Optical Engineering

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Optical Engineering 54(10), 106104 (October 2015)

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Abstract. A smoothly tunable 1.432 μm Nd∶$YAlO_3$ laser was assembled for potential remote sensing of atmospheric CO_2 at high altitudes. Continuous laser tuning from 6982.8 to 6984.6 cm^{-1} was demonstrated, and CO_2 absorption lines relatively free of atmospheric water absorption interference at 6983 and 6984 cm^{-1} were experimentally observed, confirming feasibility of atmospheric CO_2 sensing. *© 2015 Society of Photo-Optical Instrumentation Engineers (SPIE)* [DOI: 10.1117/1.OE.54.10.106104]

Keywords: optics; light; lasers; neodymium; carbon dioxide; sensing.

Paper 150719 received May 29, 2015; accepted for publication Sep. 9, 2015; published online Oct. 13, 2015.

1 Introduction

Molecules in the atmosphere have characteristic absorption spectra as shown in Fig. 1 calculated using the HITRAN-PC program[1] and 2012 HITRAN database.[2] Water absorption features dominate throughout the 4000 to 10,000 cm^{-1} band. Spectral lines of other molecules often overlap with water lines, so remote sensing measurements of a particular molecule typically involve a compromise between spectral lines of high cross-section and those with minimal interference with water. Remote sensing of carbon dioxide in the atmosphere has typically been accomplished using lasers operating at 5000 cm^{-1} (2 μm)[3] or 6200 cm^{-1} (1.6 μm).[4]

Our innovation is to use the uncommon 1.4 μm, four-level emission lines of Nd^{3+}, a well-established solid-state lasing ion, to directly generate eye-safe output, which overlaps well with a strong CO_2 absorption manifold (∼7000 cm^{-1}). Past differential absorption lidar (DIAL) research has rejected such operation for atmospheric CO_2 detection due to interference from water vapor lines.

Figure 1 shows that the 1.4 μm CO_2 lines, while stronger in absorbance than the 1.6 μm CO_2 lines, might be dominated by strong water absorption features. However, our analysis indicates that operation at high altitudes (e.g., Mauna Loa Observatory at 3.4 km) would reduce the water vapor interference. Some CO_2 lines should be observable at fortuitous spectral locations where water absorption lines are not present or in situations where water concentration is low. Figure 2 shows the HITRAN-PC (2012 HITRAN database) Voigt lineshape predicted atmospheric (mostly water vapor) and CO_2 absorption near 1.43 μm for a 5 km lidar path aimed upward from a Mauna Loa-type altitude. This transmission plot is also suitable for airborne DIAL systems.

Several promising CO_2 lines with low water interference in the 6980 to 6985 cm^{-1} (1.4316 to 1.4326 μm) region are shown in Fig. 2(b). Table 1 shows a compilation of the demonstrated Nd lasing operation in the 1.4 μm region for different hosts.

Most Nd^{3+} hosts have 1.4 μm transitions that are higher in photon energy (shorter wavelength) than the desired 1.432 μm (6983 cm^{-1}) CO_2 absorption line. No literature evidence could be found for Nd-doped YLF and KGW laser operation in the 1.4 μm region. From inspection of Table 1, both Nd:YAG and Nd∶$YAlO_3$ have emission lines in the range of 1.430 to 1.434 μm (6993 to 6974 cm^{-1}). However, there is variation in the reported lasing lines in the literature; so it is not certain which Nd transition will best match up with the CO_2 absorption. As reported by Kaminskii,[5] the Nd:YAG line at 1.432 μm would seem to be a better match than the 1.4338 μm line of Nd∶$YAlO_3$. However, Němec et al.[6] report the Nd∶$YAlO_3$ lasing value as 1.4328 μm. The 1.432 μm spectroscopic Nd:YAG line reported by Kaminskii[5] is also reported by Marling[7] to show no evidence of lasing while the neighboring lines at 1.414 and 1.444 μm do. Hence, we selected Nd∶$YAlO_3$ for this investigation to avoid the uncertainties with the usefulness of the 1.432 μm line in Nd:YAG and to avoid the stress-induced birefringence issue with YAG. Here we report experimental confirmation of Nd∶$YAlO_3$ tunable lasing as well as sensing of CO_2 absorption lines in the 1.432 μm spectral region.

