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ON THE DEVELOPMENT OF NEW VUV AND UV SOLID STATE LASER SOURCES FOR PHOTOCHEMICAL APPLICATIONS

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Laser action in the VUV and UV region of the spectrum at 172 nm and at 260 nm had been obtained from $LaF_3:Nd^{3+}$ and $LiYF_4:Nd^{3+}$ crystals respectively when they were pumped with an F_2 pulsed discharge molecular laser at 157 nm.

The fluorescence spectrum of $LiYF_4:Ho^{3+}$ monocrystals using the same pumping source has been obtained as well and it suggests that this crystal can be used for generating tunable coherent VUV and UV radiation. This finding opens new prospects for the development of a new class of solid state VUV and UV laser sources for photochemical applications.

KEY WORDS: Laser Chemistry, Tunable solid-state lasers, Interconfigurational ef transitions.

1. INTRODUCTION

Vacuum ultraviolet (VUV) and ultraviolet (UV) laser sources are useful devices for a variety of applications ranging from photochemistry[1] to photolithography.[2] Especially in the VUV region of the spectrum only very few laser devices operate with limited tunability. Besides the existing laser sources, tunable and coherent VUV radiation has been obtained with a variety of methods such as frequency mixing in gases and metal vapours.[3,4] In addition to the above methods tunable VUV radiation can in principle be obtained from a wide class of solid-state dielectric crystals activated by rare-earth ions[5] due to the allowed d-f radiative transitions.

Laser action from $LaF_3:Nd^{3+}$ (LaF:Nd) at 172 nm has been achieved previously by Waynant and Klein.[6,7] The crystal was pumped by the VUV fluorescence of excited Kr_2* dimers (excited by an e-beam). However the method of excitation of the LaF:Nd crystal was too complicated to be used on a small laboratory scale. Recently[8] we have managed to achieve laser action from the same crystal using as a pumping source an F_2 pulsed discharge molecular laser operating at 157 nm. In this paper we report on the efficient laser action from LaF:Nd and $LiYF_4:Nd^{3+}$ (YLF:Nd) single crystals around 172 nm and 260 nm respectively.

It is evident that the realization of this new pumping scheme opens the way for the wide use of rare-earth activating ions in dielectric crystals for efficient generation of coherent VUV and UV radiation. Therefore the search for new rare earth activated wide band-gap dielectric crystals to be used as potential active media for solid state lasers is of great importance and in this paper we report as well on the interconfigurational $4f^95d$-$4f^{10}$ VUV and UV fluorescence of YLF:Ho^{3+} crystals under irradiation at 157 nm from an F_2 pulsed discharge molecular laser.

The broad band features of the fluorescence in the VUV and UV regions of the spectrum and the photochemical stability of these crystals under F_2 laser pumping suggest that these materials can be used as active media for VUV and UV tunable laser sources.

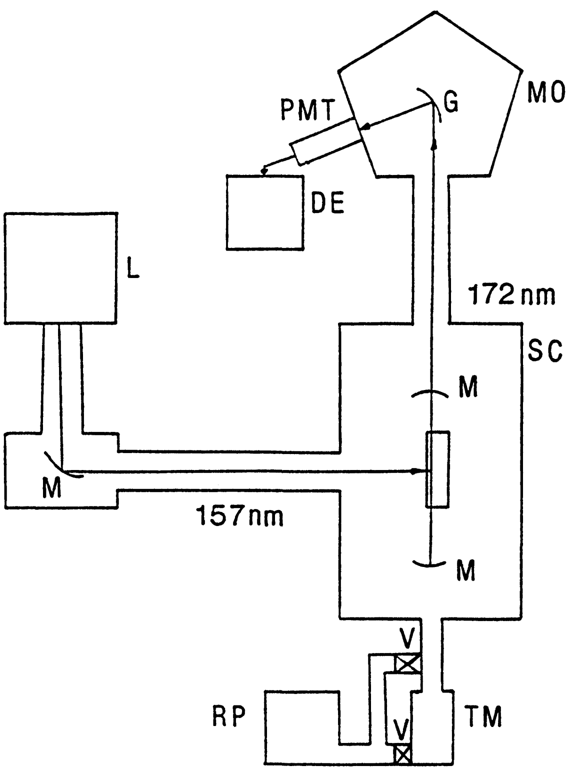


Figure 1 Experimental apparatus for obtaining laser action from $LaF_3:Nd^{3+}$ and $LiYF_4:Nd^{3+}$ crystals at 172 and 260 nm respectively. L: F_2 pulsed discharge molecular laser at 157 nm, M: mirrors, SC: stainless steel vacuum chamber, MO: VUV monochromator, PMT: solar blind photomultiplier, DE: detection electronics, G: grating 1200 lines/mm. TM: turbo molecular pump, RP: rotary pump, V: valve.

