UCRL-JC-123041
PREPRINT
CONF-960163-10

Applications of High Average Power Nonlinear Optics

S. P. Velsko
W. F. Krupke

This paper was prepared for submittal to the
Photonics West '96 Symposium
San Jose, California
January 27 - February 2, 1996

February 5, 1996

Lawrence Livermore National Laboratory

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Applications of high-average power nonlinear optics

Stephan P. Velsko and William F. Krupke

Lawrence Livermore National Laboratory
University of Caliornia
Livermore CA 94550

ABSTRACT

Nonlinear optical frequency convertors (harmonic generators and optical parametric oscillators are reviewed with an emphasis on high average power performance and limitations. NLO materials issues and NLO device designs are discussed in reference to several emerging scientific, military and industrial-commercial applications requiring ≈ 100 watt average power level in the visible and infrared spectral regions. Research efforts required to enable practical ≈ 100 watt class NLO based laser systems are identified.

Key words: nonlinear-optics; harmonic generators; optical parametric oscillators; phase matching; thermal dephasing; thermal gradients; optical loss

1. AVERAGE POWERS OF CURRENT NLO CONVERTORS

The past decade has witnessed sweeping advances in the science and technology of solid state lasers: discrete wavelength high power laser crystals (Nd:YAG, Nd:YLF, Tm:YAG, Ho:Tm:YAG, etc.); novel tunable laser crystals (Ti: Sapphire; Cr:LiSAF, etc.); semiconductor laser diode pump sources; complex, high damage threshold coatings; and high performance nonlinear optical materials (KTP, BBO, LBO, $ZnGeP_2$, etc.) for harmonic gerneration and optical parametric oscillation these advances have enabled the developmant of a variety of laboratory and commercial solid state laser devices and systems able to perform an ever-expanding array of scientific military, commercial and insustrial applications.

Figure 1 plots the output average power extrema of several laboratory and commercial laser systems operating at the 2nd, 3rd, and 4th harmonics of Nd:YAG (532, 355, and 266 nm). Generally speaking[1], there are several commercial laser products that employ electro-optic Q-switches and one or more external harmonic generators, delivering average powers of up to 40 watts at 532 nm, 25 watts at 355 nm, and 4 watts at 266 nm. Operating pulse repetition rates vary from 10 - 100 Hz; puse duration is typically 10 nsec. Another class of commercial lasers[2] utilizes an internal acousto-optic Q-switch and an internal frequency doubler, delivering an output power up to 25 watts at 532 nm with pulse repetition rates varying from several to 25 kHz; pulse duration is typically 100 - 250 nsec.

Several green lasers with average powers ≈ 100 watts have been demonstrated in the laboratory[3-5]. Dane, et. al. have reported a lamp-pumped Nd: glass slab laser system producing 15 joules at 527 nm in a 10 nsec pulse using a KD*P doubler, at 3

Hz (60 watts averape power)[3]; the system is near diffraction limited and has a 60 meter coherence length. Velsko, et. al. reported achieving 100 watts of 532 nm output from an electro-optically Q-switched diode-pumped Nd:YAG power oscillator (250 watts, 2500 Hz, 10 nsec) using an external KTP doubler[4] . St. Pierre, et. al. reported producing 5 joules at 532 nm in an 8 nsec pulse using a diode-pumped Nd:YAG slab mopa chain, and an external KTP doubler[5]. The output was near diffraction limited. Ortiz, et. al.[6] reported obtaining 97 watts of green output using AO modulation and internal harmonic generation in KTP, with pulse repetition rates up to 25 KHz. Over 23 watts of average power were also obtained at a wavelength of 659 nm (double the 1320 nm wavelength of Nd:YAG.)

Although the revival of interest in all solid-state tunable laser systems based on optical parametric oscillators is only a few years old, remarkable increases in the average powers of these devices have already been made. Generally speaking, tunable OPO's in the near IR have generated output in the ≈ 10 watt range, while those in the mid-IR, as well as those in the visible and ultraviolet have reported output powers as high as a few watts. Figure 2 collects some recent results[7-10] for laboratory OPO demonstrations in the "eyesafe" 1.5 μm and the mid-IR 3.5 μm regions. Since all of these results involve 1.06 μm pumping, they necessarily generate light in both these regions. However, we group them only according to the primary wavelength at which the power was measured, presuming that the devices were designed to optimize the output at these wavelengths.

