Large Bandwidth Frequency-Converted Nd:Glass Laser at 527 nm with $\Delta v/v = 2\%$

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Large bandwidth in lasers for inertial confinement fusion is potentially capable of controlling laserplasma instabilities. We have demonstrated the efficient generation of large bandwidth, $\Delta v/v=2\%$, in a frequency-doubled Nd:glass laser at 527 nm, using stimulated rotational Raman scattering in atmospheric pressure nitrogen gas. Its use in a fusion laser would involve the use of a multilens array to produce multiple foci where the broadband conversion takes place. This technique is also effective for frequency tripled or quadrupled lasers at 351 or 263 nm.

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It is well known that laser bandwidth can reduce resonant plasma instabilities such as stimulated Raman (SRS) and Brillouin (SBS) scattering that can occur in the long underdense plasmas present in laser fusion targets [1-4].Plasma instabilities can reduce the absorption efficiency of the laser drive and in the case of SRS can generate suprathermal electrons that preheat the fuel and cause the target performance to suffer. By suppressing these instabilities, laser bandwidth can increase the parameter space of efficient target performance, and it can also preserve target performance under conditions where instabilities would otherwise degrade it. For example, laser bandwidth may enable the option of using a wavelength of 527 nm, rather than 351 nm or shorter, and it may enable the use of significantly higher intensities and temperatures than are practical with monochromatic laser drives. The benefits of bandwidth accrue monotonically with increasing laser bandwidth [4].

It is generally believed that to reduce resonant plasma instabilities directly with laser bandwidth requires a bandwidth $(\Delta v/v)$ of several percent [3,4]. However, the bandwidth of the well-established Nd:glass laser at 1053 nm is limited by both the intrinsic bandwidth of the laser medium itself, and the bandwidth of the frequency conversion process from 1053 nm to its second and third harmonics at 527 and 351 nm, respectively. The intrinsic bandwidth is approximately $\Delta v/v=1\%$, and the bandwidth over which frequency conversion is efficient is typically less than 0.1%. A recent development in frequency conversion technology has enabled second harmonic generation with a bandwidth larger than the laser bandwidth (by partially deuterating the frequency conversion material potassium dihydrogen phosphate [5]), but the bandwidth of efficient frequency tripling to 351 nm remains of order 0.1% or less. Achieving greater bandwidths than this will require a new approach. Note that a similar discussion applies to the KrF* laser at 248 nm, which although less mature than Nd:glass technology is being developed as a laser fusion driver. While a KrF* laser does not require frequency conversion, its intrinsic gain bandwidth is somewhat less than 1%, and is comparable to that of Nd:glass [6].

In this Letter we describe and demonstrate a new method for generating large bandwidth that is suitable for use in laser drivers for inertial confinement fusion (ICF). The technique is applied at the output of the laser, and efticiently converts a narrow bandwidth laser to a multiline spectrum covering a total frequency range equal to several percent of the laser frequency. The method is based on parametric stimulated rotational Raman scattering (SRRS) in clean air or atmospheric pressure nitrogen gas [7-20]. The experimental arrangement is shown in Fig. 1. A monochromatic or narrow band laser is converted to almost circular polarization and passes through a long confocal telescope which in our experiment was filled with dry nitrogen gas at atmospheric pressure and room temperature. On passing through the focus the input pulse is modulated by the rotational excitations in the nitrogen molecules, in such a way as to develop a number of frequency sidebands, covering a frequency range of about 10 THz. The strongest sidebands are separated by about 0.5 THz in frequency, corresponding to the known rotational constants of the nitrogen molecule.

In the focused geometry the parametric stimulated ro-

FIG. 1. The layout of the experiment in which a multiline spectrum is generated in the focus of a 5 m $f/600$ lens. This experiment simulates at scale one element of a multilens array suitable for use in laser fusion drivers. A multilens array (discussed in the concluding section) would avoid optical breakdown in the Raman medium.

tational Raman process may be masked by the analogous process involving vibrational excitation of the nitrogen molecules, or by gas breakdown. (The vibrational gain is comparable to the rotational gain under the conditions of our experiment.) The Raman gain is approximately independent of the f/No . of the telescope, whereas the peak intensity varies inversely as its square. Thus there is a minimum f/N o. required to avoid breakdown. For the vibrational process we note that it is more sensitive to the phase mismatch arising from the wavelength dispersion of the refractive index of the gas, and tends to generate sidebands in cones to compensate for the phase mismatch. These cones carry the vibrational sidebands out of the focal region if the f/No . is sufficiently long, and will be suppressed, relative to the rotational process. Thus for a sufficiently long focal length, we expect to eliminate the two main competitors to SRRS. For a 10 mm diameter beam, we estimate the minimum focal length to be 2-3 ^m at 527 nm and 5-6 ^m at 1053 nm.

