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Laser emission under flashlamp pumping of a new crystal: Nd-doped strontium-lanthanum-aluminate (Nd:ASL)

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Abstract. Nd-doped strontium-lanthanum-aluminate ($Sr_{1-x}Nd_yLa_{x-y}Mg_xAl_{12-x}O_{19}$) is a new material, tunable in the 1–1.1 μm band. We present the first results of pulsed laser emission of this crystal under flashlamp pumping. We obtain a slope efficiency of 3.3% with a maximum average power of 40 W at 40 Hz in free-lasing regime. Nd:Cr codoping of ASL is also tested, but with much lower efficiency.

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The most well-known and attractive laser crystal is Nd:YAG, which combines very high efficiency and very good thermomechanical properties. Numerous attempts have been made over a long time to find a substitute for this material but it remains very difficult to compete with it. Crystals such as Nd:YAP [1] or more recently studied Nd:KGW [2, 3] present a similar or higher efficiency than YAG but at the expense of worse thermomechanical properties (higher thermal lensing for example), particularly for KGW which suffers from a three times higher thermal dioptric power than YAG [4]. In this never-ending search for new laser materials, we have tested for the first time under flashlamp pumping, a Nd-doped new material: strontium-lanthanum-aluminate (French acronym Nd:ASL or Nd:SLA in English), which is related to the previously known lanthanum-magnesium-aluminate or LMA [5] and strontium-magnesium-aluminate or ASM [6] crystals with a $Sr_{1-x}Nd_xMg_xAl_{12-x}O_{19}$ composition for this last one. In the case of ASL the composition is slightly modified by partly replacing lanthanum by magnesium leading to the formula: $Sr_{1-x}Nd_yLa_{x-y}Mg_xAl_{12-x}O_{19}$. This crystal is Czochralski-grown by Crismatec (Gières, France) who holds the patent. Unlike LMA crystals, ASL as ASM can be grown both along *a* and *c* directions [6], the *c* direction being the preferred one for laser action. ASL has a magnetoplumbite structure where the magnesium ion substitutes for aluminium for charge compensation. For lanthanide aluminates "$LnAl_{11}O_{18}$" with $Ln = La^{3+}, Nd^{3+} \ldots$, the presence of trivalent lanthanides leads to a very marked deficient nature, unfavourable to the stability and to the existence even of the phases. The introduction of M^{2+} ions leading to $LnMAl_{11}O_{19}$ compounds with $M = Mg^{2+}$, Ni^{2+}, $Co^{2+} \ldots$, has generally as a consequence stabilization of the structure and the crystallogenesis of the compounds. The most used ion is magnesium, the $LnMgAl_{11}O_{19}$ phases exist only for lanthanide ions having the largest radii, from gadolinium to lanthanum, the crystalline quality increasing when going closer to lanthanum, the biggest one among them. The compositions are chosen with $0 < x < 1$. As Nd is a constituent ion, the crystal lattice can have a high concentration of this ion, much higher than for Nd:YAG (up to 20%), without risks of fracture. Co-doping with Cr ions can also be obtained.

The first lasing effect in Nd:ASM was obtained by cw argon-ion laser pumping at 514 nm [7]. The authors found an emission spectrum with three tunable humps around 1.050, 1.062, and 1.070 μm, this last wavelength presenting the broadest laser emission range: about 12 nm. More interesting results in terms of efficiency were later obtained under cw diode-pumping at 798 nm [8]. With a Nd concentration as high as 15 at. %, this crystal presented a slope efficiency of 36% and a maximum laser power of 180 mW with a 5-mm-diameter × 5-mm-long crystal. Similar experiments on Nd:ASL under diode-pumping lead to better results with slope efficiencies up to 47% and a maximum output power of 280 mW [9]. In order to illustrate a property of this Nd:ASL crystal: rather large tunability in the 1–1.1 μm range, which makes it attractive, we present in Fig. 1 a fluorescence spectrum of this crystal compared to Nd:LMA and Nd:YAG (after [9]). This tunability may allow us to use this material in a laser oscillator as a pilot for a large amplifying chain made of Nd:glass for example.