2 Experimental Setup

Demonstration of Nd∶$YAlO_3$ tunable lasing in the 1.43 μm region was achieved using the setup shown in Fig. 3. The pump source was an 808 nm, fiber-coupled laser diode with a maximum power output of 15 W (Oclaro MEA200-808-15-001). Radiation from this diode was collimated and focused using two anti-reflection (AR)-coated plano-convex lenses. The laser cavity consisted of two fused silica mirrors M1 and M2 separated by 175 mm, with radii of curvature of 200 and 150 mm, respectively. Both mirrors were coated for >97% transmission at 808 nm, >99.9% reflection between 1420 and 1440 nm, and <1% reflection at 1080 nm. This last specification was chosen to suppress lasing of the highest gain line of Nd∶$YAlO_3$ at 1080 nm. Our lasing medium was a 0.8% doped crystal of Nd∶$YAlO_3$, oriented with the *b* axis in the laser propagation direction and the *c* axis parallel with

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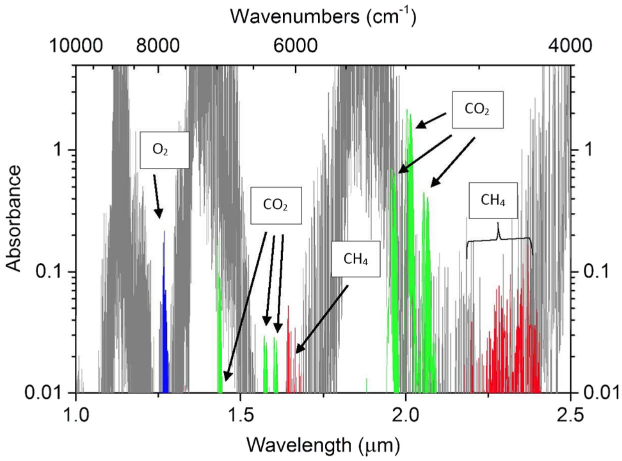

Fig. 1 Absorbance spectra calculated using HITRAN-PC program and 2012 HITRAN database for a mid-latitude winter atmosphere (1000 m horizontal path) in the 4000 to 10,000 cm^{-1} (1 to 2.5 μm) spectral region. The predominate water lines are not labelled.

the laser polarization (horizontal). The crystal had a square cross-section of 5 mm by 5 mm, and a length of 12 mm, with 1 deg of wedge on the input and output faces to prevent parasitic lasing. These faces were AR coated for 808 nm to maximize pump transmission, for 1080 and 1340 nm to further reduce the probability of parasitic lasing, and for 1420 to 1440 nm to reduce losses for our laser wavelength. The crystal was mounted in a passively air-cooled heat sink.

Lasing line selection and coarse tuning were achieved by inserting a birefringent filter (BRF) into the cavity. Rutile was selected as the BRF material after calculating birefringent tuning of several materials based upon equations published by Preuss and Gole.[8] We selected rutile because it has a relatively low tuning rate (3.5 nm/ deg), facilitating easy control with standard rotation stage resolution. Additional factors in our choice include an appropriate free spectral range of 17.8 nm for CO_2 spectroscopy with a filter of reasonable thickness (0.5 mm), and ready availability. Rutile is a uniaxial crystal, and we used a ⟨100⟩-cut plate so that the ordinary and extraordinary axes were both in the plane of the plate. The BRF was mounted at 5 deg less than Brewster's angle. Since our cavity mirrors had no output coupling, we used light ejected from the two faces of the BRF to achieve laser output coupling. By rotating this filter around its normal, lasing was selectively achieved at 10 distinct laser lines between 1328 and 1433 nm. FWHM linewidth was 0.11 nm (0.54 cm^{-1}). Further linewidth narrowing and fine wavelength tuning were then achieved using an intracavity angle-tuned 0.4 mm thick uncoated YAG etalon.

The output power from one of the two BRF faces is shown in Fig. 4 to tune smoothly over the 1.43173 to 1.43210 μm (6982.8 to 6984.6 cm^{-1}) single-line tuning range used for our CO_2 spectroscopy experiment. The diode pump power was set to 6.4 W for the duration of this experiment. We used an HP 86142A optical spectrum analyzer (OSA) to measure the spectral output of our laser; HP 86142A had not been recently calibrated to an external light wavelength, so it had (based on the manufacturer's specifications) an absolute accuracy of up to 0.5 nm (2.4 cm^{-1}) and a relative reproducibility accuracy of ∼0.003 nm (0.015 cm^{-1}).

