

Diode Pumped Solid-State Laser Oscillators for Spectroscopic Applications

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1. Introduction

Solid state laser development has been paced by the improvement of pumping sources. From the helical lamps used to pump the early Ruby lasers, to the linear arc and pulsed flashlamps used to pump Nd:YAG lasers, solid state laser pump sources have improved steadily in power and efficiency. The latest development is pumping solid state lasers with diode lasers and diode laser arrays. The development of high power, efficient, long lived diode lasers promises a revolution in solid state laser technology.

The advantages of diode laser pumping over flashlamp pumping include greater spectral brightness in the pump bands of the ions, greater efficiency compared to flashlamps, longer operating lifetime, and operation without high voltage. At this time the disadvantages are the low power of the diode lasers and the cost per watt of output power. However, the cost of diode lasers is decreasing so rapidly that diode laser pumping should be economically favored over flashlamp pumping for 1 Watt average power Nd:YAG lasers by 1989, and for 10 to 100 Watt Nd:YAG lasers by 1991.

The diode laser is inherently a cw device. The long upper state lifetime of ion levels in solid state laser materials permits energy storage which leads to potential advantages for diode laser pumping of ions. These advantages include Q-switched and modelocked operation; the possibility of extracting high peak and average power from the solid state laser medium in a diffraction limited spatial mode by summing the output of many diode laser pump sources; the ability to operate on many ion wavelengths; and the potential to convert to additional wavelengths by nonlinear optical processes. In addition, the low intrinsic loss of the crystalline medium relative to the semiconductor diode laser medium leads to a potential reduction in the Schawlow Townes linewidth of six orders of magnitude. Diode laser pumping of solid state lasers increases the power spectral brightness of the diode laser pump source without a significant loss in overall efficiency.

Advances in diode laser pumping of solid state lasers has been reviewed recently [1] as has the progress in diode lasers and diode laser arrays[2]. It is worth noting that the first suggestion for diode pumping was by Newman in 1963 who demonstrated fluorescence from the neodymium ion doped in CaWO_4 [3]. Considerable progress in the 1970's and in the early 1980's coupled with the recent rapid progress in the power and efficiency of diode lasers has led to renewed interest in this approach to solid state laser engineering.

2. Diode Laser Pumped Four Level Lasers

In 1985, Zhou et. al.[4] demonstrated efficient, frequency stable, laser diode pumped Nd:YAG laser operation. The end pumped configuration used a gradient index lens to focus the diode laser radiation into the 5 mm long monolithic Nd:YAG rod. The 2 mW threshold and 25% slope efficiency demonstrated the effectiveness of the end pumping approach.

(NASA-CR-180576) DIODE PUMPED SOLID-STATE
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APPLICATIONS (Stanford Univ.) 7 p Avail:
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Further, the 10 kHz linewidth showed promise of achieving the expected Shawlow Townes linewidth for this laser of less than 1 Hz-mW. However, spatial hole burning limited single axial mode operation to sub-milliwatt power levels in the standing wave device.

This early work was extended by Sipes who demonstrated 6% wall plug efficiency for diode laser pumping of Nd:YAG[5]. In addition, diode laser pumping of Nd:YLF with internal SHG to generate 532nm was demonstrated by Fan et. al.[6] and by Baer[7]. The lasing ion and the nonlinear material for frequency doubling were combined in diode laser pumped Nd:MgO:LiNbO₃. This self-doubling and self-Q-switching laser oscillator operated at 3.6mW threshold with a 39% slope efficiency[8]. Recently, diode laser pumping was extended to Nd:Glass with a surprisingly low threshold of 2.5mW at a slope efficiency of 42%[9]. The low loss of the glass host led to the low threshold in spite of a factor of ten less gain cross section for Nd:Glass compared to Nd:YAG. Nd:Glass operation offered the advantages of broad absorption bands for diode pumping, wide spectral bandwidth for modelocked operation, and high doping levels for side pumping with diode laser arrays.

