**Violet cw neodymium upconversion laser**

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We have observed cw laser action on a new transition ($^4D_{3/2} \rightarrow ^4I_{11/2}$) of the Nd$^{3+}$ ion at 380 nm. This was demonstrated in Nd:LaF$_3$, using an upconversion pumping scheme in which near infrared (788 nm) and visible (591 nm) pump photons from cw dye lasers produce stepwise excitation of the Nd$^{3+}$ ion. In addition, lasing was observed with a single pump source at wavelengths around 578 nm using doubly resonant, sequential absorption of two yellow photons. Single-mode cw operation with an output power of 17 mW was measured at 30 K with 1% output coupling and pump powers of several hundred mW. At 77 K the maximum power dropped to 4 mW.

Upconversion lasers, i.e., those whose output frequency is higher than that of the pump light, are emerging as attractive alternatives to intracavity frequency doubling for short-wavelength generation. Here, we report the observation of cw, upconversion lasing on the $^4D_{3/2} \rightarrow ^4I_{11/2}$ transition of Nd:LaF$_3$ in the violet (380 nm) at temperatures up to 90 K.

Stimulated emission following upconversion excitation was first reported more than 15 years ago by Johnson and Guggenheim using filtered flashlamp pump radiation to obtain red emission from the $^4F_{9/2}$ level of erbium and green emission from the $^4S_{3/2}$ level of holmium. Recently, we reported infrared laser pumping of green 550 nm Er$^{3+}$ lasers in Er:YLiF$_4$ and Er:YAlO$_3$. Spectroscopic investigations of fluorescence upconversion in Nd:LaF$_3$, using both pulsed and cw excitation, have been reported. Fan and Byer proposed the use of Nd:YLiF$_4$ as an upconversion laser at 412 nm, but no lasing was reported.

A crystal of 1% Nd:LaF$_3$ was polished to form a 3-mm-long spherical cavity of radius 2 cm. Coatings directly applied to the crystal surface were highly transmitting at the pump wavelengths (570–800 nm) with an output coupling of 1.0% at 380 nm. In addition, the coatings had sufficiently high transmission between 0.9 and 1.3 μm to suppress laser emission from the $^4F_{9/2}$ state. The laser crystal was placed in a continuous flow cryostat where the temperature could be varied between 10 and 300 K, and cw dye lasers were used as pump sources.

Two different upconversion pumping schemes were used (Fig. 1). The levels are labeled by $^{15/2} \rightarrow ^{11/2}$, and the crystal field components of a given $J$ are labeled in parentheses in order of increasing energy. The first scheme uses two-step excitation with two different frequencies, where the first pump photon around 790 nm (see Table I) is absorbed into one of the components of the $^4F_{9/2} \rightarrow ^4H_{9/2}$ manifolds from which rapid relaxation populates the metastable $^4F_{3/2}$ level whose lifetime at 77 K is 700 μs. The second, yellow, pump photon further excites the system from $^4F_{11/2}$ to the $^4D_{3/2}$ or $^4D_{5/2}$ manifolds from where it relaxes to the $^4D_{3/2}(1)$ upper laser level [Fig. 1(a)]. Note that the frequency of the violet laser is 90% of the sum of the two pump laser frequencies. The two pump beams were combined using polarization coupling, with the polarizations of the IR and yellow light perpendicular to each other. The excitation spectrum of 380 nm laser action, measured in the yellow spectral region, contains two sharp lines at 596.9 and 595.7 nm from transitions to the $^4D_{3/2}(1,2)$ levels, and a band extending from 592.7 to 591.7 nm with a maximum at 591.2 nm. The lifetime of the $^4D_{3/2}$ level has been reported as 35 μs at low temperatures.

Using upconversion excitation in 1% Nd$^{3+}$:LaF$_3$, we measured a nonexponential fluorescence decay with a time constant varying between 20 and 25 μs at 20 K. This suggests some cross-relaxation quenching and is similar to the value measured by Buisson et al. for the excited state of some Nd$^{3+}$ pair levels. Lasing occurs from $^4D_{3/2}(1)$ to the two levels $^4I_{11/2}(7,3)$ at 380.1 nm and 380.5 nm. These transitions are predominantly homogeneously broadened. The 380.1 nm transition has a higher emission cross section and is usually the only lasing line. The 1.5 cm$^{-1}$ width of the emission line is comparable to the 0.1 cm$^{-1}$ separation of the cavity modes so that the laser oscillates in one, or occasionally two, longitudinal modes at 380.1 nm in spite of spatial hole burning. The laser is polarized parallel to the crystal $c$ axis and oscillates in the fundamental TEM$_{00}$ mode.

Table I lists many of the important laser parameters. The stimulated emission cross sections were measured at 30 K from absorption data from the ground state and relative fluorescence from $^4D_{3/2}$ to the ground state and to the final laser level. To avoid reabsorption problems, a sample containing 0.1% Nd was used for these measurements.