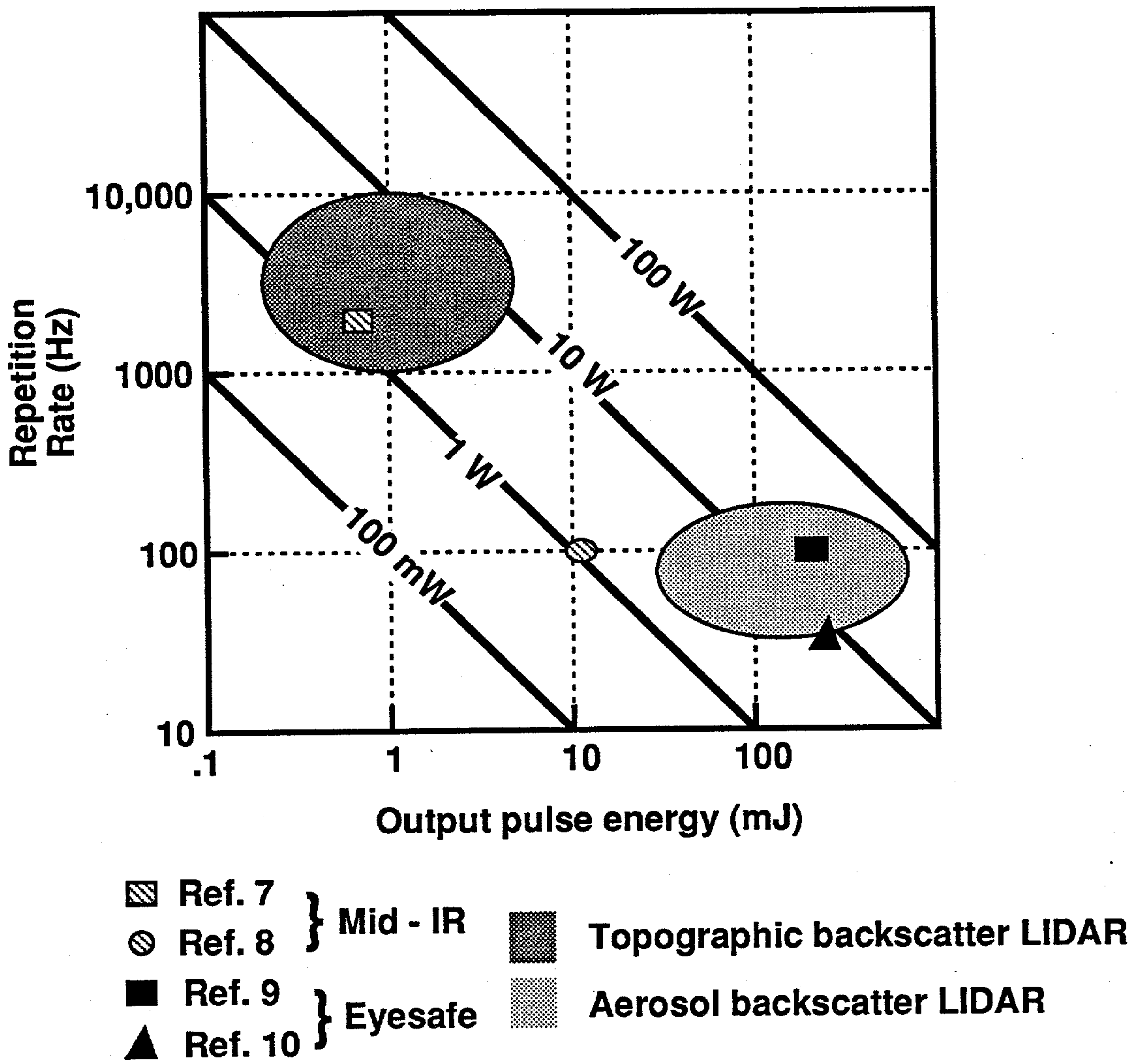


Figure 2. Applications space and average power demonstrations for near and mid-IR OPOs

The demand for average power from the infrared OPOs featured in figure 2 stems from certain long range LIDAR applications, and falls into two distinct regions of applications space. On the one hand, aerosol backscatter LIDAR requires pulse energies in the few hundred millijoule range and repetion rates up to 100 Hz, while topographic backscatter applications demand much higher repetition rates at millijoule energies.