3 Nd:YAlO$_3$ Tuning and CO_2 Absorption Results

To demonstrate feasibility of CO_2 DIAL at 1.43 μm, we used an absorption cell consisting of a 1.2 m long PVC tube with end windows of 15 μm thick plastic film stretched across the ends of the tube. The cell was filled with 1 atm of CO_2 gas. The output from our laser was transmitted through the CO_2 cell, and the laser power was measured before and after passing through the cell. We set the BRF to allow lasing in the 1432 nm region, and tuned within this region using the etalon. Figure 5 shows the transmission of the gas cell. The expected transmission calculated with HITRAN-PC is also shown for a 1.2 m path of 1 atm of CO_2 at the R16, R18, and R20 CO_2 lines. Calculated[2] CO_2 line peaks, line strengths, and linewidths as well as corresponding experimental measurements are listed in Table 2. It should be noted that the calculated ∼0.2 cm^{-1} linewidths are for CO_2 self-broadening at 1 atm pressure; normal 1 atm pressure

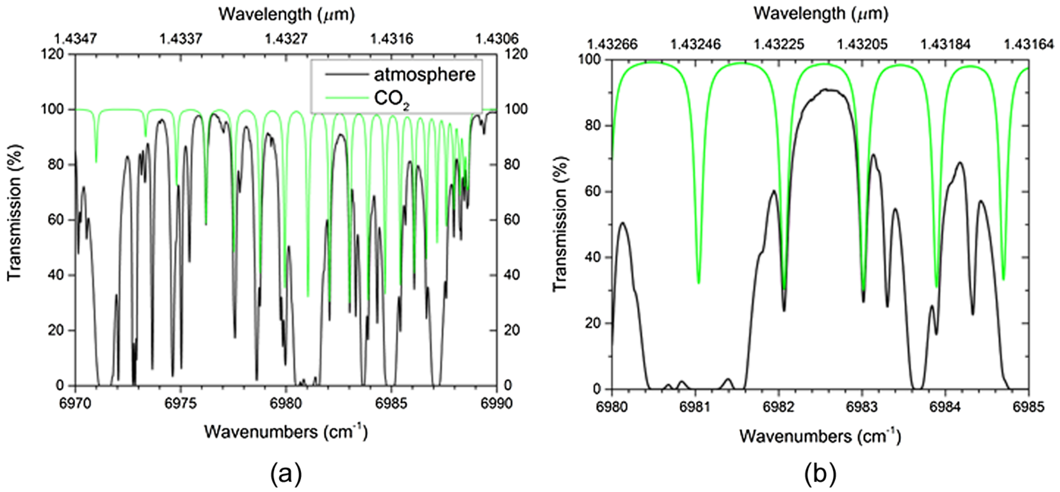

Fig. 2 (a) Atmospheric transmission for 5 km path aimed upward from 3.4 km altitude in 6970 to 6990 cm^{-1} region; black lines are composite total spectrum, and green lines are CO_2 contributions only. (b) Expanded scale 6980 to 6985 cm^{-1} region. Calculations were done using the HITRAN-PC program and 2012 HITRAN database.

Table 1 Nd laser lines operating near 1400 nm at room temperature.

Host material	Wavelength (nm)	Wavenumber (cm^{-1})	Reference	Comments
YAG	1338.1	7473.28	Ref. 5	Pulsed
YAG	1356	7374.63	Ref. 7	Continuous-wave (cw)
YAG	1414	7072.14	Ref. 7	cw
YAG	1432.0	6983.24	Ref. 5	Spectroscopy only
YAG	1444	6925.21	Ref. 9	cw
$YAlO_3$	1340	7462.69	Ref. 10	Pulsed
$YAlO_3$	1341.3	7455.45	Ref. 5	cw and pulsed
$YAlO_3$	1430	6993.01	Refs. 9 and 11	cw
$YAlO_3$	1432	6983.24	Ref. 12	Pulsed
$YAlO_3$	1432.8	6979.34	Ref. 6	Pulsed
$YAlO_3$	1433.8	6988.12	Ref. 5	Spectroscopy only
GGG	1423.4	7025.43	Ref. 13	cw
GSGG	1422.5	7029.88	Ref. 14	cw
GSAG	1421.1	7036.80	Ref. 14	cw
YSGG	1422.5	7029.88	Ref. 14	cw, but pulsed with codoped Cr^{4+}
KGW	<1400	>7143	Ref. 10	No lines >1380 nm
YLF	1321	7570.02	Ref. 15	No lines >1321 nm

