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Spectroscopy and Optical Amplification in Cr doped $LiNbO_3$

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Abstract

We present measured polarised absorption and emission cross-sections for $Cr:LiNbO_3$, and investigate the feasibility of active waveguide devices in this material system.

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Introduction

Recent efforts in the development of planar waveguide lasers and amplifiers have resulted in the demonstration of sophisticated rare-earth-doped waveguide devices in LiNbO$_3$, taking advantage of the host material electro- and acousto-optic properties [1-3]. However, RE^{3+} ions in LiNbO$_3$ typically exhibit a short tuneability range. Transition metal ions, such as chromium, have on the other hand been used extensively in the past few years to demonstrate broad tuneability in various crystalline host materials [4], other than LiNbO$_3$.

The absorption spectrum of Cr-doped lithium niobate shows two bands with peaks at 480 nm and 660 nm, approximately [5]. The fluorescence spectrum consists of a broad band with a peak at around 900 nm and half-width of about 200 nm [5]. Gain in proton-exchanged waveguides in chromium bulk-doped lithium niobate has been reported [6]. Using Ti-indiffused waveguides fabricated in Cr diffusion doped substrates internal gain was observed [7].

These results, if confirmed, indicate a significant potential for an integrated, broad-band tuneable laser in the 750-1150 nm spectral range, with the additional prospect of diode laser pumping, using Cr-doped LiNbO$_3$ substrates. However, attempts to repeat these measurements, combined with the new measurements of polarised cross-sections, cast doubt upon the accuracy of the earlier results.

Previous calculations of the gain behaviour [8] were based on published values for the cross sections and lifetimes and showed that reasonable gain could be expected in waveguides on Cr-doped lithium niobate substrates. We present here the measured absorption and emission cross sections and the resulting theoretically predicted amplification in Cr-doped lithium niobate waveguides.

Spectroscopy of Cr-Doped LiNbO$_3$

The spectroscopy of Cr^{3+} ions in octahedral crystal fields is usually discussed using the Tanabe-Sugano diagram [9]. This is a plot of the energy eigenvalues of the crystal field Hamiltonian of the d^3 configuration as a function of Dq, the strength of the octahedral field. The free ion states relevant for the optical properties are, in order of increasing energy, ^4F, ^4P and ^2G. The crystal field splits level ^4F into a ground state orbital singlet $^4A_2$ and excited state orbital triplets $^4T_2$ and $^4T_1$. The broad absorption bands are due to transitions from level $^4A_2$ to the level $^4T_2$ (band centered at 650 nm) and $^4T_1$ (band centered at 480 nm). Also, due to the crystal field, splitting of the free ion excited state ^2G gives rise to level ^2E. Transitions from the ground state $^4A_2$ to level ^2E lead to the narrow absorption band overlapping the wing of the broad absorption band due to the transition from $^4A_2$ to $^4T_2$. Depending upon whether level ^2E lies below or above level $^4T_2$, the fluorescence spectrum shows sharp R lines or a broad fluorescence band.

In the case of lithium niobate, these levels are almost degenerate and the fluorescence spectrum shows both features. The broad fluorescence band is due to the transition from level $^4T_2$ to level $^4A_2$. Transitions from level ^2E to ground state $^4A_2$ give rise to sharp R lines near 725 nm [9]. These can only be seen at low temperatures because the transition from level $^4T_2$ is vibronically broadened. The Stokes shift between absorption and fluorescence in the $^4T_2$-$^4A_2$ transition provides a possible 4-level optical amplification system.

Absorption and emission spectra

Four samples of Cr-doped LiNbO3 were used to measure absorption and emission cross sections. The concentrations for three samples, grown by the University of Tianjin (UT)-China, were supplied as 0.5, 0.9 and 1.3 ($\times 10^{19}$ cm^{-3}) of Cr^{3+}. The fourth sample was grown by Union Carbide (UC) and its concentration was 0.4×10^{19} cm^{-3}.

For sample UC, the absorption cross section is shown in fig. 1, for both σ and π polarisations together with the labelling of the corresponding transitions. The spectra were measured on a Perkin-Elmer Lambda 9 spectrophotometer. A peak value of 8.3×10^{-19} cm^2 at 657 nm was measured for σ polarisation. The shape of the polarised absorption spectra compares well with the published uncalibrated plots [10].

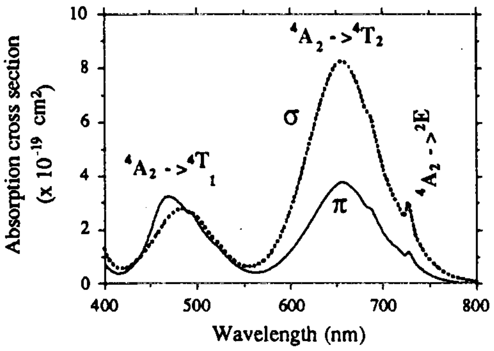

Fig. 1: *Polarised absorption cross sections for sample UC doped with 0.4×10^{19} cm^{-3}*

The polarised fluorescence spectra measured for sample UC are shown in fig. 2, and are typical of the $^4T_2$-$^4A_2$ transition. An argon ion laser at 488 nm was used to populate level $^4T_1$, from which a fast decay to level $^4T_2$ occurs.