2. EMERGING NLO APPLICATIONS AT THE 100 WATT LEVEL

Table 1 presents a summary of emerging scientific[11,12], military[13,14], and commercial/industrial[15-17] laser applications that generally require visible or infrared laser powers in the 100 Watt regime (or about an order of magnitude beyond the commercial state-of-the-art). A significant number of the applications listed call for ≈ 100 watts of green radiation at multi-kHz pulse repetiton rates, with pulse durations of ≈ 100 nsec (Na-guidestar dye laser pump source, laser isotope separation dye laser pump source, fine hole drilling, surfzone mine sensing.) Multiple applications of such a single laser technology broadens the interest in, and investment base necessary for the development of practical laser sources of this type. At the same time, it is important to recognize the significant differences in the demand pulse widths for these applications (i.e. < 5 nsec for mines vs. > 200 nsec for Na-guidestar pump source), which will likely drive the laser device architecture and the specific operating point for associated NLO convertors.

For practical use of such lasers in applications it will be necessary to extend the reliability and operating lifetime of the harmonic doublers significantly beyond present commercial experience (i.e. > 25 watts.) In the following sections we consider the prospects of higher average power harmonic generation from a fundamental point of view.

3. MATERIAL FIGURES OF MERIT FOR AVERAGE POWER FREQUENCY CONVERSION

The fundamental physical principles underlying average power effects on frequency convertors have been outlined in a number of papers[17,18,19]. The high average power potential of frequency conversion devices which depend on the $\chi^{(2)}$ nonlinear susceptability of solids is fundamentally limited only by "parasitic" optical absorbtion processes, since three wave mixing itself is intrinsically elastic (i.e. energy conserving in the electric field.) Generally one and two-photon absorbtion are the most important processes. Absorbtion of the fundamental or generated wavelengths leads to thermal gradients, whose primary effect on frequency conversion is the local phase mismatch induced by the local change in refractive indices with temperature. A secondary effect is the change in beam phasefront which leads to lensing and higher order aberrations. Gradients which are transverse to and along the beam direction are both generally present. The relative importance of these effects depends on the frequency conversion process and differs somewhat for harmonic generation and parametric oscillation.

By far, the most common way to cool frequency conversion crystals in practical devices is perpendicular (transverse) to the beam direction. If the heating caused by the laser beams was uniform, and the crystal faces were well insulated, only transverse gradients would be significant. The center to edge temperature difference is given by the well known formula:

$$\Delta T_{ce} = a_r g(\alpha/8\kappa)P_{av} \qquad (1)$$

where a_r is the beam aspect ratio (thickness/width); g is a geometry dependent constant ≈ 1, α and κ are the linear absorbtion coefficient and thermal conductivity respectively.

Significant longitudinal gradients can also arise, either because the absorption differs among the fundamental and generated waves, or when face cooled geometries (where the primary direction of heat flow is in the beam direction) are used. The latter case has been treated extensively in reference 18, but the former source is the more commonly encountered effect for small aperture, transversely cooled crystals. A good example of this is 1 µm pumped parametric oscillators using KTP which has a relatively strong absorbtion in the 3.5 µm region. The magnitude of these gradients can only be accurately estimated by a self-consistent numerical procedure. However it is straightforward to show that equation (1) with the absorbtion coefficient of the relevent harmonic wave and the incident power at the fundamental is a very conservative upper bound to the end-to-end thermal gradient expected in such a situation. Expression (1) is also an upper bound to the *transverse* gradient when α refers to the largest absorbtion coefficient for any of the frequencies involved, and P_{av} is the average power of the fundamental.

The phase mismatch associated with the temperature gradient ΔT_{ce} is given by

$$\Delta k = (1/2)\beta_T \Delta T_{ce} \tag{2}$$

where $\beta_T = d\Delta k/dT$ is the thermal sensitivity parameter for the three wave mixing process of interest. The efficiency of the conversion process is severely reduced when the ratio of the dephasing Δk to the nonlinear drive parameter $CLI_D{}^{1/2}$ exceeds a critical value[18]. These considerations lead directly to an upper bound on P_{av} in transversely cooled harmonic generators given by:

$$(g/a_r)P_{av} \leq 16\kappa C I_D{}^{1/2}/\beta_T\alpha = P_{tl} \tag{3}$$

If the drive irradience I_D is taken to be a material parameter (i.e. a characteristic "safe" operating value for the crystal of interest) the right hand side of equation (3), denoted P_{tl}, is a useful material figure of merit for average power harmonic generation. Generally, I_D is determined by the damage threshold of the crystal, but may also be a function of two photon absorbtion coefficient (see below). The $I_D{}^{1/2}$ dependence of the limiting average power reflects the fact that if a crystal with a given nonlinear coupling parameter C can be driven at a higher irradience, its length L (over which the dephasing accumulates) can be made shorter. The geometrical factors multiplying the average power on the left hand side of equation (3) represent the increase in average power made possible by beam shaping ($a_r < 1$).

