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TUNABLE LASER EFECT IN ASN A NEODYMIUM-ACTIVATED STRONTIUM-MAGNESIUM HEXAALUMINATE

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Abstract - Crystal growth and characterization, optical properties and laser action in ASN - a new laser material - are reported. Compared with the related compound LNA, ASN crystals have the significant advantage to growth spontaneously along the $\vec{c}$ crystal direction, which is the optimal one for the laser effect. ASN has also the broadest tunability range so far reported for a neodymium activated laser crystal.

1.- Introduction.

Several neodymium doped lanthanide aluminates such as YAG: Nd and $La_{1-y}Nd_yMgAl_{11}O_{19}$ (LNA) /1/ are of great interest as solid state laser materials. At the present time, only the YAG:Nd has achieved commercial importance. There are continuing efforts to find other neodymium doped crystalline hosts which can offer some advantages with respect to YAG:Nd for particular applications.
ASN (Aluminate de Strontium Neodyme) is a new laser material formed by partial replacement of Sr^{2+} ions by Nd^{3+} in the magnetoplumbite-type (MP) strontium hexaaluminate $SrAl_{12}O_{19}$, isostructural with LNA. In this matrix electroneutrality is restored by simultaneous replacement of Al^{3+} by Mg^{2+} leading to the formulae $Sr_{1-x}Nd_xMg_xAl_{12-x}O_{19}$ $(0 < x < 1)$. This charge-compensation process was found to be the most suitable.
The following paper reports the synthesis of large-sized single crystals with good optical quality by the Czochralski pulling technique. ESR and optical absorption spectroscopies are used to study the localization of Nd^{3+} ions in ASN and CW Laser tests are performed.

2.- Crystal growth.

Preliminary optical studies were made on single crystals grown by floating zone technique using an arc image furnace. The melting point is close to 1850°C. The floating zone technique is very convenient for preparation of small crystals because it is rapid (1 $cm.h^{-1}$), non polluting and needs only a small amount of material. We used it for testing many samples of different compositions in order to determine the compounds exhibiting the optimal properties.
Then single crystals of these selected compounds were grown by Czochralski method. According to the high melting point of the compound, iridium crucible (50 mm in length; 50 mm in diameter) under argon or nitrogen atmosphere was used. The crystal was pulled at a speed of 0.5 mm/h and rotated at 25 rpm.
At first a crystal is pulled from the melt with an iridium rod without seed.

Crystals of ASN grow spontaneously along the $\vec{c}$ axis of the hexagonal structure which corresponds to the optimal direction for laser effect. Of course, this result was confirmed by pulling experiments with $\vec{c}$ oriented seed. These seeds were obtained by cutting cylindrical rods from the first

Czochralski boules of ASN.

It is a major difference of ASN with respect to LNA that it can be grown easily in the $\vec{c}$ crystal direction.

3.- Spectroscopic studies.

ESR

The spectra were performed for different orientations of the static magnetic field B_0 with respect to the $\vec{c}$ axis of the crystals.

$B_0 \,//\, \vec{c}$: Spectra are always constituted of a narrow central line (associated with Nd^{3+} ions of zero nuclear spin) corresponding to a single type of site occupied by neodymium. On the contrary, in the LNA case, there are two central lines which reveal the multisite character of the neodymium ions.

$B_0 \perp \vec{c}$: When B_0 rotates into the $(\vec{a}, \vec{b})$ plane, the ESR spectrum is mainly constituted of a broad line, nearly isotropic. However, its position exhibits a very small fluctuation with a 60° periodicity. This indicates that the main site for the Nd^{3+} ion in ASN is not perfectly axial, even for very low x values, but is very close to the theoretical large cation site (2d), in Wyckoff notation, of the magnetoplumbite structure with D_{3h} symmetry.

Optical absorption.

Electronic absorption spectra of Nd^{3+} give informations about the localization of Nd^{3+}. At very low temperature (4 K), there is only one line for the transition $^4I_{9/2} \rightarrow {}^2P_{1/2}$ for each type of Nd^{3+}. In LNA, one can observe 3 lines corresponding to this transition (figure 1) and can conclude that Nd^{3+} ions are distributed among 3 different sites /2/. On the contrary in ASN only one line is observed when $0 < x < 0.2$, for higher x values a second site appears and the corresponding line at 429 nm grows with the neodymium content (figure 1). In ASN, Nd^{3+} ions tend to be localized in only one site.

