Optical Characterization and 2,525 μm Lasing of Cr^{2+}: $Cd_{0.85}Mn_{0.15}Te$

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1. Introduction

Transition metal doped solids are of significant current interest for the development of tunable solid-state lasers for the near and mid-infrared (1-4 pm) spectral region [1,2]. Applications of these lasers include basic research in atomic, molecular, and solid-state physics, optical communication, medicine, and environmental studies of the atmosphere.

In transition metal based laser materials, absorption and emission of light arises from electronic transitions between crystal field split energy levels of 3d transition metal ions. The optical spectra generally exhibit broad bands due to the strong interaction between dopant and host (electron-phonon coupling) [3]. Broad emission bands offer the prospect of tunable laser activity over a wide wavelength range, e.g. the tuning range of Ti:Sapphire extends from 700-1100 run [1,2]. The only current transition metal laser operating in the mid-infrared wavelength region (1.8-2.4 μm) is Co^{2+}:MgF_2, but its performance is severely limited due to strong nonradiative decay at room temperature. Based on lifetime data, the quantum efficiency is estimated to be less than 3°/0 [1,2]. In general, the probability for non-radiative decay via multi-phonon relaxation increases with decreasing energy gap between ground and excited state. Therefore, efficient transition metal lasers beyond -1.6 μm are rare [1,2].

Recently, tunable laser activity around 2.3 μm was observed from Cr doped ZnS and ZnSe [4,5]. The new lasing center in these materials was identified as Cr^{2+} occupying the tetrahedral Zn site. Tetrahedrally coordinated optical centers are rather unusual among transition metal lasers [1,2]. Their potential usefulness, however, has been demonstrated by the recent development of near infrared laser materials such as Cr:forsterite and Cr:YAG, which are based on tetrahedrally coordinated Cr^{4+} ions [6,7]. According to the Laporte selection rule, electric-dipole transition within the optically active 3d-electron shells are parity forbidden [3]. However, a static acentric electric crystal field or the coupling of asymmetric phonons can force electric-dipole transitions by the admixture of wavefunctions with opposite parity. Tetrahedral sites lack inversion symmetry which provides the odd-parity field necessary to relax the parity selection rule. Therefore, high absorption and emission cross sections are observed. An enhanced radiative emission rate is also expected to reduce the detrimental effect of non-radiative decay [4,5]. Motivated by the initial results on Cr doped ZnS and ZnSe, we have started a comprehensive effort to study Cr^{2+} doped II-VI semiconductors for solid-state laser applications. In this paper we present the optical properties and the demonstration of mid-infrared lasing from Cr doped $Cd_{0.85}Mn_{0.15}Te$.

2. Experimental details

The undoped and Cr doped $Cd_{0.85}Mn_{0.15}Te$ samples were prepared at Brimrose Corporation using a modified Bridgman growth method. The nominal Cr concentration was $5x10^{19}$ atoms/cm³. Absorption measurements were carried out using a Cary 5E spectrophotometer. Luminescence was excited with the 1.9 μm output of a Q-switched Nd:YAG pumped Optic-al Parametric Oscillator. The infrared emission was dispersed with a 1m monochromator and detected with a liquid nitrogen cooled InSb detector. The emission signal was processed with a boxcar averager and personal computer. All spectra were corrected for the spectral response of the luminescence setup. Lifetime measurements were performed by directly monitoring the InSb signal on an averaging digitizing oscilloscope. Cooling was achieved by mounting the sample on the cold finger of a closed cycle helium refrigerator. Laser experiments were performed using the pulsed (10Hz) 1.9 μm output of a H_2-Raman shifted Nd:YAG laser.

3. **Results and Discussion**

Absorption:

The unpolarized absorption spectra of undoped (upper trace) and Cr doped (lower trace) $Cd_{0.85}Mn_{0.15}Te$ are shown in Figure 1. The incorporation of Cr into the $Cd_{0.85}Mn_{0.15}Te$ lattice results in a strong absorption band centered at around 1900 nm with a width of ~400 nm (FWHM). This absorption feature is similar to that measured for other Cr doped II-VI's semiconductors [8] and is attributed to the only spin-allowed transition $(^5T_2 \rightarrow {}^5E)$ of Cr^{2+} ions ($3d^4$ electronic configuration) in a tetrahedral telluride coordination. Using the nominal Cr concentration of $5 \times 10^{19}/cm^3$ the peak absorption cross section was calculated to be $4.4 \times 10^{-19} cm^2$. This value has to be considered as a lower limit because the actual concentration of Cr^{2+} ions in the sample is unknown.