broadening of CO_2 in air is ~0.15 cm^{-1}. As can be seen in Fig. 5, the experimental and calculated values of CO_2 absorption lines agree quite well. There is an ~0.23 cm^{-1} difference in experimental and theoretical absorption peaks, but this is well within the 0.5 nm (2.4 cm^{-1}) uncalibrated absolute accuracy of the OSA. Note that the 6983.01 cm^{-1} R16 CO_2 line lies at a location with reduced water line interference in Fig. 2.

From Fig. 5, the measured transmission for the background level (in air) was a value of ~0.76 due to 24% absorption from the plastic film windows. The measured transmission at the line center/peak absorption for the left R16 CO_2 line is a value of ~0.45. The transmission is thus $T = 0.45/0.76 = 0.59$ (which is a 41% absorption at line center).

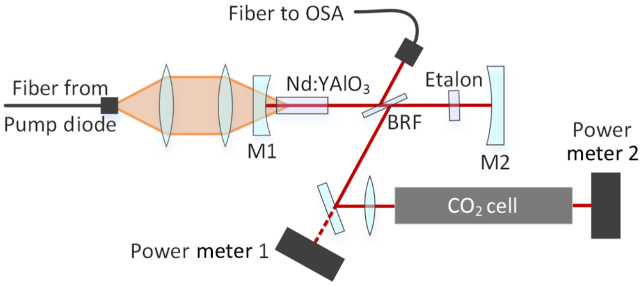

Fig. 3 Nd:$YAlO_3$ laser and CO_2 transmission measurement schematic. OSA, optical spectrum analyzer; BRF, birefringent filter; M1 and M2, laser resonator mirrors.

In order to compare the measured spectrum to the calculated spectrum, the data of Fig. 5 were rescaled in Fig. 6 for 100% transmission relative to the window loss and also shifted in wavelength for peak alignment with the known HITRAN line positions. For both the left R16 CO_2 line near 6983.01 cm^{-1} and the right R18 CO_2 line near

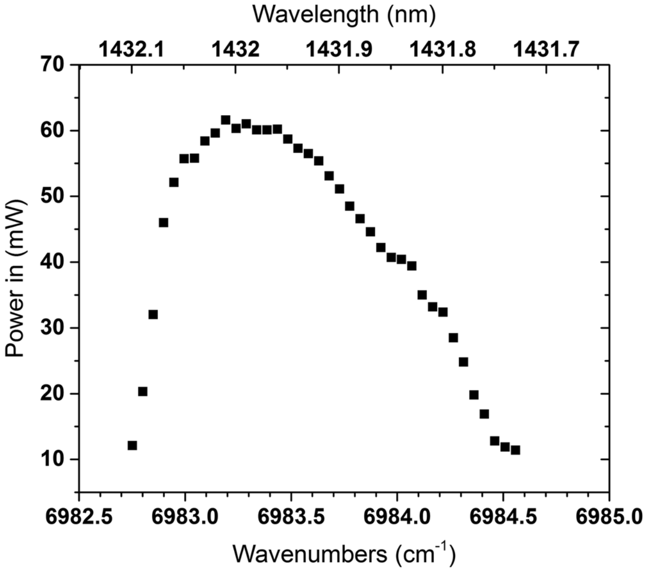

Fig. 4 Nd:$YAlO_3$ laser power versus wavelength, which was etalon tuned across the 1432 nm emission line.