We used for the first time large-size crystals of Nd:ASL coming from the same boule grown by Crismatec as the one used for the rod tested under diode-pumping. The Nd^{3+} concentration is 10 mol. % and the rod size is 6.35×101 mm ($1/4'' \times 4''$). The exact composition of the tested rod is $Sr_{0.7}Nd_{0.1}La_{0.2}Mg_{0.3}Al_{11}O_{19}$. The rods are flat-ended, parallel, but without any AR coating on the end faces. These crystals, relatively large compared to the core size of the boule, which is about 3–4 cm in diameter, have subsequently a rather poor optical quality, this is confirmed by the strong deformation of a He-Ne beam after propagation through the

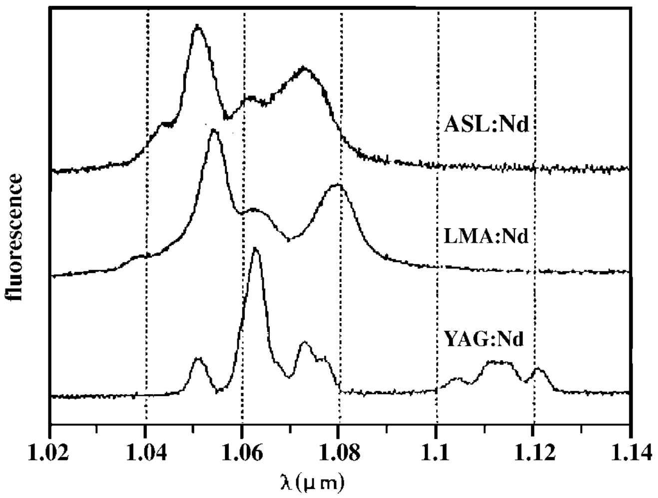

Fig. 1. Nd:ASL, Nd:LMA, and Nd:YAG fluorescence spectra near 1.06 μm (after [9])

rod. Unfortunately these boules were made to obtain small-size crystals suitable for diode pumping and not for large rods as we needed.

These rods were tested under flashlamp pumping and we will present the results obtained at different pump energies and repetition rates up to 50 Hz. We used different cavity configurations with various mirror reflectivities and radii of curvature. By optimizing these configurations we obtained under free-lasing mode, energies up to 1.1 J and average output power of 40 W at 40 Hz. We will present results on the thermal dioptric power of Nd:ASL and on the losses through a Findlay–Clay analysis. Some results on a co-doped Cr-Nd crystal will be presented leading to relatively bad results due to a non-matched concentration of the doping ions for flashlamp pumping.

1 Experimental set-up

The Nd:ASL crystal is set in a commercial single-lamp laser head (Kigre model FE254KK) with a close-coupled diffuse reflector surrounding a double flow tube made of a KTF2 material, allowing a good filtration of the UV lamp emission. The laser head holds a single flashlamp with a cerium-doped quartz envelope, a pumping length of 3.5″ and a Xe pressure of 400 torr.

We use a capacitor-charging power supply (Converter Power Inc. model RCS 1500) with a maximum voltage of 2 kV and a charging power of 1500 J/s at repetition rates up to 100 Hz. The lab-made storage and fast discharge system is made of a PFN (pulse-forming network) with eight similar L-C cells with $C = 4\,\mu$F and $L = 14\,\mu$H. Such a PFN delivers quasi-rectangular current pulses which allows one to obtain a constant lighting level of the crystal by the flashlamp. The L-C cells can be connected in series or in parallel (or suppressed) to obtain the desired pulse duration between 60 and 120 μs. Here in the case of Nd:ASL rod, we use the longer pulse duration but we can note that it is not well matched to the fluorescence lifetime of the crystal which is about 185 μs for a 10 mol. % Nd concentration. A low dc simmer current of about 60 mA maintains the lamp conduction in order to decrease its impedance and to improve laser energy stability and efficiency. Initial lamp ignition is obtained through a parallel discharge in a high-voltage transformer coupled with an automatic detection of the lamp conduction. The PFN discharge is obtained through a high-voltage, high-current thyristor (Semikron, model SKT 520-24E) and presents a very reproducible rectangular shape for the current.

We tested two Nd:ASL rods cut from the same boule but at different locations towards the core, showing a slightly better optical quality of one of the rods. This is confirmed by its laser emission performances, which are about 20% better than with the other one, and we will present now only the results obtained with the better rod. As said in the introduction, the rod has a diameter of 6.35 mm and a length of 101 mm without AR coating on the parallel flat end-faces. As the index of refraction of ASL is quite high: $n_e = 1.759$ and $n_0 = 1.805$ (@ 1067 nm) along the c axis, we expect significant losses on these faces.

