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A NEW GENERATION OF LASER ACTIVE IONS FOR TUNABLE LASER OPERATION: THE TRANSITION - METAL IONS IN TETRAHEDRAL COORDINATION

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Abstract : We report the polarized absorption spectra, and the emission or stimulated emission cross-section spectra of several Cr^{4+} , Mn^{5+} and Co^{2+} doped crystals. The emission lifetimes as a function of temperature are reported as well.

The absorption characteristics of many inorganic materials doped with transition-metal ions have been often discovered a long time before their fluorescence properties. This is due sometimes to misinterpretations of the spectra which have led to investigations in wavelength domains where nothing should appear or to technical barriers linked for example to the difficulty to register infrared spectra at a time where the detectors were not sensitive enough.

Now a number of these crystals have been lased efficiently or have been studied for their potential laser properties. In the last decade, it has been the case of materials doped with transition-metal ions such as Ti^{3+} , Cr^{3+} , V^{2+} , Co^{2+} and Ni^{2+} and a number of them such as alexandrite ($\text{BeAl}_2\text{O}_4 : \text{Cr}^{3+}$), Ti : saphir ($\text{Al}_2\text{O}_3 : \text{Ti}^{3+}$) and $\text{MgF}_2 : \text{Co}^{2+}$ are now commercially available. However in these systems, the active ions site in nearly octahedral environments so that the cross-sections of the optical transitions are relatively weak (10^{-21} - 10^{-19} cm^2) which make them relatively low gain media. That is why the search in this domain still remains very active. In particular, in the recent years, a new solution has appeared with the tetrahedrally coordinated transition-metal ions. In this case the optical transitions can be electric-dipole allowed thus give rise to larger emission cross-sections (10^{-19} - 10^{-17} cm^2) and possibly higher gain materials. This applies to the now famous Cr^{4+} doped forsterite (Mg_2SiO_4) ⁽¹⁾ and might be the case of the recently discovered ⁽²⁾ Mn^{5+} doped spodiosite ($\text{A}_2(\text{PO}_4)\text{B}$) and apatite ($\text{A}_2(\text{PO}_4)_3\text{B}$) crystals ($\text{A}=\text{Ca}, \text{Sr}$ $\text{B}=\text{Cl}, \text{F}$). In this paper, we present the main spectroscopic and fluorescence data of such systems.

The two $3d^2$ ions Cr^{4+} and Mn^{5+} doped systems are characterized by an interesting emission band - figure 1 to 3 - around 1200 nm, due in the case of $\text{Mg}_2\text{SiO}_4 : \text{Cr}^{4+}$ to the $^3\text{T}_2 \rightarrow ^3\text{A}_2$ transition, and to $^1\text{E} \rightarrow ^3\text{A}_2$ transition in the case of Mn^{5+} doped systems. Their fluorescence lifetimes and their temperature dependence are given in figure 6.

In the case of Co^{2+} doped LGO (LiGa_5O_8) or LMA ($\text{La MgAl}_{11}\text{O}_{19}$) - figure 4-5 - we observe three emission bands around 700, 900, and 1100 or 1200 nm. The calculated stimulated emission cross-section is about $2.4 \cdot 10^{-17} \text{ cm}^2$ at 690 nm in LGO : Co^{2+} and $1.6 \cdot 10^{-18} \text{ cm}^2$ at 670 nm in LMA : Co^{2+} . Nevertheless, the fluorescence lifetime in these two systems is relatively short - figure.7 -, and decreases drastically with increasing temperature in the case of LMA : Co^{2+} , and its room temperature quantum efficiency is certainly low.

Fig.1. Room temperature polarized absorption and stimulated emission cross-section of $\text{MS}(\text{MgSi}_2\text{O}_4) : \text{Cr}^{4+}$.

Fig.2. Low temperature polarized absorption and room temperature stimulated emission cross-section of $\text{CPC}(\text{Ca}_2\text{PO}_4\text{Cl}) : \text{Mn}^{5+}$.

Fig.3. Low temperature polarized absorption and room temperature stimulated emission cross-section of $\text{SPC}(\text{Sr}_5(\text{PO}_4)_3\text{Cl}) : \text{Mn}^{5+}$.

Fig.4. Absorption and emission spectra of LGO(LiGa₅O₈): Co²⁺ at room temperature.

Fig.5. Absorption and emission spectra of LMA(LaMgAl₁₁O₁₉): Co²⁺ at room temperature.

Fig.6. Temperature dependence of the fluorescence lifetime in CPC : Mn⁵⁺ (•), SPC:Mn⁵⁺ (o) and MS : Cr⁴⁺ (Δ).

Fig.7. Temperature dependence of the fluorescence lifetime in LMA : Co²⁺ (•) and LGO :Co²⁺ (o).

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