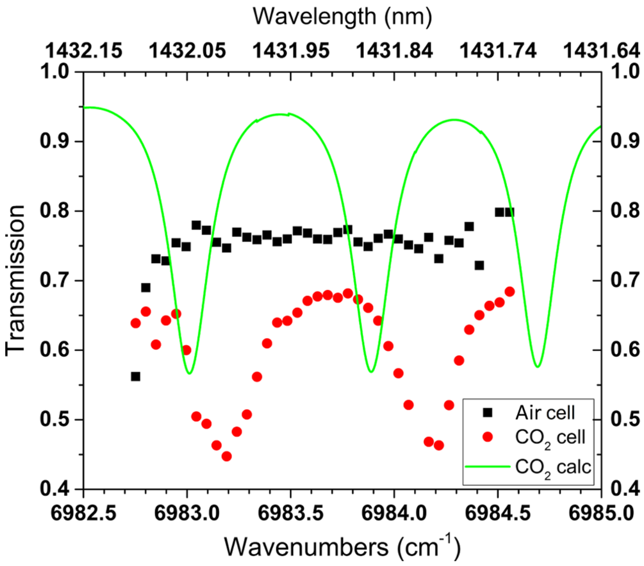

Fig. 5 Transmission of Nd:$YAlO_3$ laser through 1.2 m absorption cell filled with 1 atm of CO_2 as laser wavelength was smoothly tuned and comparison with CO_2 calculated spectrum; room air alone shows no spectral features.

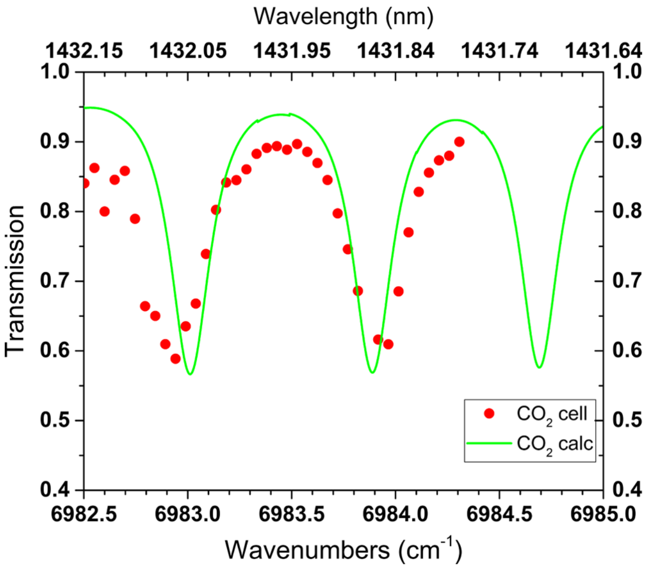

Fig. 6 Comparison of line strengths and linewidths for calculated and measured CO_2 absorption lines. The 24% window loss was factored out of the experimental values and experimental wavenumber values were shifted 0.23 cm^{-1} to match the calculated 6983.9 cm^{-1} peak.

6983.89 cm^{-1} in Fig. 6, the HITRAN-PC prediction is $T = 0.57$ at line center (appropriate for a single frequency laser with linewidth of <0.001 cm^{-1}). Our measured absorption is slightly less than that predicted (see Table 2 for a numerical comparison) and the lines are slightly wider than the predicted lines; we suspect this may be due to laser line broadening. For pressure broadened line measurements, the peak linecenter transmission is related to the pressure broadened linewidth by $T = \exp[-A * S/(\Delta\nu)]$, where A is a constant (path length, concentration) and $\Delta\nu$ is the pressure broadened linewidth.[1] The experimental linewidth is approximately the square root of the combined sum of the squares of the CO_2 linewidth and laser linewidth. This increased linewidth leads to reduced absorbance (and increased transmission); we can use this information to estimate the laser linewidth. Using an increase in peak transmission from 0.57 to 0.59 for R16 (0.57 to 0.61 for R18) and the R16 CO_2 self-broadened linewidth of 0.204 cm^{-1} (0.203 cm^{-1} for R18), we estimate the laser linewidth FWHM to be on the order of 0.08 cm^{-1} near R16 (0.11 cm^{-1} near R18).

The laser cavity length of 175 mm, 169 mm of that in air, corresponds to a longitudinal mode separation of 0.03 cm^{-1}. Thus, both CO_2 absorption linewidth measurements above indicate that the laser probably operated on three to four laser modes for an FWHM laser linewidth of ~0.08 to 0.11 cm^{-1}. It is interesting to note that laser power and linewidth could be better stabilized in future studies by including a PZT drive on the cavity mirror to better allow single frequency operation over the 0.2 cm^{-1} tuning range required for a CO_2 line. Motorized control and tracking of the BFT and etalon could also provide improved wavelength accuracy during a scan.