The laser cavity is a simple two-mirror cavity, made of a R_{max} concave end-mirror with different radii of curvature, from -3 m down to -0.75 m. The output mirror is plane and we have tested different reflectivities. Due to the high thermal lensing of the ASL crystal it may be necessary to add inside the cavity, above a certain value of pump power, an AR-coated convergent lens (focal length of about 1 m) just in front of the output coupler, in order to stay in the stable part of the resonator under high pump powers.

2 Experimental results

The first experiments were made with a concave R_{max} mirror with a radius of curvature $R = -3$ m and with an output mirror of 60% reflectivity. The laser emission was then very weak and by testing different output mirrors with reflectivities of 60%, 70%, 80%, and 90% we obtained the best results with either the 70% or the 80% reflectivity. These values have been used in the following experiments.

We present in Fig. 2 the output energy at 1.06 μm as a function of pumping energy, obtained at a 20-Hz repetition rate and for three values of the radius of curvature of the R_{max} mirror. We can see that the best results are obtained with the shorter radius of -1 m. Complementary experiments

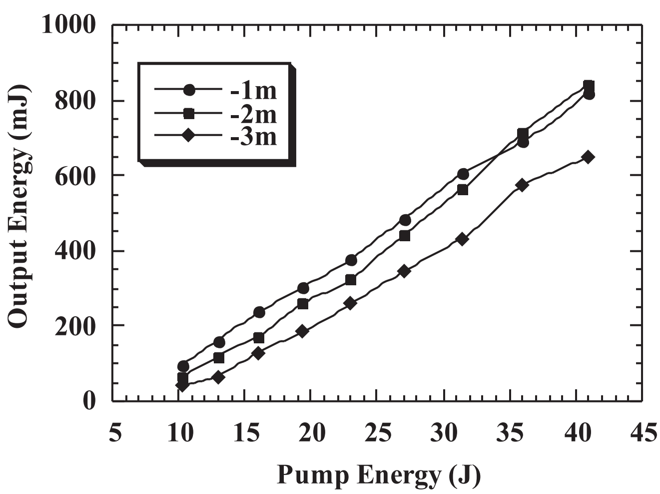

Fig. 2. Nd:ASL output energy at 1.06 μm vs. pumping energy (@ 20 Hz) for three radii of curvature end-mirrors

made with a −0.75-m radius did not show any improvement over the −1 m mirror and are not presented on the figure for clarity. When we increase either the repetition rate above 20 Hz (up to 50 Hz) or the pump energy, there appears quickly a dramatic instability of the resonator for the longer radii of curvature and the −1-m radius allows us to stay in the stable regime only with pump powers up to 1000 W.

By using this −1-m mirror and a plane $R = 70\%$ output mirror we obtain the results presented in Fig. 3 at various repetition rates from 10 to 50 Hz and pumping energies up to 41 J. We can see that the output power stays linear as a function of the pump energy indicating that we do not saturate the material and that the resonator remains stable. The maximum output power in this case is 27 W at a pumping energy of 31 J and a 40-Hz repetition rate, corresponding to a pump power of 1250 W. The total efficiency is then about 2.2% and as the lasing threshold is about 7 J, the maximum slope is close to 2.8%.

At high pumping powers, above 1000 W, we note an inflexion of the output power curve leading to a fast decrease of this power. This indicates a strong thermal lensing effect that can no longer be compensated by the R_{max} mirror concavity: we fall in the instability domain of the resonator. It is then necessary to replace the plane output mirror by a curved one or to put into the resonator a convergent lens set close to the flat semi-reflective mirror. We chose this simpler and more flexible solution and we used a plano-convex lens with a focal length of 1 m. The faces of the lens are AR-coated for 1.06 μm, so it does not introduce any noticeable loss in the cavity. We also replace the −1-m end-mirror by a slightly more curved one having a −0.75-m radius. The output mirror has then a 80% reflectivity for 1.06 μm. We tested several cavity lengths: 30, 35, and 40 cm. The best results are obtained with a length of 35 cm and are presented in Fig. 4 for various repetition rates and we can see that even at low pump energy, the results are much better than without lens. The maximum output power is 39.7 W, obtained with a pump energy of 36 J and at a 40-Hz repetition rate: this corresponds to a pump power of 1440 W. We are very close to the limit of our charging power supply and we do not see any saturation or cavity instability effect.