In our experiment we used both the second harmonic at 527 nm and the laser fundamental wavelength at 1053 nm. We selected the focal length of the telescope to be 5 m at 527 nm and 6 m at 1053 nm. The laser pulse is better than $1.5 \times$ diffraction limited and its pulse length was 1.5 ns. The maximum fluence in this experiment is 2 $J/cm²$, so that the intensity in the focus is less than $I_{\text{max}} = 3 \times 10^{11} \text{ W/cm}^2$. At first we occasionally observed gas breakdown at 1053 nm in the room air, which was not devoid of small particulates. We therefore replaced the room air at the focus with pure dry nitrogen gas, passed through a long open-ended tube containing the focus at its midpoint. No breakdown was observed with the dry nitrogen system. The dry nitrogen system is a good representation of the conditions in the beam tubes of NOVA [15,16) and projected future ICF laser drivers $[21 - 23]$.

The output was dispersed with a grating and recorded with a streak camera. A typical streak record of the output spectrum at 527 nm is shown in Fig. 2. The spectrum contains many lines separated by about 0.5 THz, covering a total bandwidth exceeding 2% of the pump frequency. Nearly all the output occurs in the Stokes lines, and there are evidently no significant anti-Stokes components in the output spectrum. This is in sharp contrast to experiments on SRRS in hydrogen where significant energy is observed blueshifted from the input frequency [9,18]. The lack of significant anti-Stokes lines is not fully understood at this time.

After an initial transient in the nitrogen gas of about 250 ps, the output spectrum essentially appears at full bandwidth. The bandwidth achieved was 2% at a fluence of 2 J/cm². Future laser fusion drivers are likely to operate at higher fluences than this and would be expected to achieve higher bandwidth. The 250 ps transient is associated with the buildup of a coherent molecular rotation in the nitrogen gas, determined by the molecular col-

FIG. 2. A streak camera record of the temporal shape of each frequency present in the output pulse. Note that the entire spectrum appears after about 250 ps, and multiple Stokes orders are produced simultaneously, demonstrating the parametric nature of the process. Note also the absence of anti-Stokes lines. Top: Streak record generated by a 0.8 3, 526 nm, elliptically polarized $(2.2:1)$ input pulse. Bottom: Intensity vs time for the strongest lines, labeled by the frequency shift in cm^{-1} .

lision rate, whereas plasma processes typically appear very rapidly, in a time scale of picoseconds, and hydrodynamic instabilities, which are somewhat slower, are sensitive to the initial behavior of the laser pulse. These processes are highly nonlinear in the laser intensity and are therefore sensitive to the laser pulse shape. They are also strongly dependent on the pulse history through the plasma temperature. The temporal development of the bandwidth must be integrated with the time-dependent plasma conditions to evaluate the effect of the initial transient. Initially the underdense plasma (where the major instabilities occur) has a short scale length which grows at about the ion thermal velocity. Accordingly we expect there to be a small time interval between 10 and 100 ps (depending on the laser pulse shape) at the front end of the pulse where instabilities are suppressed by a

 $.5 -$

 \equiv 1.0 \rightarrow

 (b)

-200 0 200 DIVERGENCE ANGLE (µrad)

FIG. 3. The depletion of energy at the input frequency as a function of the pulse energy. These data are consistent with the interpretation that there is an energy threshold of a fraction of a joule which represents the energy required to generate significant coherent molecular rotation in the gas. After the molecular rotation is established, the input energy is spread over a wide spectrum by the parametric process.

steep plasma profile, and where bandwidth is not needed. If the bandwidth reaches a magnitude sufficient to reduce instabilities within this time then the instabilities will be completely suppressed. The 250 ps transient observed in our experiment implies that there is a small time interval early in the pulse where plasma instabilities are not suppressed by SRRS. However, for the remainder of the pulse the bandwidth would be sufficient to suppress them, by design.