Figure 2(a) shows the output power of the upconver-
TABLE I. Parameters of the 380 nm Nd:LaF₃, upconversion laser.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Upper level (D_{3/2}) lifetime</td>
<td>20 μs (30 K)</td>
</tr>
<tr>
<td>Intermediate (G) lifetime</td>
<td>40 μs (30 K)</td>
</tr>
<tr>
<td>Intermediate <em>F</em>₃/₂ lifetime</td>
<td>700 μs (77 K)</td>
</tr>
</tbody>
</table>
| Stimulated emission cross section           | D_{5/2}(1) → I_{1/2}(2): 4 × 10⁻⁸ cm³⁻¹
|                                              | D_{5/2}(1) → I_{1/2}(3): 1 × 10⁻⁹ cm³⁻¹ |
| Threshold for yellow pump                    | 230 mW         |
| Temperature of operation                     | up to 90 K (two-laser pump)  |
| Output                                       | cw, TEM₀₀, see Fig. 2 for power. |
| Wavelengths of operation                     | 380.06 nm (380.52 nm) |
| Pump wavelengths (nm)                        | 577.4, 577.7, 578.8, 579.3 |
| one-laser operation (σ pol.)                 | 577.5, 577.4, 578.8, 579.3 |
| two-laser operation (σ pol.)                 | 596.9, 595.7, 592.7, 581.7 |
| IR (σ pol.)                                  | 787.7, 788.9, 790.5, 791.6 |
|                                              | 792.1, 793.7, 792.5, 794.5 |

*Measured on 1% sample.
*Measured at 30 K (see text).
*Strongest lines.

Laser operation as a function of the two input powers. The 380 nm laser power varies linearly with the yellow and infrared pump power when one of the pump lasers is kept at constant power. The absorption of the infrared radiation is very strong and essentially all of the incident 788 nm light is absorbed in the 3-mm-long laser crystal. With the IR pump laser providing 110 mW, 15% of the 591 nm pump light is absorbed. The lasing threshold is then reached with 27 mW of incident 591 nm which corresponds to an absorbed threshold pump power of 4 mW. Under these conditions, i.e. with a fixed infrared pump power of 110 mW, the slope efficiency is 4.5% when expressed in terms of incident yellow pump power, and 30% when expressed in terms of absorbed pump power. When the yellow pump power is fixed at 280 mW, variation of the infrared pump power, which is completely absorbed, results in a slope efficiency of 10%.

Assuming that each yellow pump photon absorbed populates the D_{3/2} level, the threshold pump power for the second absorption step can be expressed as

\[
p_{th} = \frac{f_i(w_i^2 + w_o^2)\sigma_o(T+1)}{4\sigma_o(T+1)} \times (1 - \exp(-\alpha(T,P_L))),
\]

where \(f_i\) is the fraction of the D_{3/2} population that resides in the lowest component D_{3/2}(1), \(L\) is the cavity loss, \(\tau\) the transmission of the output mirror, \(\sigma_o\) the photon energy of the pump laser, \(\sigma_o(T)\) the temperature-dependent emission cross section, \(\tau(T)\) the lifetime of the upper laser level, and \(1 - L\) the length of the laser crystal. The absorption coefficient \(\alpha(P_L, T)\) for the yellow light depends on temperature and pump power \(P_L\) of the infrared laser. Note that the geometry factor describing the spatial overlap of the laser field and the transition population is exactly valid only for Gaussian distributions for both the inversion and laser profiles with waist \(w_o\) and \(w_{l,2}\), respectively. The waist of the Gaussian laser mode is \(w_o = 22 \mu m\). Due to the difficulty of achieving exact spatial overlap of the two pump beams and the laser beam, the saturation effects associated with the strong absorption of the infrared laser, and the nonlinear excitation mechanism that populates D_{3/2}, the inversion density distribution will be nonuniform both along and transverse to the laser axis. For approximation purposes, we assume a radial excitation distribution that is Gaussian with a waist \(w_o = 10 \mu m\) averaged over the length of the crystal. From Eq. (1) we obtain a reasonably low value of \(L \approx 1\%\) for the laser cavity losses. The onset of the 4f^3 - 4f^2 5d absorption from the ground state occurs at approximately 165 nm in Nd:LaF₃, so that parity allowed excited-state absorption of the 380 nm laser radiation to these levels is expected to be small.

Laser oscillation was observed up to 90 K. At 77 K the highest output power achieved was 4 mW. As temperature increases, two of the factors that reduce the laser performance are the reduction in peak absorption and emission cross sections, and the sharing of population between adjacent thermally accessible levels. Absorption of both IR and yellow pump beams was essentially constant up to 90 K. The stimulated emission cross section was observed to decrease by a factor of 5 between 20 and 90 K. Together with the thermally induced population decrease of the D_{3/2}(1) level, this gives an overall reduction of a factor of 8 in the effective cross section between these two temperatures. This accounts for most of the increase in laser threshold, which was found to be 290 mW at 90 K. Another potentially important source of temperature-dependent loss is the buildup of population in the lower laser levels I_{1/2}(2,3), which are 59 and 90 cm⁻¹ above I_{1/2}(1). The lifetime of I_{1/2}(1) is not known but can be estimated from the energy-gap dependence of the multiphonon emission rates in LaF₃ to be < 5 μs.6,12 The I_{1/2} manifold is populated by relaxation from F_{3/2} and by the laser action itself. We estimate that the upper limit to the steady-state populations of I_{1/2}(2) and I_{1/2}(3) for the temperatures below 90 K and for 380 nm laser powers below

![FIG. 2. Violet output of the neodymium upconversion laser as a function of pump power: (a) two-step excitation with the IR power fixed and yellow power varied and vice versa; (b) single pump laser at 577.7 nm.](image-url)
12 mW. Reabsorption losses should then be negligible for our experimental conditions.