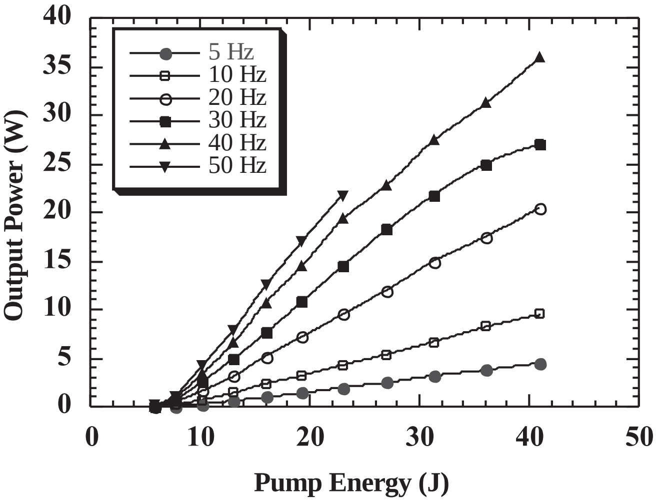

Fig. 4. Nd:ASL output power at various repetition rates with an intra-cavity convergent lens

The lasing threshold is about 4 J and under these conditions, the total efficiency is 2.7% and the slope efficiency is about 3.3%. The maximum energy per pulse is 1.15 J with a pump energy of 41 J. The pulse duration corresponds to the PFN discharge curve and is about 120 μs which is, as mentioned before, not completely well matched to the fluorescence lifetime of the crystal (185 μs for a 10-mol. % Nd concentration.). We note the large increase on the laser performances and efficiency by adding the intra-cavity lens in front of the output mirror. No attempt was made to tune the emission wavelength around 1.076 μm, but future experiments will be made to verify the tunability of this material as found under diode pumping which is an interesting property of these crystals.

We have calculated the optical losses of the cavity through a Findlay–Clay analysis [11]. The curve showing the dependence between the output reflectivity and the laser threshold is given in Fig. 5 for reflectivities varying from 90% to 30%. The optical losses are then calculated and we obtain a value of 29%. They include in addition to the usual losses by diffusion, absorption, etc. the losses due to the lack of AR coating

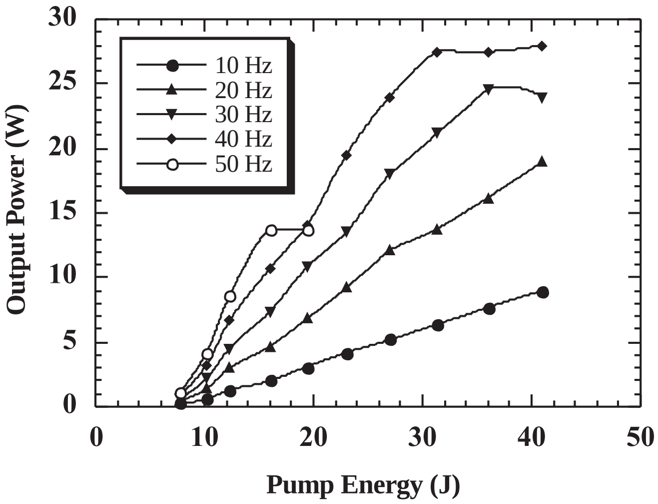

Fig. 3. Nd:ASL output power at various repetition rates without intra-cavity lens (@ 1.06 μm)