The "thermally limited power" figure of merit, P_{tl}, is the average power analogue of the "threshold power" figure of merit proposed by Eimerl[18]:

$$P_{th} = (\lambda\beta_\theta/C)^2 \tag{4}$$

where β_θ is the angular sensitivity parameter for the phasematching process of interest. P_{th} represents the peak power necessary for efficient conversion in a critically phasematched harmonic generator.

For simple harmonic generation, the thermal dephasing effect on conversion efficiency is generally the most important average power limiter. Thermal lensing can change beam quality, but it is unusual to expect severe beam quality effects to be present before strong efficiency reductions also manifest themselves (an exception may be thermally insensitive doublers discussed below.) For OPOs the effect of thermal gradients is more complex. Numerical modelling suggests that absorbtion of the non-resonated wave can actually enhance conversion at low average powers by reducing back conversion. At high average powers, phasematching condition can be satisfied even in the presence of strong transverse thermal gradients by frequency pulling effects. For these reasons, OPOs under thermal load may still convert pump light efficiently, but with severe degradation of linewidth and beam quality. As yet, no generalized figures of merit analogous to equation (3) for OPO average power, brightness or spectral brightness have been proposed.

2. AVERAGE POWER POTENTIAL OF AVAILABLE MATERIALS

Table 2 lists parameters for frequency doubling of 1.06 μm for most of the crystals commonly used for this purpose. From these parameters are calculated the "limiting average power" values P_{tl} from equation (3). This comparison reveals dramatic variations among the intrinsic average power handling capabilities of these materials. The precision of the values of P_{tl} listed in table 2 depend, of course, on the values chosen for the two "extrinsic" material parameters α and I_D, as estimated from damage threshold data. Both optical absorbtion and damage threshold are often a function of crystal growth and fabrication, and can vary from crystal to crystal in the same material.

Table 2. Material parameters and figures of merit for average power frequency doubling of 1.06 μm.

	C $GW^{-1/2}$	κ mW/cm°C	β_T $cm^{-1}/°C$	β_θ $cm^{-1}/mrad$	I_D MW/cm^2	α cm^{-1}	P_{th} MW	P_{tl} W
DLAP II	1.76	6	0.56	3.7	250	0.013	50	12
KD*P I	0.81	12	0.24	4.9	250	0.002	400	175
KD*P II	0.98	12	0.30	2.5	250	0.002	75	169
$LiIO_3$ I	5.0	15	0.24	16.3	25	0.0005	120	1600
BBO I	5.2	13	0.11	10.6	250	0.001	47	4900
BBO II	4.1	13	0.15	7.0	250	0.001	33	2800
LBO I	2.08	33	0.88	1.38	250	0.0004	5.0	1700
LBO II	1.72	33	0.68	0.55	250	0.0004	1.2	1800
KTP II	7.2	30	0.20	0.60	100	0.01	0.08	600

Nonetheless, materials can vary widely in their potential on the basis of their C, κ and β_T values. It can be noted, for example that the large nonlinear coupling constant of KTP offsets its relatively large absorbtion coefficient (taken as the absorbtion at the second harmonic.) Similarly, a small thermal conductivity and large 1 μm absorbance give the organic crystal DLAP very poor average power properties. It is also interesting to note that of the materials in table 2, only LBO and KTP combine the features of low threshold power P_{th} and high P_{tl}. This makes them potentially far more useful than the other crystals for the multikiloherz, low pulse energy applications which dominate the applications space outlined in table 1.