Figure 1 also shows that the bandedge of Cr doped $Cd_{0.85}Mn_{0.15}Te$ exhibits a strong absorption shoulder extending nearly down to 1000 nm which is not observed in the undoped $Cd_{0.85}Mn_{0.15}Te$ sample. Since all transitions from the $^5T_2$ ground state of Cr^{2+} to higher excited states are spin-forbidden, it is unlikely that the 1000 nm absorption shoulder arises from an intra 3 d transition. Further work on the identification of the Cr induced near bandedge absorption is in progress.
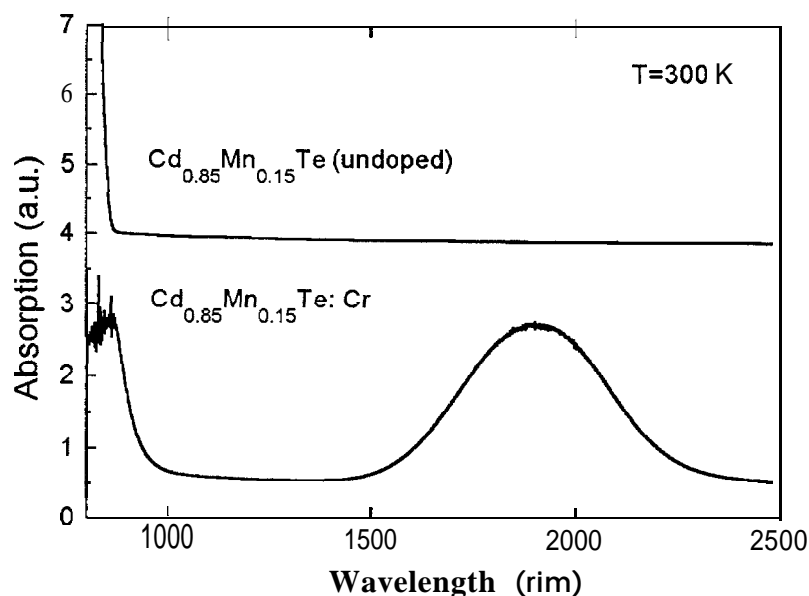

Figure 1: Unpolarized absorption spectra of undoped (upper trace) and Cr doped (lower trace) $Cd_{0.85}Mn_{0.15}Te$. The strong absorption band centered at 1900 nm is attributed to tetrahedrally coordinated Cr^{2+} ions.

Emission:

The room temperature luminescence spectrum of $Cr:Cd_{0.85}Mn_{0.15}Te$ excited at 1900 nm shows a strong band centered at 2250 nm with a FWHM of 450 nm and a lifetime of 1.4 µs. When cooling the sample to 15 K, the emission band narrows and consists of a main peak at 2200 nm and a shoulder at 2400 nm. The double peak in the low temperature spectrum indicates the

derivation of the local Cr^{2-} environment from perfect tetrahedral symmetry [3]. At 15 K the lifetime increases to 3.7 μs. Under the assumption that the radiative decay rate is constant with temperature, any change in he measured lifetime is interpreted as being due to the onset of non-radiative decay [3,9], This yields a promising high quantum efficiency of -38% at room temperature.
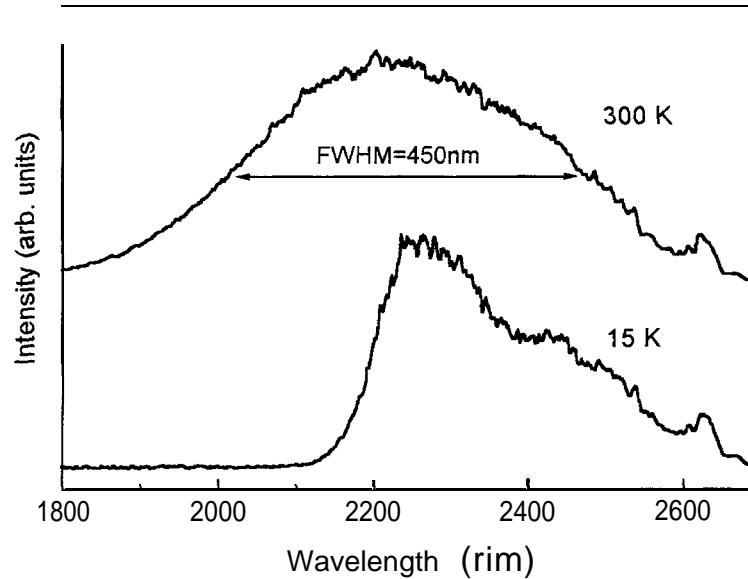
Figure 2: Mid-infrared luminescence spectra of Cr^{2+}:CdMnTe at low and room temperature. The luminescence was excited with the 1900 nm output of a Q-switched Nd:YAG pumped Optical Parametric Oscillator.

Using the estimated quantum efficiency and lifetime τ, the room temperature emission cross section σ_{em} was calculated according to McCumber's theory [10,11]:

$$\sigma_{em} = \eta \cdot \sqrt{\frac{\ln 2}{\pi} \frac{1}{4\pi cn^2}} \frac{1}{\tau} \cdot \frac{\lambda^4}{\Delta\lambda}$$

where n is the refractive index, c is the speed of light, λ is the emission wavelength, and $\Delta\lambda$ is the full width half maximum (FWHM) of the emission band. It is implied in McCumber's theory that the emission spectrum has a nearly Gaussian band-shape. For the case of $Cr:Cd_{0.85}Mn_{0.15}Te$, we obtained an emission cross section of $2.7\times10^{-18}\,cm^2$, using η=38%, λ=2250 nm, $\Delta\lambda$=450 nm, τ=1.4μs, and n=2.7 (CdTe). The emission cross section of Cr^{2+}:CdMnTe is significantly larger than that of the commercial laser material Ti:Sapphire [1,2].