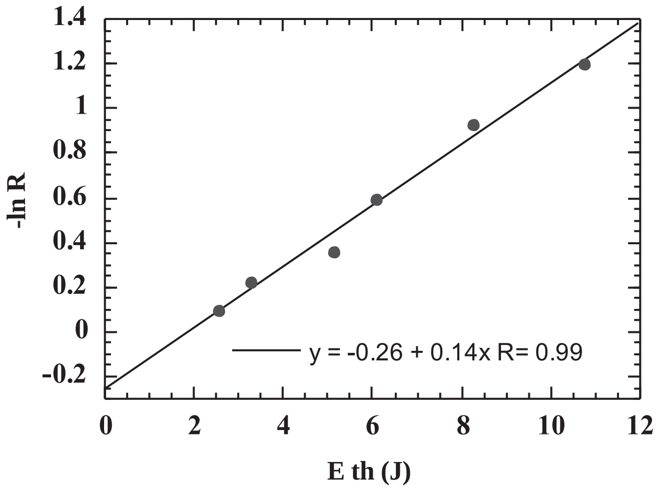

Fig. 5. Findlay and Clay analysis of the resonator

on the rod faces (about 7.6% per face). The pumping parameter is deduced from the data and equal to $6.7 \times 10^{-2}\ \mathrm{J}^{-1}$. As we know the optical losses L, the slope efficiency η and the energy threshold $E_{\mathrm{threshold}}$ for an output mirror of reflectivity R, the fluorescence lifetime τ_{f} and the pump pulse duration τ_{p}, we can calculate the intensity of saturation I_{s}:

$$I_{\mathrm{s}} = \frac{h\nu}{\sigma_{21}\tau_{\mathrm{f}}} = \frac{-\eta E_{\mathrm{threshold}}}{\tau_{\mathrm{p}} A \ln \sqrt{R(1-L)}} .$$

We can also estimate the stimulated-emission cross section. The calculated value is in fact an effective cross section rather than a real measurement. The uncertainty is then relatively high but allows us nevertheless to compare this value to the ones of more well-known crystals. We obtain an "effective" cross section of about $9.5 \times 10^{-20}\ \mathrm{cm}^2$ which is about three times less than for Nd:YAG. The effective cross section × fluorescence duration product is $17 \times 10^{-24}\ \mathrm{cm}^2\,\mathrm{s}$ for Nd:ASL versus $72 \times 10^{-24}\ \mathrm{cm}^2\,\mathrm{s}$ for Nd:YAG. This crystal appears clearly less favourable than Nd:YAG in term of energy storage.

Unfortunately it was impossible to measure the thermal focal length of the rod according to the poor optical rod quality, which did not allow us to use the usual method of thermal dioptric power estimation with the help of a He-Ne laser probe. The He-Ne laser beam was strongly distorted by passing through the rod, even without pumping. Figure 6 presents the slope efficiency as a function of the repetition rate at 36 J of input energy. We observe a variation of 20% between the low value of 2.6% at 180 W and the maximum value of 3% at 720 W. For a pump power higher than 720 W the slope efficiency decreases and falls to 2.7% for a pump power of 1440 W. This variation demonstrates a high thermal dependence of the crystal efficiency and consequentely a high dioptric power.

It was also impossible to obtain a lasing effect in the Q-switched mode either with a passive element (Cr^{4+}:YAG) or with an active Pockels cell, due to the strong depolarization appearing in the material under flashlamp pumping. This is confirmed in free-lasing regime: if we insert a dielectric polarizer in the laser cavity we can note a dramatic decrease of the laser efficiency (slope falling from 3.3% to about 1.2%). In this case the losses were measured at the energy threshold to be equal to 74% (29% for the rod and 45% for the losses introduced by the polarizer). This high value shows the poor optical quality of the rod and is characteristic of the material depolarization with a strong induced birefringence. The nature of this birefringence is partly thermal, as verified by increasing the pumping load of the rod with higher repetition rates, and partly due to the crystal growth.

We had also the possibility to test another ASL rod but codoped with Nd and Cr. Codoping of laser materials is a good possibility to increase the emission efficiency by radiative or non-radiative energy transfer from a sensitizer ion to an activator one [11]. We have tested a Cr:Nd:ASL rod with the following composition: $Sr_{0.7}Nd_{0.1}La_{0.2}Mg_{0.3}Cr_{0.05}Al_{11.7}O_{19}$ cut in a boule of 32 mm in diameter and 170 mm in length obtained by Czochralski method. The rod has similar dimensions to the previous rods and the dopant concentrations given by the manufacturer are: Nd: 10 mol. % and Cr: 5 mol. %. This very high Cr doping level was essentially suited for diode pumping.