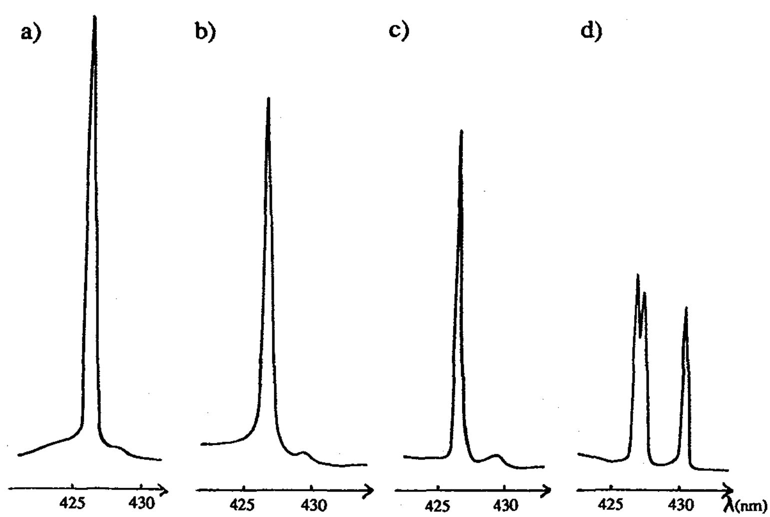
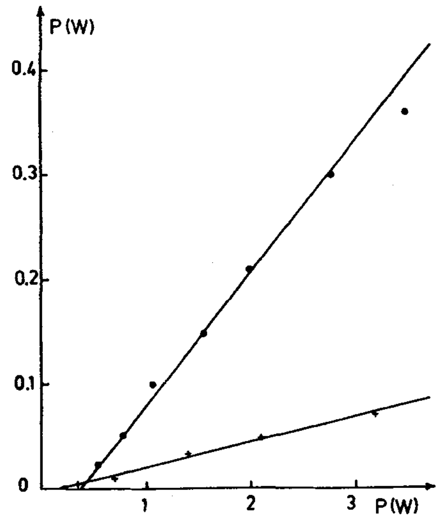


Fig. 1. Absorption spectra at 4K of Nd^{3+} in ASN ($Sr_{1-x}Nd_xMg_xAl_{12-x}O_{19}$)
a) x = 0.2
b) x = 0.4
c) x = 0.6
d) LNA $La_{0.9}Nd_{0.1}MgAl_{11}O_{19}$ ref. /2/

4.- Laser tests.



Laser tests were performed on a sample of $Sr_{0.8}Nd_{0.2}Mg_{0.2}Al_{11.8}O_{19}$, cleaved from a Czochralski boule, with uncoated faces perpendicular to the $\vec{c}$ axis. In this crystal the room temperature neodymium excited state lifetime was about 110 µs. The crystal was CW pumped using the 514.5 nm line of an argon ion laser. Free running laser emission was observed at 1049.8 nm. The slope efficiency was for example 21 % for a 10 % transmitting output mirror (Figure 2). By inserting a Lyot filter in the laser cavity, it was possible like for LNA /1/ to tune the laser emission for about 1.5 nm around 1049.8 and 1061.8 nm, and 12 nm around 1074 nm (Figure 3). To our knowledge, this is the largest tunability range reported, so far for a neodymium activated single crystalline laser material.

Fig. 2. Output power of the ASN laser as a function of the pump power at 514.5 nm (crystal thickness: 7mm; rod // $\vec{c}$). Output mirror transmission: 1-1% 2-10%

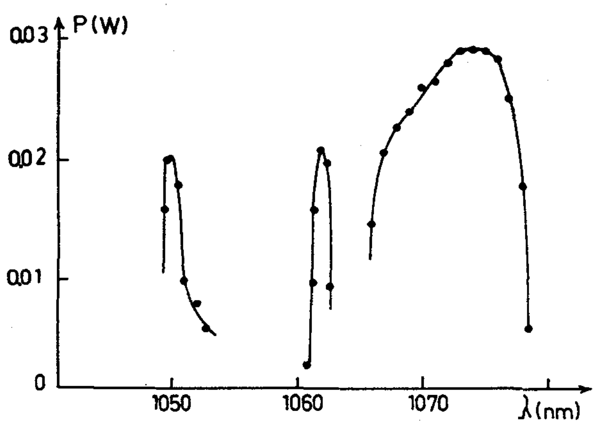


Fig. 3. Tuning curves of the ASN laser (pumping source: Ar^+ laser (514.5 nm))

5.- Conclusion.

ASN presents several originalities:

-The spontaneous crystal growth direction is the optimal one for laser effect ($\vec{c}$ axis).

-Nd^{3+} ions are localized on one site because of the presence of Sr^{2+} ions and of the simultaneous charge compensation by Al/Mg substitution. Attemps to use other charge compensation processes resulted in the distribution of neodymium over several sites as with the parent compound LNA /2/.

-It exhibits a broad tunability range.

It presents also some other advantages:

- High concentration of active ion (Nd^{3+}).
- No segregation of Nd^{3+} during crystal growth.
- Emission wavelength differency from the YAG:Nd one (1.064 nm), and which could be interesting for specific applications.

Further investigation of the laser properties and optimisation of the ASN characteristics are in progress.

6.- References.

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