2. EXPERIMENTAL

The experimental apparatus is indicated in Figure 1. The laser crystals were cylindrical rods 50 mm long with 3 mm radius and they were placed inside an optical cavity consisted of two mirrors been 150 mm apart. The back mirror was totally reflecting at 172 nm and 260 nm and the front mirrors were 65% transparent at 172 nm and at 260 nm. The laser beam from the F_2 laser was focused on the crystals using an off axis concave mirror at right angles. A polished flat window was made on the side of the cylindrical surface of the crystal for efficient excitation of the pumped medium. The maximum energy density of the pumping source on the surface of the crystal was 120 mJ/cm^2. The optical path of the laser radiation from the pumping source at 157 nm and the excited solid-state laser was kept within stainless steel vacuum lines at 10^{-5} mbar using a turbo molecular pump. The laser output from the solid-state laser was detected using a VUV monochromator with resolution of 0.1 nm and a solar blind photomultiplier. Signals were recorded using a Tektronix 7104 fast oscilloscope and a box-car integrator interfaced to a computer. The F_2 pump laser has been described previously[9,10] and it can deliver 12 mJ (as it is measured with a Gentek PR100-type pyroelectric detector) in a single pulse with 12 ns at FWHM.

3. RESULTS AND DISCUSSION

a) LaF_3:Nd^{3+}

Laser action from the LaF:Nd crystal at 172 nm has been obtained when the crystal was pumped at 157 nm. The concentration of the Nd^{3+} ions was 0.5 at %. The pumping threshold for obtaining stimulated emission was 3 mJ.

Stimulated emission from the LaF:Nd crystal was observed previously by Waynant[7] at 0.1 at % concentration of the Nd ion. The differences between our and Waynant's data are attributed to the different pumping geometry. The transverse pumping geometry of our experiment requires the active medium to have higher absorption coefficients of the pumping radiation, which are unattainable at low doping levels. The temporal evolution of the pumping pulse at 157 nm and the excited laser pulse at 172 nm is indicated in Figure 2. Saturation in the Laf:Nd-laser output vs input dependence is taking place, Figure 3, while the mechanisms of this process are probably similar to those that occur for the YLF:Ce^{3+} laser,[11] such as formation of transient colour centers by excited state absorption of the pumping radiation. The laser output at 172 nm was due to the transitions from the $4f^25d$ mixed configuration, Figure 4, to the $^4I_{11/2,13/2}$ levels of the $4f^3$ configuration of the Nd^{3+} ion.

3b) $LiYF_4$:Nd^{3+}

Laser action at 260 nm has been obtained from the YLF:Nd crystal as well using the same excitation geometry and optical cavity as for the LaF:Nd crystal. The

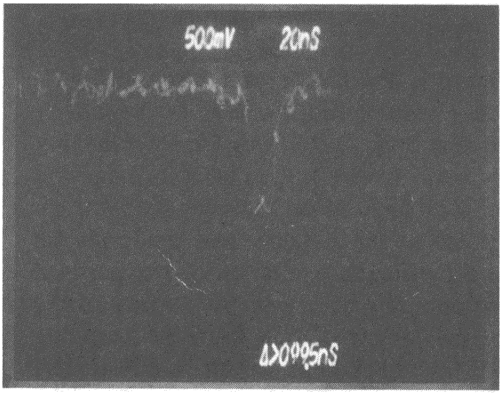


Figure 2a Temporal evolution of the pumping pulse at 157 nm.

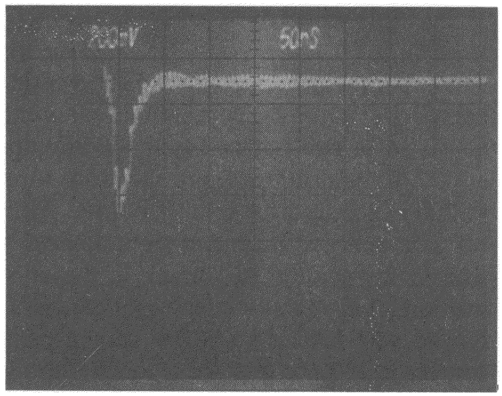


Figure 2b Temporal evolution of the excited laser pulse at 172 nm from LaF_3:Nd^{3+}.