3. Diode Laser Pumped Three Level Laser Systems

Many of the potential useful ions for near infrared laser operation are three level systems with residual population in the lower level. There was skepticism about whether these laser systems could be pumped by low power diode lasers. Fan and Byer[10] showed theoretically that under proper conditions diode laser pumping was possible. These conditions were realized with the successful diode laser pumping of the 946 nm transition in Nd:YAG[11] which was subsequently frequency doubled to generate blue radiation at 473 nm[12].

The eyesafe requirement of many laser radar applications including global remote wind sensing, led to the search for lasers that operate at wavelengths longer than 1400 nm. Following early work by Duczynski et. al. [13], Fan et. al.[14] successfully demonstrated cw, diode laser pumped, room temperature operation, of the three level laser Tm:Ho:YAG at 2090nm. Diode laser radiation was absorbed at 781nm by the Tm ion with subsequent transfer to the holmium ion which oscillated at 2090nm. This transfer laser should allow one absorbed pump photon to generate up to two inverted holmium ions for a potential pump quantum efficiency of two. A threshold of 4.4mW with slope efficiency of 19% were observed in early diode laser pumping experiments. This laser system is more complex than Nd:YAG and requires additional research to optimize the doping concentrations. However, the 8 msec upper level energy storage time of the holmium ion makes possible extraction of high peak power pulses by Q-switching.

4. Q-Switched and Modelocked Operation

The potential to store energy and extract high peak power pulses is a significant advantage for diode laser pumping of crystal and glass lasers. Both Nd:YLF and Nd:YAG have been Q-switched. The cw-pumped laser oscillators were acousto-optically Q-switched at kilohertz repetition rates with peak power levels of greater than 2.5 kW for Nd:YLF and 1.8 kW for Nd:YAG[15]. Q-switched pulse widths of 10 nsec were observed for these short cavity, compact, laser diode pumped oscillators. The higher peak power and larger pulse energy of Nd:YLF compared to Nd:YAG reflected the longer storage time of the Nd:YLF system. The kilowatt peak powers of these lasers allows direct external frequency doubling.

Diode laser pumping of Nd:Glass with active modelocking to achieve pulses of 45psec width was reported by Basu and Byer[16]. The laser oscillator utilized a 3 mm thick Brewster plate of Nd:Glass in an astigmatically compensated three mirror folded resonator similar to the early cw dye laser cavity configuration. The 8 mW diode laser pump threshold of this actively modelocked Nd:Glass laser allowed pumping by a single stripe 30 mW diode laser. Modelocked operation was extended to Nd:YAG which operated at a 90 psec pulsewidth.

5. Single Axial Mode Non-Planar Ring Oscillator

Spatial hole burning in the standing wave monolithic diode laser pumped oscillators led to multi-axial mode operation at powers above the milliwatt level. The desire to maintain the inherent stability of the monolithic structure and achieve single axial mode operation at high power levels led to the consideration of a ring oscillator. To eliminate spatial hole burning and to assure operation in a single direction, the ring oscillator should contain the three elements of an optical diode in its monolithic structure. These elements are a Faraday rotation, a wave plate, and a polarizer. Kane and Byer[17] proposed and demonstrated the monolithic, unidirectional, single axial mode, non-planar ring oscillator to meet these requirements. The key element in this invention is the breaking of planar symmetry to achieve the combination of Faraday rotation, waveplate and polarizer within the monolithic structure. The nonplanar ring oscillator has operated at up to 163 mW of single axial mode output when argon ion laser pumped, and up to 25 mW of single axial mode power when diode laser pumped. Using single mode fibers to combine the radiation of an NPRO with a single axial mode standing wave oscillator, Kane et. al.[18] have demonstrated that the device operates with free running linewidths of less than 10 kHz.

The advantages of the nonplanar ring oscillator include single frequency operation with frequency stability due to the monolithic construction and immunity to feedback. This last factor is critical in many applications of the NPRO and is an inherent property of the nonplanar ring design[19].