For frequency conversion into the ultraviolet, two photon absorbtion becomes a more important source of thermal loading, and can impose a new limit on the drive irradience I_D. When two photon absorbtion is taken into account we have:

$$\alpha = \alpha_1 + \beta I_D \tag{5}$$

where α_1 is the normal 1 photon (linear) absorption coefficient and β is the two-photon absorbtion coefficient. Equation (3) becomes

$$(g/a_r)P_{av} \leq (16\kappa C/\beta_T\alpha_1)I_D{}^{1/2}/(1 + I_D/I_2) \tag{6}$$

where $I_2 = \alpha_1/\beta$. The right hand side of equation (6) has a maximum for $I_D = I_2$, and a new definition of P_{tl} obtains:

$$P_{tl} = (8\kappa C/\beta_T\alpha_1)(\alpha_1/\beta)^{1/2} \tag{7}$$

Along with this fundamental limitation, it is also true that linear absorbtion due to impurities is generally higher in the U.V. and damage thresholds are usually lower than in the visible and near infrared. Table 3 gives estimates of P_{tl} for KD*P and BBO for the generation of 266 nm by doubling of 532 nm light, assuming that two photon absorbtion, rather than damage threshold is the irradience limiting phenomenon. It is clear that U.V. average powers are constrained to much lower values than those which pertain to frequency doubling to visible wavelengths.

Table 3. Thermally limited powers for 4th harmonic generation of 1.06 μm, assuming 2-photon limited drive intensities.

	β_T $cm^{-1}/^oC$	α cm^{-1}	β cm/GW	I_2 MW/cm2	P_{tl} W
KD*P I	3.1	0.01	0.06	160	5
BBO I	1.4	0.01	1	10	10

4. TOWARD HIGHER AVERAGE POWERS.

There are several routes to extending the average power capabilities of harmonic generators:

Material improvement. The slow, but continuous improvement of absorbtion and damage properties of existing nonlinear crystals, as well as the occasional discovery of new useful materials, can clearly be expected to continue. The P_{tl} figure of merit shows that increasing the damage threshold has less leverage for improving average power than decreasing the absorbtion. In addition, a larger nonlinear coefficient can translate into a larger average power handling capability. However, as the above discussion implies, while there may be considerable headroom in the area of frequency doubling into the visible, there are more fundamental limitations in the ultraviolet which are less likely to be overcome by material development.

Thermally insensitive phasematching. Certain biaxial crystals such as DLAP[20] and LBO[21] have orientations where the thermal sensitivity β_T for certain frequency mixing processes vanishes to first order. In this case, the average power handling capacity of the crystal will be determined by lensing or second order thermal dephasing effects, or possibly thermal fracture. While this phenomenon has been known for some time, to date it has not been used in practice. In any case, it is likely to remain only a "point solution" to particular average power problems where such a crystal and orientation happen to exist.

High Aspect ratio geometries. This solution is well known[17] and has generally been implemented in slab laser systems where elongated apertures are naturally encountered. A related variant uses a moving crystal to spread out heat generation with low aspect ratio beams. In systems with round beams, or if the aspect ratio is required to be greater than 10:1, the implementation of the necessary beam shaping optics can be awkward.

Face cooled geometries. While this architecture has the best scaling properties[18], the engineering of multiplate devices represents some daunting challenges in coatings and precision fabrication. Eventually, as the demand for average powers begins to exceed several hundred Watts in the visible, or perhaps several tens of Watts in the U.V., such devices will be necessary.

6. CONCLUDING REMARKS

There are a growing number of applications for high average power solid state lasers which will stress the state-of-the-art in frequency conversion technology. These applications tend to cluster in the high repetition rate, low pulse energy regime which requires materials with the ability to convert beams with low peak to average power ratios with high efficeincy. The number of such materials is small, but there are excellent prospects for increasing average powers in the visible into the few hundred watt range as appropriate pump laser sources become available.