In order to calculate the emission cross section we used the Fuchtbauer-Ladenburg relation [11]. The peak emission cross section is given by:

$$\sigma_e^{peak} = \frac{\lambda_{peak}^4}{8\pi c n^2 \tau_{rad} \Delta\lambda_{eff}}$$

where λ_{peak} is the wavelength at the peak of fluorescence , c is the light velocity in vacuum, n is the refractive index of the medium at the peak wavelength, τ_{rad} is the radiative lifetime and $\Delta\lambda_{eff}$ is the fluorescence effective linewidth, defined as:

$$\Delta\lambda_{eff} = \frac{\int I_{FLR}(\lambda)d\lambda}{I_{Peak}}$$

where $I_{FLR}(\lambda)$ is the fluorescence intensity spectrum measured in arbitrary units and I_{peak} its peak value.

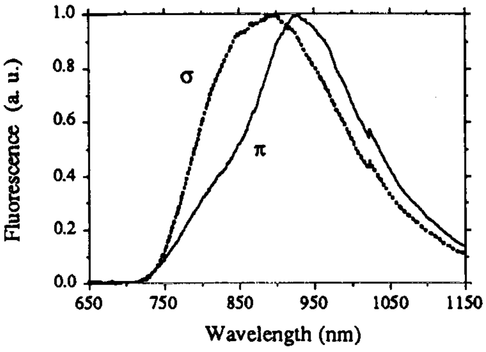

Fig. 2: *Polarised fuorescence spectra for sample UC doped with 0.4×10^{19} cm^{-3} showing the $^4T_2 \to ^4A_2$ transition.*

The fluorescence lifetime was previously measured as a function of temperature in the same UC sample [7]. We measured a constant value of approximately 9 µs at temperatures below 100 K and 1 µs at room temperature. We assume that the radiative lifetime is 9 µs, and that the room temperature lifetime is due to multiphonon nonradiative decay, indicating the room temperature fluorescence quantum efficiency to be ≈ 0.11.

Using the above relation and the measured data, we calculated, for the σ polarised emission- cross section, 8.4×10^{-20} cm^2 at 898 nm, and 1.1×10^{-19} cm^2 at 927 nm for the π polarised emission cross section. In [12] Babadjanyan et al presented a value of 2×10^{-19} cm^2 , but did not mention a specific polarisation.

Cr:LiNbO$_3$ waveguide amplifiers

A simple model of laser amplification was applied to estimate the behaviour of Cr:LiNbO3 channel waveguides in both bulk- and diffusion-doped structures [5], using the measured cross-sections as above and reasonable estimates for the scattering losses and waveguide modal profiles for pump and signal. The rate equations for a 4-level laser system in the steady state were used, as in [13], for the cases of bulk- and diffusion-doped amplifiers.

Due to the high peak value of the absorption cross section we chose a pump wavelength of 750 nm, for which we measured an absorption cross section of

8.9×10^{-20} cm^2 in σ polarization. We assumed a signal wavelength of 927 nm, which is the peak of the emission cross section. A surface Cr concentration of 5.0×10^{20} cm^{-3}, which can be readily achieved in diffusion-doped samples [5], was used. We assume scattering losses to be on the order of 0.4 dB/cm, an effective pump area of 20 μm^2 and a lifetime of 1 µs (at room temperature).

The gain evolution as a function of cavity length is shown in fig.3 for three values of launched pump power. It was found that, for a launched pump power of 100 mW, a theoretical maximum net gain of 0.85 dB can be obtained for a waveguide length of around 1 mm.

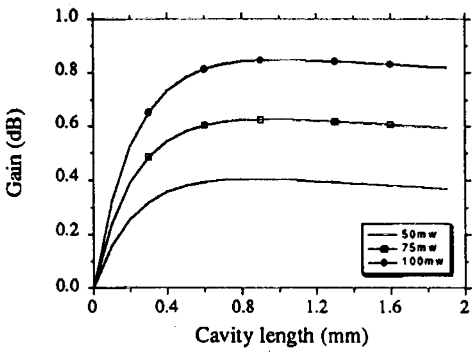

Fig. 3: *Simulated gain evolution in a waveguide on Cr:LiNbO₃.*

As it is difficult to fabricate a device as short as 1 mm, further detuning of the pump absorption is desirable, along with a decrease in the Cr concentration. If a pump wavelength of 800 nm is used, an optimum cavity length of 4.5 mm was calculated, for which a maximum gain of 0.68 dB could be achieved for 100 mW of launched pump power. A decrease in the Cr concentration has not been considered, because it would also reduce the gain.

Based on the above calculation we can conclude that Cr:$LiNbO_3$ is, at best, a low gain material, yielding a gain below 1dB for a 1 mm sample, when a reasonably high pump power is launched, thus requiring waveguides with very low loss. Moreover, the short wavelengths that have to be used to excite the Cr^{3+} ions are known to induce photorefractive damage in $LiNbO_3$, increasing the losses and further reducing the gain. Proton-exchange in conjunction with MgO doped $LiNbO_3$ may potentially overcome the problems associated with optical damage.

Conclusions

The polarised absorption and emission cross sections of Cr doped $LiNbO_3$ were measured for the first time. Using this accurate spectroscopic data, an estimate of the gain behaviour of a waveguide amplifier has been carried out using a simple model. The combination of a relatively small emission cross section with a short lifetime shows that this material exhibits low gain for high pump power density in contrast to previous expectations.

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