The Nd:YAG nonplanar ring oscillator offers a narrower linewidth and improved frequency stability compared to the diode laser. For example, the Nd:YAG NPRO with scatter loss of 0.001 cm^{-1} has a projected Schawlow Townes linewidth of 0.3 Hz-mW for a one centimeter diameter ring oscillator. Work is in progress to reduce the technical noise dominated 10 kHz linewidth of the NPRO to the subHertz Schawlow Townes linewidth by using power stable diode pump sources and by using rapid feedback frequency control through an applied magnetic field to offset the slow temperature tuning of -3.1 GHz per degree.

The stable, single axial mode, feedback resistant output of the Nd:YAG nonplanar ring oscillator has allowed efficient SHG by external resonance doubling in LiNbO_3 [20]. In the early experiments, 15mW of cw 1064nm input was doubled to generated 3mW of single axial mode green output at 15% SHG efficiency. With improvements in the optical coatings, SHG efficiencies approaching 50% are predicted.

6. Amplification

In many applications high peak power radiation is desired but at linewidths that remain single frequency within the Fourier transform limit of the sampling time. We have investigated linear amplification of milliwatt power level sources using a multipass, flashlamp pumped, slab geometry Nd:YAG configuration[21]. The measured 62 dB gain for a 30 microsecond duration, was limited by the onset of superfluorescence. The linear amplification approach to generate high peak power levels offers an alternative to Q-switching. The advantages of linear amplification are the ability to select the pulse width and thus the Fourier Transform linewidth of the output radiation independent of Q-switch dynamics.. The nonplanar ring oscillator, Nd:YAG slab amplifier, and single mode optical fiber for heterodyne mixing was used for a successful demonstration of coherent laser radar at 1064nm[22].

7. Summary

The rapid improvement in diode laser pump sources has led to the recent progress in diode laser pumped solid state lasers. To date electrical efficiencies of greater than 10% have been demonstrated[23]. As diode laser costs decrease with increased production volume, diode laser and diode laser array pumped solid state lasers will replace the traditional flashlamp

pumped Nd:YAG laser sources. The use of laser diode array pumping of slab geometry lasers will allow efficient, high peak and average power solid state laser sources to be developed.

Perhaps the greatest impact of diode laser pumped solid state lasers will be in spectroscopic applications of the miniature, monolithic devices. The nonplanar ring oscillator offers the potential of sub-Hertz linewidths in a size that matches the recently developed ion traps. It may be possible, one day, to carry in your watch the laser source and single ion that are the basis for a highly accurate miniature clock.

This work was supported by U. S. Army Research Office contract DAAG29-84-K-0071 and NASA grant NAG 1-182. W. J. Kozlovsky and C. D. Nabors gratefully acknowledge the support of the Fannie & John Hertz Foundation.

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Continuous-wave operation of a room-temperature, diode-laser-pumped, 946-nm Nd:YAG laser

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Received June 4, 1987; accepted July 21, 1987

Single-stripe diode-laser-pumped operation of a continuous-wave 946-nm Nd:YAG laser with less than 10-mW threshold has been demonstrated. A slope efficiency of 16% near threshold was shown with a projected slope efficiency well above a threshold of 34% based on results under Rhodamine 6G dye-laser pumping. Nonlinear crystals for second-harmonic generation of this source were evaluated. KNbO_3 and periodically poled LiNbO_3 appear to be the most promising.

