Table I also shows the minimum lifetime due to upconversion, for ions 0.35 nm apart, which presumably reflects the lifetime in clusters. Estimates in the literature^{2,10} for the upconversion lifetime in clusters range from 3.5 μ s to 100 μ s. This suggests that our calculations are reliable to within better than an order of magnitude.

Results and discussion

Waveguide absorption/gain was measured with the set-up in fig. 1, as a function of pump power. The time-dependence of the fluorescence decay was measured by mechanically chopping the pump and directing the waveguide output at a detector connected to a digital storage oscilloscope.

Fig. 2 shows measurements of absorption/gain versus transmitted pump power, along with two model predictions. The models differ in the upconversion coefficient; A uses the value $7x10^{-18}$ cm³ s⁻¹ predicted as discussed above, and B, which matches the data well, uses $120x10^{-18}$ cm³ s⁻¹, almost 20x greater. Model B also matches the instantaneous decay rate, as shown in table II. Other parameter changes which might account for the observations include reduced excited state lifetime or higher clustering levels. The numerical model has shown these not to match the data.

There is thus a large difference between our predicted UU coefficient and that needed to fit the data. As noted above, our prediction has large uncertainties, but it is hard to accommodate the error required to make the model parameter agree with the theory. We are now evaluating the exchange interaction and magnetic dipole contributions, and the significance of phonon-assisted upconversion.

Pump power	Expt	Model
14 mW	1.5 ms	1.2 ms
3 mW	2.5 ms	2.2 ms
14 μW	4.9 ms	5.2 ms

Table II: Instantaneous fluorescence lifetime when pump turned off

Another possibility is energy migration from "isolated" ions to clusters where quenching is

rapid. The rate is roughly proportional to the inversion level, because an excitation migrating to a cluster will be annihilated only if another is present. Macroscopically, the process mimics UU. We can estimate a quenching coefficient (for paired clusters) as $C_Q = 0.5x$ [migration coefficient]x[fraction of ions that are clustered], where the migration coefficient is computed in a manner analogous to the upconversion coefficient. For this host C_Q could be as high as $60x10^{-18}$ cm³ s⁻¹ (Note that C_Q has the same units as C_{UC} and is directly comparable). However, we need to set the minimum migration radius higher than that for upconversion, so as to count only "effective" jumps which move to a different neighbourhood. If we arbitrarily use a minimum radius twice that for C_{UC} , C_Q is reduced by a factor of 8 to a value close to C_{UC} , which is insufficient to account for the discrepancy.

It is also conceivable that the glass is inhomogeneous, so that the local dopant concentration seen by an individual Er^{3+} ion is substantially higher than the average value. It is perhaps noteworthy that this glass composition shows phase separation at twice the doping concentration used here. In our model, the apparent C_{UC} increases in proportion to the ratio of local to average concentrations, so a doping level of about 5×10^{20} cm⁻³ would be required to account for the data.

Note that this upconversion discrepancy has not been seen in the only other host - a barium silicate - studied in detail. However, the value of C_{UC} used to model this second glass has not yet been directly determined, but inferred from another with very similar ground-state spectroscopy.

Conclusion

In developing approaches to host glass evaluation and planar amplifier modelling, we have found a discrepancy with experimental results for an Er³⁺-doped glass. Our model matches data only with an anomalously large upconversion coefficient. This discrepancy has come to light because we have established an independent estimate for the upconversion coefficient. It is unclear whether its source lies in the upconversion theory or in the glass properties, and work is in progress to elucidate this.

Acknowledgements

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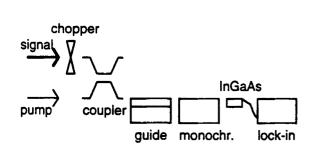


Figure 1: Experimental set-up to measure absorption/gain

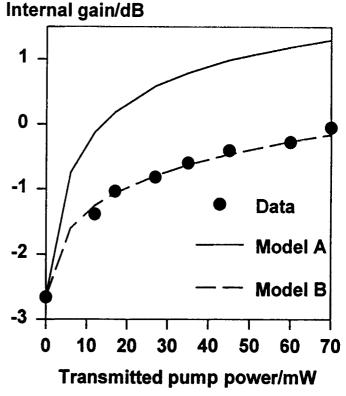


Figure 2: Internal gain of amplifier vs. transmitted 980 nm pump