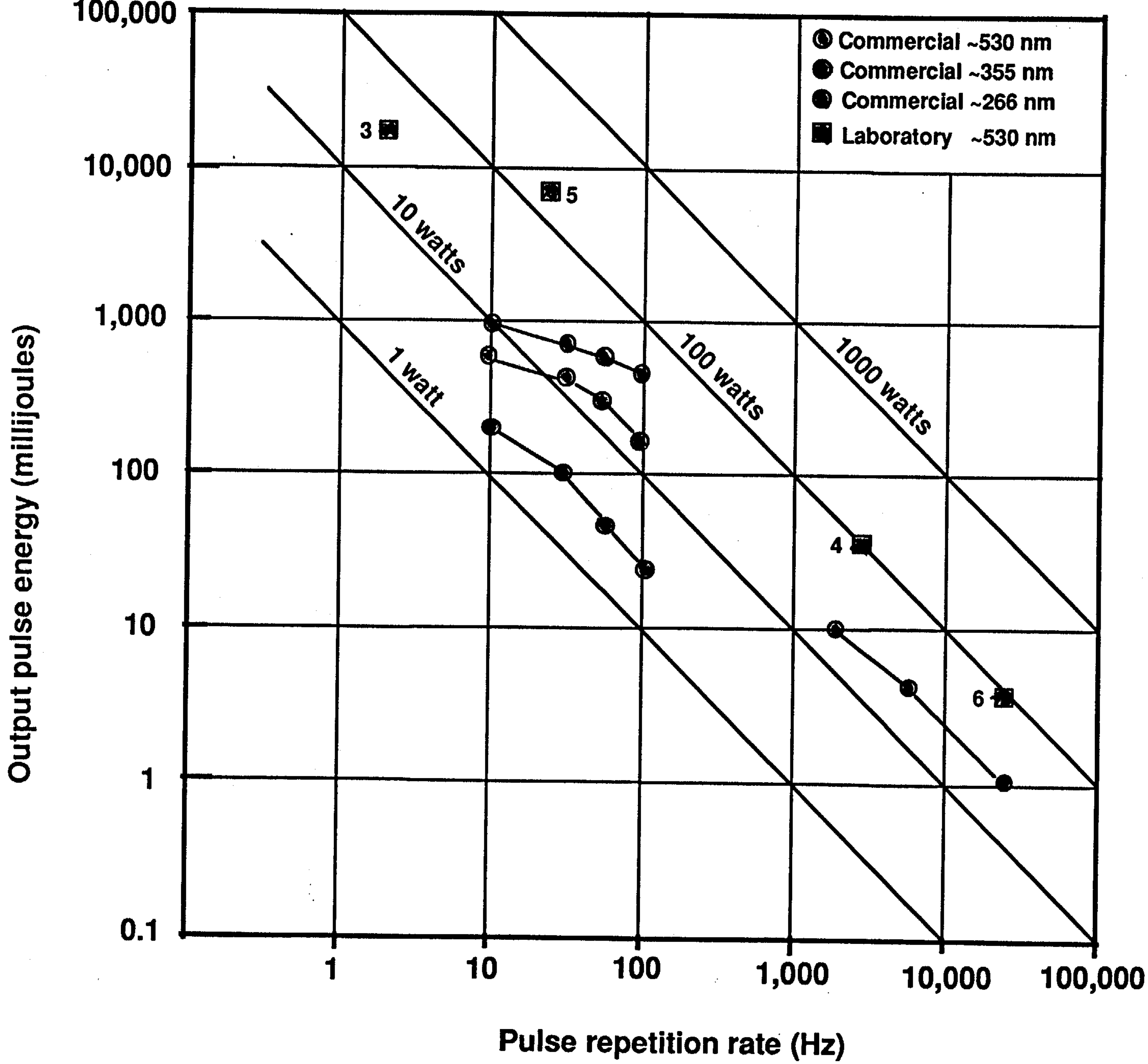
Some High-Average-Power NLO-based Laser Applications

Application	Wavelength (nm)	Av. Power (watts)	Pulse-Rep Rate (Hz)	Pulse Energy (millijoules)	Pulse Width (nsec)	BQ (xDL)
Scientific						
Na-Guidestar Laser Pump	~530	100	~5000	20	>200	<few
Global Windsensor (LAWS)	~2000	100	~10	10,000	>600	~ 1
Military						
Surfmine Sensor	~530	100	~5000	20	<5	~few
Biochemical Sensor	1550	40	80	500	100	~few
Missle-countermeasure	2000 5000	10 100	~25,000 100	0.4 1000	100 <1000	~few ~few
Industrial-Commercial						
Fine Hole Drilling	~530	40-100	~5000	8-20	<60	<fe
Isotope Separation (Pump)	~530	100-200	>5000	20-40	<100	~few
FPD Lithography	~355 (I) ~436 (G)	200 100	~4000 ~4000	50 50	<100 <100	~1 ~1
IC Lithography	248 193	10 10	~1000 ~1000	~10 ~10	>100 >100	~1 ~1

WFK 1/96

Table 1

HAP extrema performance levels of some nonlinear optical devices



WFK 1/96

Figure 1

1. REFERENCES

1. S.G. Anderson, "Diode-Pumped Nd:YAG Laser Provides High Energy At High Repetition Rates", Laser Focus World, February, 1994. p15.