There has been a revival of interest in using diode lasers as pump sources for solid-state lasers. While much of the recent work has focused on the ${}^4F_{3/2}$ - ${}^4I_{11/2}$ 1.06- μm Nd^{3+} transition, there are other transitions of interest in Nd^{3+} , such as the ${}^4F_{3/2}$ - ${}^4I_{9/2}$ Nd^{3+} transition in the 900–950-nm range. The difficulties with this transition are a typically lower stimulated emission cross section than the ${}^4F_{3/2}$ - ${}^4I_{11/2}$ transition and a populated lower laser level at room temperature since the lower laser level is less than 1000 cm^{-1} above the ground state. A way to overcome these difficulties is to confine the pump beam and the laser mode in a Nd-doped single-mode glass fiber to achieve high pump densities and gain.^{1,2} Alternatively, we recently demonstrated through modeling and experiment that diode-laser pumping of the 946-nm transition in Nd:YAG in a bulk device at room temperature is possible.^{3–5} We also proposed second-harmonic generation (SHG) of this source to generate coherent blue radiation in an all-solid-state device^{4,5} similar to solid-state green sources by frequency doubling of the ${}^4F_{3/2}$ - ${}^4I_{11/2}$ Nd^{3+} transition.^{6–9} Blue generation was subsequently demonstrated by intracavity SHG of a 946-nm Nd:YAG laser pumped by a Rhodamine 6G (R6G) dye laser.¹⁰ In this Letter we report on experimental results of the first diode-laser-pumped 946-nm Nd:YAG laser at 300 K and discuss some aspects of SHG of this source.

The primary difficulties with this transition in Nd:YAG are an approximately order-of-magnitude-lower stimulated emission cross section than the 1.064- μm line¹¹ and a lower laser level at 857 cm^{-1} above the ground state. The first laser operation on this transition was demonstrated at 77 K to reduce the lower laser level population.¹² In 1969, Wallace and Harris demonstrated laser action on this transition with flash-lamp pumping at room temperature.¹³ The question is whether diode-pumped laser operation at room temperature is possible. The low stimulated emission cross section of $4 \times 10^{-20} \text{ cm}^2$ itself is not a problem; low threshold and high slope efficiency have been demonstrated in Nd:glass at 1.053 μm , which has a similar cross section.¹⁴ However, one difficulty with the low cross section is parasitic oscillation on the 1.064- μm transition, which previously pre-

vented operation at 946 nm in a room-temperature, end-pumped Nd:YAG laser.¹⁵ We have shown that when proper design is used, parasitic oscillation in cw operation is not a problem.⁴

The lower laser level population, 0.0074 of the dopant concentration at 300 K, leads to an increase in threshold. The increase in threshold can be viewed in either of two equivalent ways. Either the laser must have enough gain to overcome reabsorption at the laser wavelength in addition to the other cavity losses or additional pump power is needed just to reach population inversion. Modeling of quasi-three-level laser transitions showed that thresholds of less than 10 mW at 300 K are possible under diode pumping.⁴

Figure 1 shows our experimental apparatus. The output of a single-stripe diode laser operating at 808.5 nm is combined with the output of a R6G dye laser at 588 nm with part of the diode-laser beam sent to a monochromator to allow its wavelength to be monitored. The beams pass through a polarizer and a Fresnel rhomb to provide isolation against feedback for the diode laser. The beams are focused to spots with slightly less than $\sim 20\text{-}\mu\text{m}$ radii in the Nd:YAG rod by a 4-cm focal-length lens. The YAG rod is doped with nominally 1% Nd and is $\sim 0.13 \text{ cm}$ long with 1-cm radius-of-curvature faces polished on the ends. One face is coated with a higher reflector (HR) at 946 nm and the other with an antireflection coating at 946 nm. For this rod length, 87% of the incident power at 588 nm and 67% at 808.5 nm is absorbed. The output coupler located approximately 5 cm from the rod has a 5-cm radius of curvature and is either a HR or a 99% reflector at 946 nm with greater than 60%

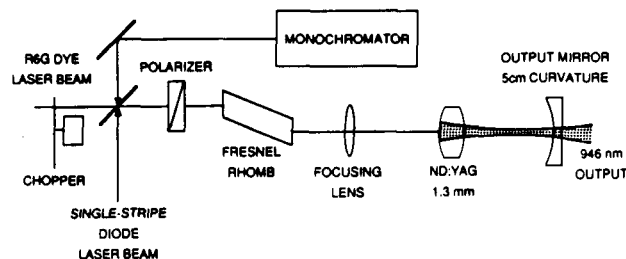


Fig. 1. Schematic diagram of the experimental apparatus.