Table 2 R-line calculations and measurements for 100% CO_2 at 1 atm.

	R16	R18	R20	Comments
Line position (cm^{-1})	6983.01	6983.89	6984.69	Calculated
	6983.2	6984.2		Measured
Line strength S (cm^{-1} mol^{-1})	5.980E-23	5.858E-23	5.580E-23	Calculated
Self-pressure broadened linewidth FWHM (cm^{-1})	0.204	0.203	0.199	Calculated
Line center transmission (%)	57	57	57.5	Calculated
	45	46		Measured (raw)
	59	61		Measured (corrected for window losses)
Line center absorption (%)	43	42.8	42.5	Calculated
	41	39		Measured (corrected for window losses)

4 Summary

We have demonstrated continuously tunable laser operation of an Nd:$YAlO_3$ laser throughout the 6982.8 to 6984.6 cm^{-1} (1.43173 to 1.43210 μm) region. We have also measured CO_2 absorption lines in this spectral region, thus demonstrating feasibility of using 1.432 μm Nd:$YAlO_3$ DIAL for atmospheric CO_2 remote sensing. Relative to the state of the art, our approach may offer higher efficiency and power scaling since it is a true four-level laser. Hence, it may be suitable for air and space platforms and free-space optical communications, including higher pulse energy, average power, beam quality, and spectral properties.

Acknowledgments

We gratefully acknowledge financial support for this work by NASA-Goddard Contract NNX14CG48P.

References

1. D. K. Killinger, W. E. Wilcox, and D. Pliutau, "HITRAN-PC: 25 years of academic development and commercialization of laser atmospheric transmission software for environmental remote sensing," *J. Technol. Innov.* **14**, 303–327 (2012).
2. L. S. Rothman et al., "The HITRAN 2012 Molecular Spectroscopic Database," *J. Quant. Spectrosc. Radiat. Transfer* **130**, 4–50 (2013).
3. U. N. Singh et al., "Development of a pulsed 2-micron integrated path differential absorption Lidar for CO_2 measurement," *Proc. SPIE* **8872**, 887209 (2013).
4. A. Ramanathan et al., "Spectroscopic measurements of a CO_2 absorption line in an open vertical path using an airborne lidar," *Appl. Phys. Lett.* **103**, 214102 (2013).
5. A. A. Kaminskii, *Laser Crystals: Their Physics and Properties*, Springer-Verlag, Berlin (1990).
6. M. Němec et al., "Multiline possibility of Nd:$YAlO_3$ laser in spectral range 1.3–1.5 μm," *Proc. SPIE* **8959**, 895920 (2014).
7. J. Marling, "1.05–1.44 pm tunability and performance of the CW Nd^{3+}:YAG laser," *IEEE J. Quant. Electron.* **14**, 56–62 (1978).
8. D. R. Preuss and J. L. Gole, "Three-stage birefringent filter tuning smoothly over the visible region: theoretical treatment and experimental design," *Appl. Opt.* **19**, 702–710 (1980).
9. H. M. Kretschmann et al., "High-power diode-pumped continuous-wave Nd^{3+} lasers at wavelengths near 1.44 μm," *Opt. Lett.* **22**, 466–468 (1997).
10. R. Moncorgé et al., "Nd doped crystals for medical laser applications," *Opt. Mater.* **8**, 109–119 (1997).
11. S. Yiou et al., "High-power continuous-wave diode-pumped Nd:$YAlO_3$ laser that emits on low-gain 1378- and 1385-nm transitions," *Appl. Opt.* **40**, 3019–3022 (2001).
12. S. Wang et al., "Pulsed Nd:YAP laser at 1432 nm pumped with high power laser diode," *Opt. Commun.* **283**, 2881–2884 (2010).
13. H. Zhang et al., "High-efficiency continuous-wave Nd:$Gd_3Ga_5O_{12}$ eye-safe laser operating at 1423.4 nm," *Appl. Opt.* **52**, 5469–5472 (2013).
14. A. l. Zagumennyi et al., "A comparison of diode pumped 1.4 mm lasers based on the different compositions Nd^{3+}-doped garnet crystals," presented at the CLEO Europe2003.
15. C. C. Andressen, "A 1.32 micron, long-range, solid-state, imaging LADAR," *Proc. SPIE* **1694**, 121–131 (1992).

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