2. Clark Instrumentation, Inc., ORC-1000 Product Sheet, 1993 Rochester, NY.

3. C.B. Dane, et. al., "Long Coherence-Length 600 nsec Solid State Laser Illuminator at 527 nm", Technical Digest, CLEO, 1995 paper CTuG4, p80.

4. S.P. Velsko, et. al., "100 Watt Average Power At 530 nm By External Frequency Conversion of an Electro-Optically Q-Switched Diode-Pumped Power Oscillator", Appl. Phys. Letters, 64, 3086 (1994).

5. R.J. St. Pierre, et. al., "High Average Power Frequency Doubling", LEOS Annual Meeting Technical Digest, paper SLL3.1, p694, San Jose (1993).

6. M.V. Ortiz, et.al., "High Average Power Second Harmonic Generation with KTP", OSA Proceedings on Advanced Solid State Lasers, Vol 13, p361 (1992).

7. G.T. Bennett, P.M. Noblett, and R.M. Morrison, "1.5 Watt Average Power Optical Parametric Oscillator at 3.8 μm", OSA Proceedings on Advanced Solid State Lasers, T.Y. Fan and B.H.T. Chai, eds, (Optical Society of America, Washington, D.C. 1994), Vol. 20, p. 419.

8. S.P. Velsko, M.S. Webb, W.M. Cook, J.M. Halpin, and W.A. Neuman, "Simultaneous Dual-line Seeded "Non-resonant" Parametric Oscillator for Mid-Infrared DIAL", in Conference on Lasers and Electro-0ptics, Vol. 15, 1995 OSA Technical Digest Series (Optical Society of America, Washington, D. C. 1995), CTh7.

9. A. Englander and R. Lavi, "A Multi-Watt Mid-IR OPO Using a Top-Hat Spatial Profile Pump Laser", OSA Proceedings on Advanced Solid State Lasers, T.Y. Fan and B.H.T. Chai, eds, (Optical Society of America, Washington, D.C. 1994), Vol. 20, PD2-1.

10. G. Rines, D. Rines, and P. Moulton, "Efficient, High Energy Optical Parametric Oscillators Pumped with 1 Micron Nd-lasers", OSA Proceedings on Advanced Solid State Lasers, T.Y. Fan and B.H.T. Chai, eds, (Optical Society of America, Washington, D.C. 1994), Vol. 20, p. 461.

11. J.R. Morris, "Efficient Excitation of a Mesopheric Solium Laser Guide Star by Intermediate-Duration Pulses", (Optical Society of America, A11, 832 (1994).

12. M.M. Sokoloski and R.J. Curran, "The Earth Observing System: A Mission for Lidar Remote Sensing Technology", SPIE, 868, 120 (1987).

13. W.B. Scott, "Lidar System to Detect Biological Warfare Agents", Aviation Week, p44, November 13, 1995.

14. Missile countermeasures reference

15. J.J. Chang, et. al., "Precision Micro Drilling with Copper Vapor Lasers", ICALEO, 13th International Congress, Orlando, Florida, October 1994, p323-332.

16. J.L. Emmett, et. al., "Laser R&D at the Lawrence Livermore National Laboratory for Fusion and Isotope Separation Applications", IEEE J.Q.E., 20, 591 (1984).

17. D. Hon, "High Average Power, Efficeint Second Harmonic Generation", Laser Handbook, Vol. 3, p.421, (Elsevier, North Holland, N.Y., 1979).

18. D. Eimerl, "High Average Power Harmonic Generation", I.E.E.E. J. Quantum Electron. QE-23, p. 575, (1987).

19. N.P. Barnes and J.A. Williams-Byrd, "Average Power Effects In Parametric Oscillators and Amplifiers", J. Opt. Soc. Am. B, Vol. 12, 124, (1995).

20. C.E. Barker, D. Eimerl, and S.P. Velsko, "Temperature Insensitive Phase Matching for Second Harmonic Generation in Deuterated l-arginine Phosphate", J. Opt. Soc. B, Vol. 8, p. 2481, (1991).

21. M. Dreger, private communication.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.