We also note that the 250 ps delay observed in this experiment implies that SRRS will not smooth out early beam nonuniformities that lead to the nonuniform startup problem in direct drive ICF.

Figure 3 shows the output energy at the frequency of the 0.527 nm pump as a function of the input energy. The line with the most energy at the output is the input frequency, and Fig. 3 shows that this line can be depleted by the SRRS process, to about 10% of the input energy, without exciting breakdown in the gas. In the case of strong depletion a small amount of energy remains in the nitrogen gas as rotational excitation. The losses associated with broadband conversion are typically between 1% and 3%. We measured a 4% loss at 10 times the threshold fluence.

Figure 4 shows the far field of the full broadband output in the case of strong depletion. It clearly demonstrates that the focal spot size of all the sidebands is essentially the same as that of the input beam. The near field shows a more complex structure, which in a fusion laser driver must be compensated by using a Raman cell with a magnification slightly less than unity. The full details of the temporal and spatial structure of the output spectrum are too complex to be reported in this Letter

2.0 $-$ 2.0 $-$ 2.0 $-$

 (a)

 $\begin{array}{c}\n\overline{1} \\
\overline{1} \\
0\n\end{array}$

The effect of a multiline spectrum on the resonant plasma instabilities is believed to be very similar to that produced by a broadband continuous spectrum, but the effective increase in the intensity threshold for the instability depends on the line separation and the instability growth rate. If the lines are separated by more than the growth rate of the instability, then each line operates independently of the others and drives just its own plasma wave. If, on the other hand, the line separation is much less than the instability growth rate, the multiline laser behaves as if its spectrum were continuous. The line separation for SRRS in nitrogen is approximately 0.5 THz which is generally faster than filamentation, slightly slower than SBS, and quite slow compared to SRS growth rates in ICF plasmas. Thus we can estimate the effect of a multiline spectrum on both SRS and SBS on the instability threshold and growth rate as similar to that expected for a continuous spectrum [3,4]. For example, in the special case of a convective instability above threshold in a uniform quiescent plasma, the growth rate for the instability is reduced by $1/\Delta\omega$, where $\Delta\omega$ is the total bandwidth of the laser:

$$
\gamma_0' \simeq \gamma_0^2 / \Delta \omega \,. \tag{1}
$$

The threshold is given by

$$
\gamma_0^2 \approx \gamma_1(\gamma_2 + \Delta \omega) \tag{2}
$$

where γ_0 is the threshold, and γ_1 (γ_2) is the larger (smaller) damping rate of the plasma wave and the daughter electromagnetic wave. These factors are modified in a nontrivial manner if the plasma density, velocity, and temperature profiles are not uniform or if they depend on the laser bandwidth. Another effect, which is more difficult to assess, is the effect of the different spatial structure of the different lines on the instability growth rates. This question is currently under investigation, both theoretically and experimentally. However, there appears to be little disadvantage to using multiline spectra compared to continuous spectra in that for closely spaced lines they have similar effects, and even when the line spacing is greater than optimal, there is a substantial reduction in the effective intensity driving the instability.

In conclusion, our experiment has demonstrated at scale that a Raman cell placed at the output of a frequency converted Nd:glass laser will generate a useful multiline spectrum with $\Delta v/v=2\%$. We believe that such a multiline spectrum can be effective in controlling the growth of resonant plasma instabilities, although the initial turn on transient of 250 ps limits its usefulness for early time effects. The development of techniques for reducing the turn on time such as operation at high pressure or low temperature, if feasible, will make SRRS even more effective in suppressing plasma instabilities, and in reducing nonuniform start-up in direct drive ICF. In a multikilojoule laser, optical breakdown in the Raman cell can be avoided by using a "Ay's eye" arrangement where the input and exit optics of the Raman cell are matched arrays of small aperture (1-2 cm) lenses. This brings the beam to many foci whose energy is below the breakdown limit of the gas, while allowing the total beam energy to be unconstrained. The broadband beam must be focused into the target using an achromatic optic to avoid focal spot spreading due to wavelength dispersion.

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