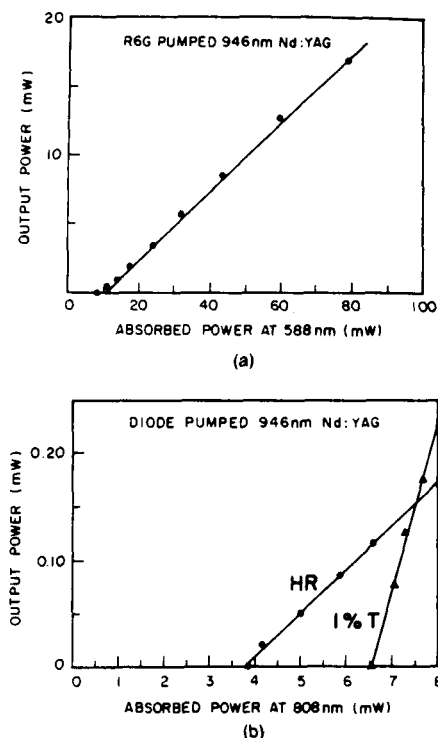


Fig. 2. (a) Output at 946 nm under R6G dye-laser pumping at 588 nm with 99% reflectivity output coupler. Line is a least-squares fit to the highest five data points. (b) Output at 946 nm under single-stripe diode-laser pumping at 808.5 nm with both HR and 99% reflective output couplers. The lines are least-squares fits to the data.

transmission at $1.064 \mu\text{m}$. Gaussian beam radii for the TEM_{00} mode of slightly less than $20 \mu\text{m}$ in the Nd:YAG are achieved in this cavity.

Figure 2(a) shows the output at 946 nm for 1% output coupling under R6G dye-laser pumping at 588 nm. The slope efficiency well above threshold of 24% is substantially higher than that demonstrated previously⁴ because of increased output coupling and lower losses other than output coupling. The absorbed pump power threshold of 8.1 mW is in agreement with the calculated value of 7.9 mW assuming $20\text{-}\mu\text{m}$ -radii pump and cavity modes and 1% round-trip losses. Figure 2(b) shows the results for diode-laser pumping for both HR and 1% output couplers. The thresholds of 3.9 and 6.6 mW are again in good agreement with the calculated values of 3.0 and 5.7 mW for the HR and 1% output couplers. The slope efficiencies near threshold are 4% for the HR and 16% for the 1% transmission mirrors.

While one would expect the slope efficiency for diode pumping to be 1.4 times larger than that under dye-laser pumping because of a smaller quantum defect, this is not the case because we are comparing slope efficiency barely above threshold under diode-laser pumping with that well above threshold under dye-laser pumping. A better comparison is the relative outputs near threshold since it has been shown that near threshold the slope efficiency is lower than at well above threshold,¹⁶ especially in lasers with a populated lower laser level⁴ where the reabsorption that is

due to this population appears as a real loss near threshold. The slope efficiency near threshold is dependent on the amount of reabsorption loss due to the lower laser level population⁴ and on the tightness of focus of the pump beam¹⁶ with higher slope efficiencies for tighter focus and less reabsorption loss.⁴ The output under diode-laser pumping rises at least 1.1 times faster than that under R6G dye-laser pumping near threshold. The threshold numbers indicate that the dye-laser pump is focused tighter than the diode-laser pump, so it appears that the only difference between R6G dye and diode-laser pumping is the quantum defect. If this is the case, the slope efficiency well above threshold should approach 34% under diode-laser pumping based on the 24% slope efficiency under R6G dye-laser pumping.

We have analyzed a number of nonlinear materials for SHG of the 946-nm light. Efficient SHG of continuous-wave 946-nm radiation is more difficult than at $1.06 \mu\text{m}$ because a number of crystals with relatively high nonlinearity at $1.06 \mu\text{m}$ cannot be used for 946-nm SHG. KTiOPO_4 (KTP) has high effective nonlinearity at $1.06 \mu\text{m}$ but low nonlinearity at 946 nm because Type I instead of Type II phase matching must be used.¹⁷ $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ does not have enough birefringence to phase match at 946 nm. Regular LiNbO_3 does not phase match for 946 nm, but Li-diffused stoichiometric LiNbO_3 phase matches at -37°C for doubling 954 nm and may phase match 946 nm at lower temperatures.¹⁸ Thus it is important to evaluate materials appropriate for 946-nm SHG.