The second upconversion pumping scheme uses a single pump wavelength around 578 nm resonant with one of the components of the 4f_{5/2} \rightarrow 4g_{7/2} absorption. A number of pump wavelengths between 575 and 579 nm resulted in laser operation (see Table 1). Figure 3 shows the results of measuring the spontaneous emission and the onset of laser action when the pump light was turned on with an acousto-optic modulator having a rise time of less than 1 μs. The spontaneous emission rises slowly compared to its lifetime [Fig. 3(b)] showing that the 4d_{5/2} level is being fed by upconversion from the long-lived 4f_{5/2} level [Fig. 1(b)]. Accordingly, the onset of laser oscillation is delayed by about 60 μs with respect to the pump onset [see Fig. 3(c)] and this delay varied between 50 and 200 μs in our experiments depending on the pump power. The delay occurs while population builds up in 4f_{5/2} to the value required for threshold. The relaxation oscillations visible at the onset of lasing in Fig. 3(c) have frequencies between 200 and 400 kHz depending on the excitation intensity. Following the damping of the relaxation oscillations [Fig. 3(c)] the 380 nm laser power continues to rise slowly, since the upconversion excitation rate continues to increase until the population of the long-lived 4f_{5/2} level reaches its steady-state value.

Two processes can lead to population of 4d_{5/2}, both using the 4f_{5/2}, population reservoir. The first is sequential two-step absorption of yellow pump photons, similar to the first pumping scheme but now involving a doubly resonant excitation step: 4i_{5/2} \rightarrow 4g_{7/2}, and 4f_{5/2} \rightarrow 4d_{5/2}. A first yellow photon excites 4g_{7/2}, from which rapid nonradiative relaxation populates the metastable 4f_{5/2} level, a second yellow photon of the same energy excites the system further to the 4d_{5} manifolds. The other process is phonon-assisted energy transfer, involving one ion in the 4f_{5/2}, state and the other in 4g_{7/2}. The 4g_{7/2} level has a lifetime, which we measured to be 40 ns using selective pumping, in agreement with Buisson et al. and much shorter than the value of Ref. 8. This short lifetime favors two-step absorption as the dominant upconversion process. We found, however, evidence that some energy transfer upconversion occurs. In the excitation spectrum of spontaneous violet emission, there was only a factor of three drop in the violet emission when the condition for doubly resonant absorption was not satisfied.

Buisson et al. have studied upconversion processes in Nd:LaF₃ using pulsed excitation in which the 4f_{5/2} level is strongly populated. In this case, upconversion of pair systems via energy transfer dominated that due to single ions. Under our conditions of cw pumping, 4g_{7/2} is only weakly populated. This, together with the double resonance for the transitions 4i_{5/2} \rightarrow 4g_{7/2}, and 4f_{5/2} \rightarrow 4d_{5}, does not selectively favor upconversion of pairs over that of isolated ions. In general, ion-pair states can play a major role in upconversion excitation. Their contribution will depend on the details of the energy levels and on the pumping scheme being employed.

The output power of the 380 nm upconversion laser as a function of a single, π-polarized pump laser power at 577.7 nm is shown in Fig. 2(b). Over the range of pump powers available, the laser power was found to depend linearly on the pump power. This may be an indication of saturation effects due to depopulation of the ground state. Similar results were obtained at three other pump wavelengths: 577.4, 578.8, and 579.3 nm corresponding to 4i_{5/2} \rightarrow 4g_{7/2} transitions. The threshold pump power of 230 mW is considerably higher than that for the resonant two-step pumping with two lasers. Depletion of the yellow photon flux by ground-state absorption is probably a contributing factor to this.

In conclusion, we have demonstrated the operation of a cw, violet, neodymium laser using upconversion pumping schemes. Upconversion excitation by sequential two-step absorption of IR and yellow photons resulted in cw laser oscillation at 380 nm. At 20 K, 12 mW of single-mode, cw, violet output was obtained using several hundred milliwatts of pump power and very conservative output coupling of 1%. At 77 K the output power decreased to 4 mW. In addition, single laser pumping around 580 nm using sequential, doubly resonant absorption results in laser action but with a somewhat higher threshold. Excited-state absorption did not appear to cause large losses for this solid-state laser operating at a violet wavelength. These results indicate that upconversion excitation schemes can offer significant advantages for the pumping of violet and ultraviolet laser transitions, since most of the convenient and powerful pump sources are in the near infrared and visible spectral regions. Such upconversion lasers may become attractive devices for the generation of coherent short-wavelength radiation.