Laser experiments

Room temperature laser operation of Cr^{2+}: CdMnTe was demonstrated with a 3-4 mm thick sample placed in a cavity consisting of a flat high reflector (R>99%@ 2350 nm) and a curved output coupler (R=95%@ 2350nm). For the excitation of Cr^{2+} ions the 1907 nm output from a H_2 Raman-Cell pumped by a Nd:YAG laser was used (Figure 3). Laser activity centered at 2525 nm was achieved with a slope efficiency of 4. 5°/0 (Figure 4). More detailed studies on the laser performance of Cr^{2+}:$Cd_{0.85}Mn_{0.15}Te$ are currently in progress and will be published in a forthcoming paper [12].

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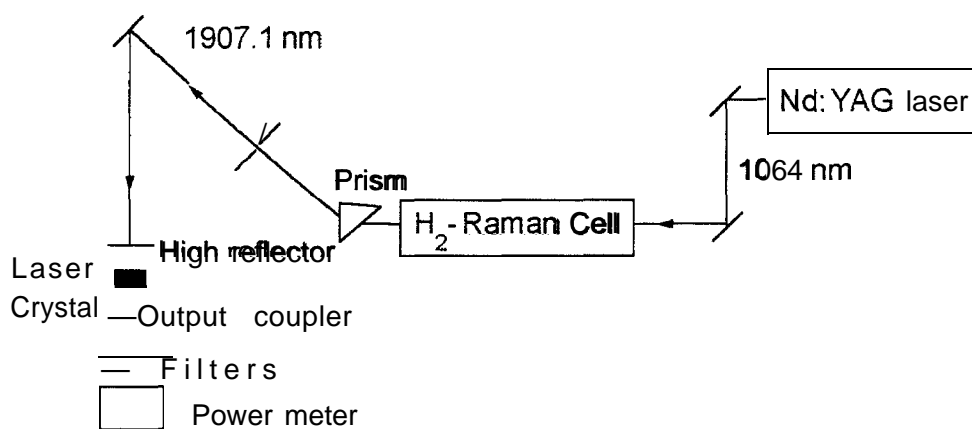
Figure 3: Experimental setup for testing the laser activity of Cr^{2+}:CdMnTe.
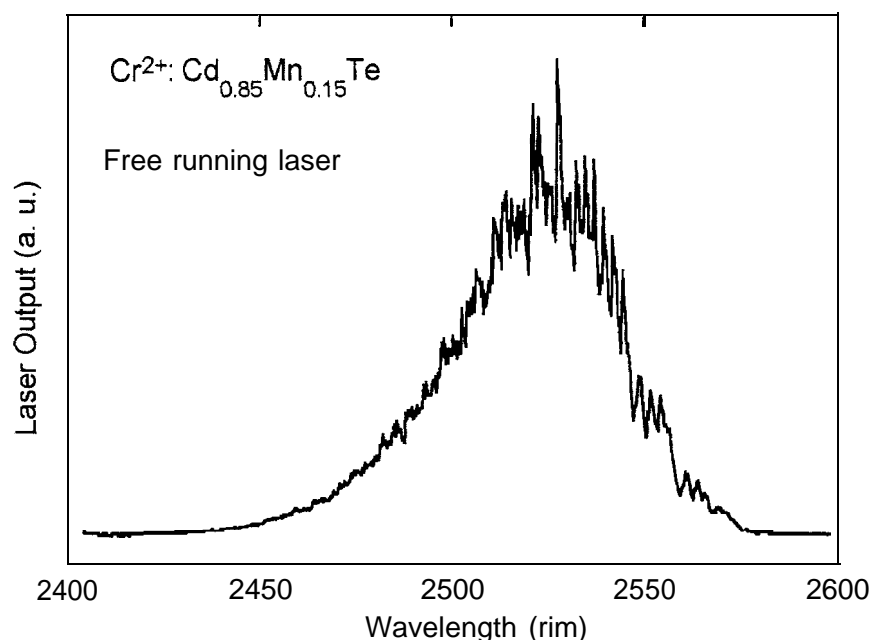
Figure 4: Spectrum of the free running Cr^{2+}:$Cd_{0.85}Mn_{0.15}Te$ laser at room temperature.

4. Conclusion

The optical properties of Cr doped $Cd_{0.85}Mn_{0.15}Te$ were presented. This system shows a broad absorption band centered at 1900 nm which is attributed to an intra 3d transition of tetrahedrally coordinated Cr^{2+} ions. Direct excitation into this absorption band resulted in a strong mid-infrared luminescence centered at 2250 nm. Based on lifetime measurements the quantum efficiency was estimated to be 3 8°/0. Initial laser experiments have been carried out and room temperature laser activity at 2525 nm with a FWHM of -50 nm was demonstrated. Under less than optimal conditions the slope efficiency was measured to be 4. 5%.

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Acknowledgment

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