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OPTICAL PROPERTIES OF Cr^{3+} ION IN $LaMgAl_{11}O_{19}$ and $La_{1-x}Nd_xMgAl_{11}O_{19}$

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Abstract

Lanthanum-magnesium hexaaluminate $LaMgAl_{11}O_{19}$ with magnetoplumbite-like structure is a remarkable good laser host for Nd^{3+} ions (LNA). In this study Cr^{3+} ions are introduced as an activator ($LaMgAl_{11-x}Cr_xO_{19}$) or as a sensitizer ($La_{1-x}Nd_xMgAl_{11}O_{19}$) in this matrix. ESR and optical spectra of $LaMgAl_{11-x}Cr_xO_{19}$ crystals reveal 3 kinds of sites for Cr^{3+} : (4f), (2a) and (12k). The presence of $^4T_2 \rightarrow {}^4A_2$ broad band fluorescence makes this material a possible candidate for a red or IR emitting vibronic laser. Optical properties of $La_{1-x}Nd_xMgAl_{11-y}Cr_yO_{19}$ reveal that both radiative and non-radiative $Cr^{3+} \rightarrow Nd^{3+}$ energy transfers occur in this material. This allows to increase the laser emission ratio compared with single doped LNA.

Introduction

Lanthanum-magnesium hexaaluminate $LaMgAl_{11}O_{19}$ with magnetoplumbite like structure (1) is a remarkable good host for laser materials. Doped with Nd^{3+} it forms the LNA ($La_{1-x}Nd_xMgAl_{11}O_{19}$) a new performant IR laser material (2). This paper deals with the optical properties of $LaMgAl_{11}O_{19}$ doped with Cr^{3+} acting
- either as an activator in $LaMgAl_{11-x}Cr_xO_{19}$
- or as a sensitizer in $La_{1-x}Nd_xMgAl_{11-y}Cr_yO_{19}$.

Large rod-shaped single crystals of these 2 kinds of materials are obtained by the flame fusion method. Their structure is hexagonal with space group $P6_3/mmc$ (magnetoplumbite type). They exhibit a cleavage along (001) planes. Optical measurements are performed on cleavage platelets.

$LaMgAl_{11-x}Cr_xO_{19}$ (3)

* ESR studies are carried out in the 120-300 K range. Angular variation of the spectra reveals that Cr^{3+} ions are mainly localized into the (4f) antiprisms, a few of them being situated in (12 k) octahedra. Small satellites flanking the central ESR line are due to Cr-Cr pairs in (4f)-(4f) sites. Their intensity increases with chromium concentration.

* The D_q parameter obtained from absorption spectra is lower than for ruby, higher than for GSGG and almost identical to that of alexandrite ($BeAl_2O_4$:Cr). This may allow to observe the fluorescent transitions both from ^2E and $^4T_2$ levels (4).

* Fluorescent spectra (excitation λ = 532 nm) were performed between 4K and 295K (see fig. 1). They exhibit, on the short wavelengths side $^2E \rightarrow {}^4A_2$ narrow lines revealing the existence, from their respective wavelength 695.3 nm, 688.6 nm and 691.5 nm, of 3 kinds of sites for Cr^{3+}:(4f) trigonal antiprism (main site) but also (2a) regular and (12 k) distorted octahedra, both with low occupancy ratio.

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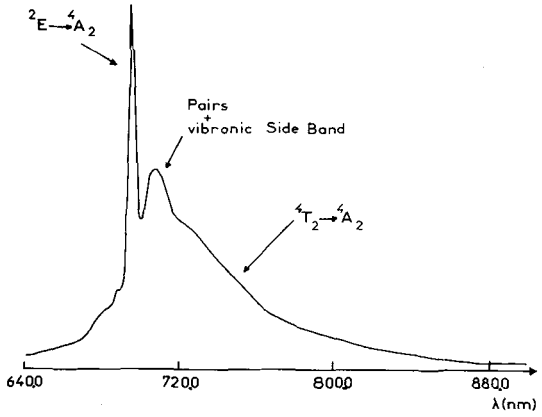

Figure 1
Fluorescence spectrum of Cr^{3+} in $LaMgAl_{11-x}Cr_xO_{19}$

One observes also the $^4T_2$ - $^4A_2$ broad band emission responsible of the long tail of the fluorescence spectrum toward the long wavelengths. Satellite lines around 720 nm are due both to vibronic side bands and Cr-Cr pairs transitions.

* Chromium fluorescence decays were followed using a time scale corresponding to the ^2E - $^4A_2$ transition (a few ms). The fluorescence decays are exponential at 4.4 K for low Cr^{3+} concentration but not when the concentration increases. From these results it turns that the presence of a broad band luminescence extending towards the long wavelengths in $LaMgAl_{11-x}Cr_xO_{19}$ makes this material a possible candidate for red or near IR emitting vibronic laser.

$La_{1-x}Nd_xMgAl_{11-y}Cr_yO_{19}$

In single doped neodymium laser materials, the continuous emission spectrum of pumping lamp and the narrow absorption bands of Nd^{3+} are not well matched. The introduction in the Nd^{3+} doped matrix of a second ion exhibiting large absorption bands in the visible range (like Cr^{3+} ions) allows to sensitize the fluorescence of Nd^{3+} (In GSGG:Nd^{3+} the codoping by Cr^{3+} leads to a yield of 4.5 % compared to the single doped YAG:Nd crystal (about 1,5 %) (5)). Cr^{3+} - Nd^{3+} transfers are demonstrated in the hexaaluminate from Nd^{3+} fluorescence excitation spectrum and optical properties of chromium (intensities measurements and fluorescence intensity decay on single doped and codoped samples).

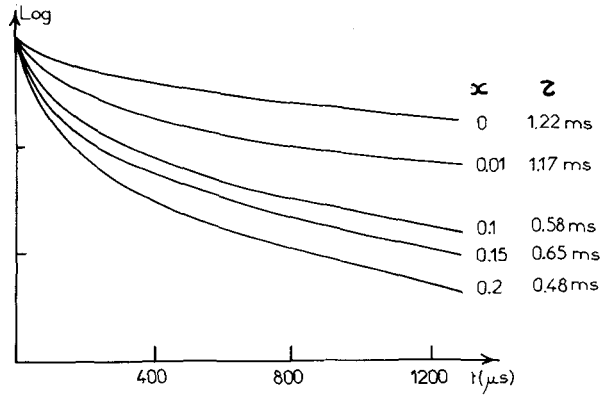

Figure 2
Fluorescence decays and lifetimes of Cr^{3+} in $La_{1-x}Nd_xMgAl_{10.89}Cr_{0.11}O_{19}$

Transfer occurs by radiative mode because the dips in the Cr^{3+} emission broad band correspond to the Nd^{3+} absorption lines.
Transfer occurs too by non radiative mode, the integrated fluorescence of Cr^{3+} ion is decreasing in codoped samples. The Cr^{3+} fluorescence decays are non exponential and get faster as Nd^{3+} concentration increases (see Fig. 2).
Mean parameters are used to characterize the transfer : the transfer efficiency is high (73 % for $La_{0.9}Nd_{0.1}MgAl_{10.89}Cr_{0.11}O_{19}$) but the time transfer reachs a high value (210 μs for $La_{0.9}Nd_{0.1}MgAl_{10.89}Cr_{0.11}O_{19}$).
From these results it turns that Cr^{3+} sensitizes the Nd^{3+} fluorescence in the $LaMgAl_{11}O_{19}$ matrix however the transfer is efficient but very slow. The consequence is that the coactivation will be much more efficient in CW operation than in Q-switched one.

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