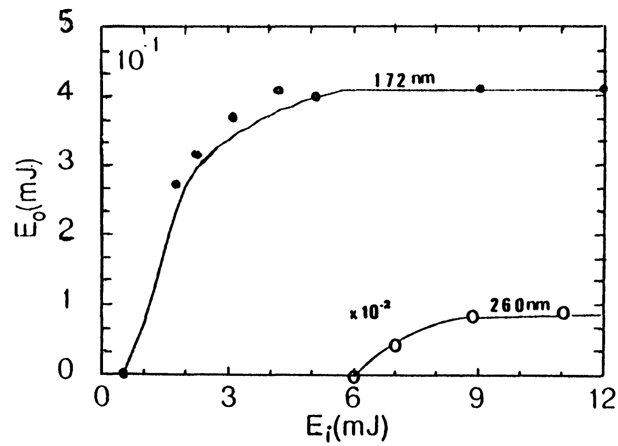


Figure 3 Output energy E_o at 172 nm • and at 260 nm o as a function of the input E_i of the pumping laser pulse at 157 nm.

fluorescence spectrum for this crystal is indicated in Figure 5. Three peaks have been identified in the VUV region of the spectrum around 182, 186 and 196 nm. Emission at these wavelengths corresponds to transitions between the lowest levels of the excited $4f^25d$ mixed configuration and the lowest ground state levels of $4f^3$ configuration, Figure 6. The peak around 182 nm corresponds to the transition between the levels of $4f^25d$ mixed configuration and the $^4I_{9/2}$ ground state level of the $4f^3$ configuration. Similarly the two peaks at 186 and at 196 nm correspond to transitions between the $4f^25d$ mixed configuration and the $^4I_{11/2}$ and $^4I_{13/2}$ levels of the $4f^3$ configuration. The peak around 260 nm can be assigned to transitions between the levels of $4f^25d$ mixed configuration and the $^2G_{7/2,5/2}$ and $^2H_{11/2}$ levels of $4f^3$ single configuration. The threshold for laser action for this crystal was 6 mJ of pumping energy and the maximum output at 260 nm for 0.5 at % concentration of the Nd^{3+} ion was 1 μJ.

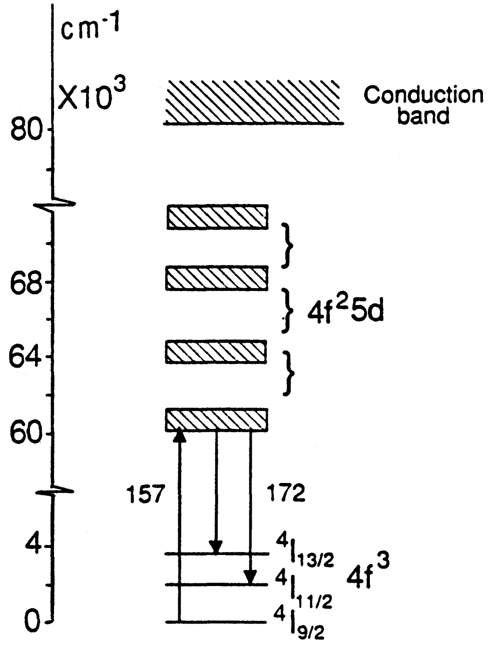
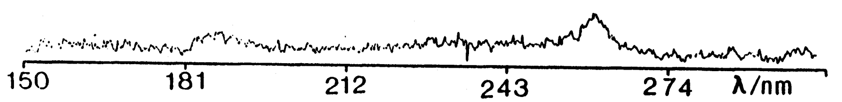


Figure 4 Simplified energy diagram of the Nd^{3+} ions in the LaF_3 host.