The experimental results are presented in Fig. 7 with output mirrors of 80% and 90%. We can note that the efficiency is very low, far away from the expected values resulting from codoping. The maximum output energy is about 60 mJ with a pump energy of 42 J. We present in Fig. 8 the output power vs. pump energy at various repetition rates. Here again the results are very bad, we obtain a maximum power of 1.4 W at a 30-Hz repetition rate with a slope between 0.1% and 0.2%. We have observed a strong red fluorescence around 690 nm (corresponding to a fluorescence peak of Cr). For short times, the fluorescence spectrum of $Sr_{0.7}Nd_{0.1}La_{0.2}Cr_{0.05}$ is made of the Nd emission between 850 and 920 nm and of Cr between 690 and 950 nm, diminished by the Nd absorption around 720–750 nm and 790–850 nm. So there is a radiative transfer from chromium to neodymium, made possible thanks to the good matching between Nd^{3+} absorption and the broad emission band of chromium. For long times, the presence of neodymium modifies strongly the fluorescence spectrum. We then distinguish the weak neodymium fluorescence from 850 to 920 nm and a part of the chromium fluorescence between 720 and 850 nm was removed, which corresponds to the Cr-to-Nd transfer.

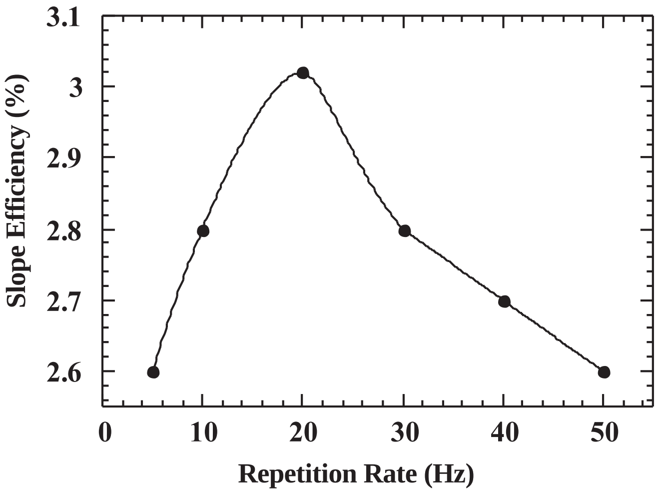

Fig. 6. Slope efficiency of Nd:ASL vs. pump power

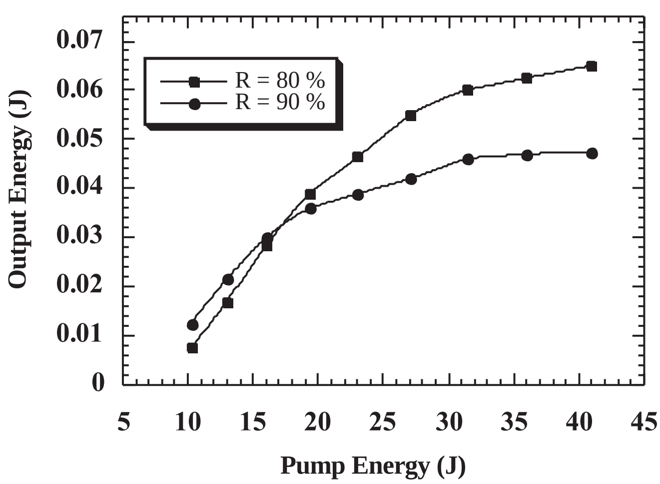

Fig. 7. Cr:Nd:ASL output energy vs. pumping energy at 1.06 μm

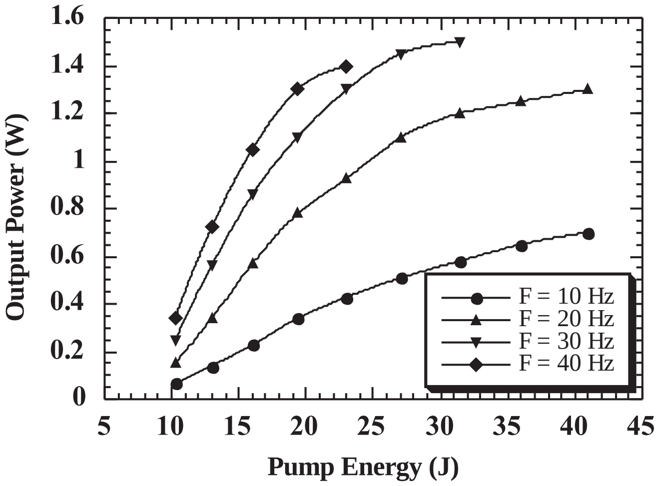

Fig. 8. Cr:Nd:ASL output power for various repetition rates

Moreover the crystal has a deep red colour and is completely opaque, it means that the doping level of Cr introduced in the crystal is too high and inadequate for an efficient transfer, particularly in the case of lamp pumping. However it seems very difficult to estimate the optimum Cr doping level in the lack of knowledge about absorption and stimulated-emission cross sections of Nd in the ASL matrix. Another possibility explaining these disappointing results may be the poorer crystalline quality of the rod, inducing very high optical losses.