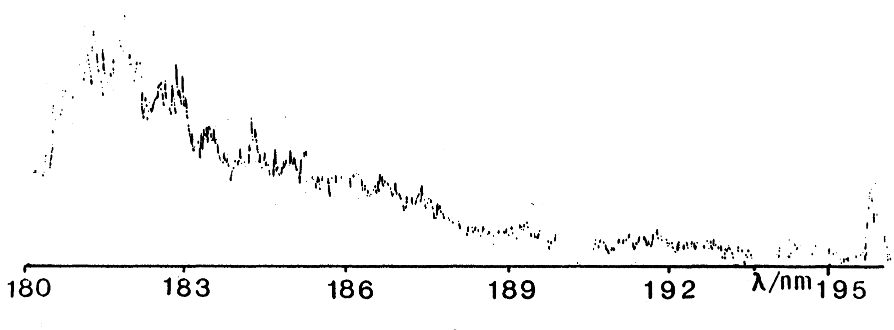


Figure 5 Fluorescence spectrum of the $LiYF_4$:Nd^{3+} under F_2 laser pumping

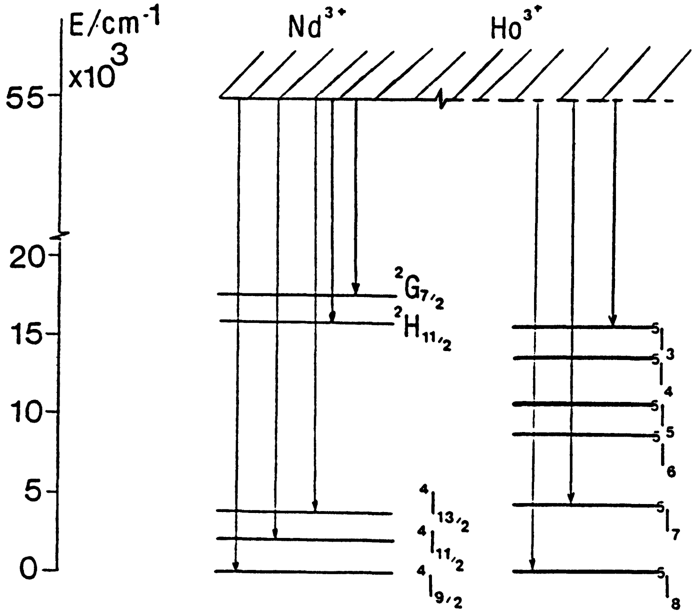


Figure 6 Simplified energy diagram of Nd^{3+} ions in $LiYF_4$ host.

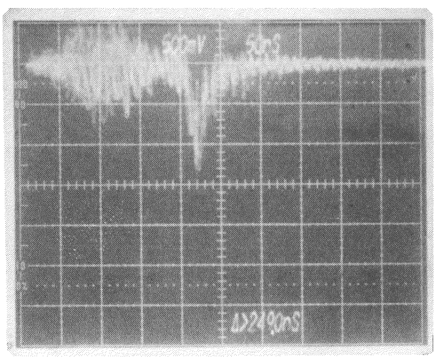


Figure 7 Temporal evolution of the excited laser pulse at 260 nm under F_2 laser pumping of $LiYF_4$:Nd^{3+} crystal.

The temporal evolution of the excited laser pulse is indicated in Figure 7.

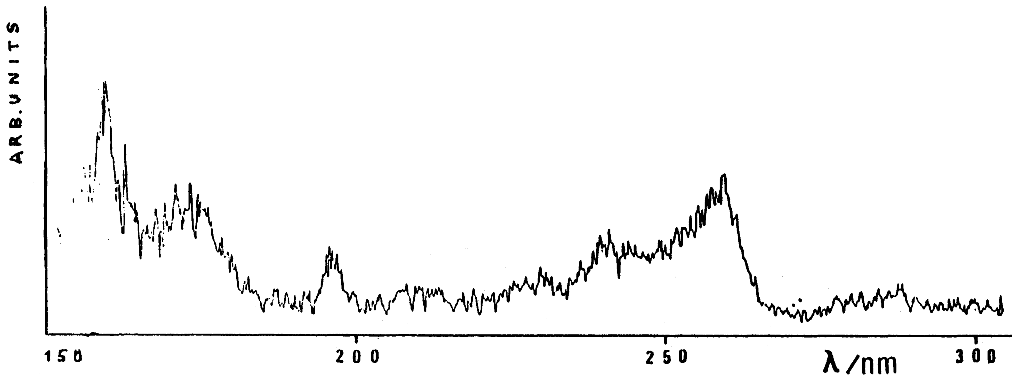


Figure 8 Fluorescence spectrum of $LiYF_4$:Ho^{3+} under F_2 laser pumping.

3c) $LiYF_4$:Ho^{3+}

The laser induced fluorescence spectrum of some rare earth activated YLF crystals have been studied in the spectral region between 157–300 nm. The short pulse duration of the exciting laser pulse with respect to the measured fluorescence lifetime for all the samples on the one hand and the fact that the YLF:Ho^{3+} crystals absorb strongly at 157 nm on the other hand, suggest that the molecular F_2 laser source can efficiently pump these crystals for generating VUV and UV radiation.

The fluorescence spectrum of YLF:Ho^{3+} crystal under the F_2 laser pumping is indicated in Figure 8. Four broad fluorescence peaks have been observed around 170, 194, 240 and 258 nm. Emission at 170 and 194 nm can be tentatively assigned to transitions between the lowest levels of the excited $4f^95d$ mixed configuration and the $^5I_6$ and $^5F_5$ levels of $4f^3$ configuration. The broad bands around 240 and 258 nm can be assigned to transitions between the $4f^95d$ mixed configuration and the $^5G_4$ and $^5G_{3,2}$ levels of $4f^3$ single configuration of Ho^{3+} ion.

The fluorescence spectrum remains unchanged over 10 hours of constant laser irradiation at 157 nm with 10 Hz repetition rate.

The absence of any visible colouration was encouraging for the crystal growth of this material to be used as VUV and UV active laser medium.

4. CONCLUSIONS

Laser action at 172 nm from LaF_3:Nd^{3+} and at 260 nm from YLF:Nd crystals have been obtained when these crystals were pumped with an F_2 pulsed discharge laser at 157 nm. The rare-earth activated dielectric crystals such as YLF:Ho offer the possibility to be used as an active media in tunable VUV and UV laser sources for photochemical applications.

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