3 Conclusion

We have tested for the first time under flashlamp pumping new crystals with Nd doping or Cr:Nd codoping. Despite the poor optical quality of the Nd:ASL crystals obtained from a first-attempt boule, we have observed a rather good lasing efficiency in free-lasing regime, with a maximum output energy of about 1.2 J with a pump energy of 41 J and a maximum power of 40 W at a 40-Hz repetition rate, corresponding to a slope efficiency of 3.3%. The lasing threshold with 70% to 80% output mirrors is low: about 4 J. The Nd:ASL crystal presents a strong thermal effect and to fight this effect it is necessary to use a specially designed laser cavity with a concave end-mirror ($R_{cc} = -1$ or -0.75 m) and a flat output mirror combined with a convergent lens (focal length of 1 m). Moreover this material presents a strong depolarization under thermal loading not allowing us to obtain a Q-switched emission.

We have realized a Findlay–Clay analysis allowing us to obtain an estimated value of the stimulated-emission cross section of Nd:ASL. The value we have calculated is about three times lower than the corresponding one for Nd:YAG.

A Cr:Nd:ASL codoped crystal was also tested but in this case the laser efficiency is very low, with a maximum energy of 60 mJ, a maximum power of 1.4 W at 40 Hz and a slope efficiency less than 0.2%. Inadequate doping level in Cr ions is certainly responsible of these disappointing results and work is to be done to estimate the optimum codoping level.

Nevertheless, Nd:ASL presents some encouraging possibilities and its potential possibilities of tuning emission in the range 1.05–1.08 μm as found under diode pumping, makes it interesting for applications where the fixed wavelength of Nd:YAG can be a problem. An example of possible application could be the use of this crystal for a pilot oscillator tuned to the exact frequency of a high-energy Nd:glass amplifying chain. To investigate all the potentialities of this crystal under flashlamp pumping, it is necessary to obtain from the manufacturer rods of good optical quality in large sizes and with different Nd doping levels, the actual values of 10% to 20% being more devoted to longitudinal diode-pumping. Future work will be done to verify the tunability of this material in the 1–1.1 μm range and also to see possible laser emission in the 1.3-μm range.

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References

1. Kh.S. Bagdasarov, A.A Kaminskii: JETP Lett. **9**, 303 (1969)
2. A. Kaminskii, S.E. Sarkiso, A. Pavlyuk, V. Lyubchenko: Izv. Akad. Naut. (SSSR) **10**, 501 (1980)
3. K.A. Stankov, G. Marowsky: Appl. Phys. B **56**, 239 (1993)
4. O. Musset, J.P. Boquillon: Appl. Phys. B **65**, 13 (1997)
5. R. Collongues, D. Gourier, A. Kahn-Harari, A.M. Lejus, J. Théry, D. Vivien: Annu. Rev. Mater. Sci. **20**, 51 (1990)
6. V. Delacarte, J. Théry, D. Vivien: J. Lumin. **62**, 237 (1994)
7. S. Alablanche, R. Collongues, M. Leduc, A. Minvielle, J. Théry, D. Vivien: J. de Phys. IV, Colloque C7, 275 (1991)
8. S. Alablanche, R. Collongues, J. Théry, D. Vivien, A. Minvielle, M. Leduc, R. Romero, C. Wyon: OSA Proc. on Advanced Solid-State Lasers **13**, 231 (1992)
9. V. Delacarte, J. Théry, J.M. Benitez, D. Vivien: OSA Proc. on Advanced Solid-State Lasers **24**, 123 (1995)
10. D. Findlay, R.A. Clay: Phys. Lett. **20**, 277 (1966)
11. B. Henderson, G.F. Imbusch: In *Optical Spectroscopy of Inorganic Solids* (Clarendon Press, Oxford 1989)