It has been shown that the SHG conversion efficiency for Gaussian beams is a function of the Poynting vector walk-off angle between the fundamental and the second harmonic, ρ , and $|d_{\text{eff}}|^2/n^3$, where d_{eff} is the effective nonlinearity and n is the refractive index.¹⁹ For plane waves in the limit of undepleted fundamental power, the conversion efficiency is proportional to $|d_{\text{eff}}|^2 l^2/n^3$, where l is the length of the nonlinear crystal; but for finite diameter beams, both diffraction and double refraction limit the effective interaction length of the crystal. For nonzero Poynting vector walk-off little increase in conversion efficiency is possible for a crystal longer than the aperture length l_a , which is equal to $\sqrt{\pi w/\rho}$, where w is the Gaussian beam radius. Thus for nonzero Poynting vector walk-off the conversion efficiency goes as $|d_{\text{eff}}|^2 l_a^2/n^3$. For $\rho = 0$, the effective interaction length is limited by diffraction and is proportional to the confocal distance for the beam. The general case of SHG with Poynting vector walk-off and focusing into the SHG crystal taken into account has been treated by Boyd and Kleinman.¹⁹

Table 1 lists a summary of calculations for the Poynting vector walk-off angle ρ , l_a for $w = 30 \mu\text{m}$, relative conversion efficiency given by $|d_{\text{eff}}|^2 l_a^2/n^3$ normalized to that of noncritically phase-matched KNbO_3 for which $|d_{\text{eff}}|^2/n^3 = 28.7 \text{ pm}^2/\text{V}^2$, and phase-matching angles where θ_m is the angle of propagation relative to the z axis and ϕ_m is the angle to the x - z plane. The references used to calculate these quantities are listed. All the calculations are for Type I SHG. Noncritically phase-matched KNbO_3 has a high relative conversion efficiency, but the phase-

Table 1. Relative Conversion Efficiency, ρ , l_a , and Phase-Matching Angles for 946-nm SHG

Nonlinear Crystal	ρ (deg)	l_a (cm)	Rel. $ d_{\text{eff}} ^2 l_a^2 / n^3$	θ_m, ϕ_m	Ref.
KTiOPO ₄ (KTP)	2.7	0.11	1.5×10^{-4}	$\theta_m = 53^\circ, \phi_m = 45^\circ$	17, 20
LiIO ₃	4.6	0.066	1.7×10^{-3}	$\theta_m = 35^\circ$	21
β -BaB ₂ O ₄	3.4	0.089	1.5×10^{-3}	$\theta_m = 25^\circ, \phi_m = 30^\circ$	22
KNbO ₃ (angle)	0.85	0.36	0.44	$\theta_m = 90^\circ, \phi_m = 60^\circ$	23
KNbO ₃ (noncritical)	0	0.5 ^a	1	$\theta_m = 90^\circ, \phi_m = 90^\circ$	24
Periodically poled LiNbO ₃	0	0.5 ^a	1.6	$\theta_m = 90^\circ$	21, 27

^a These are not values of l_a . See text for explanation.

matching temperature is 180°C.²⁵ One intriguing possibility is periodically poled LiNbO₃.^{24,26,27} The calculated d_{eff} is that for the optimum case, periodically poled using d_{33} with flipped domains every coherence length. In practice, the lengths of these two crystals are limited by available crystal size and increasing loss for longer crystals. Thus the lengths of these two crystals were chosen to be 0.5 cm although their confocal distances for $w = 30 \mu\text{m}$ are greater than 1 cm. Angle-tuned KNbO₃, which reduces the phase-matching temperature, also has a relatively large conversion efficiency. KTP, LiIO₃, and β -BaB₂O₄ are poor choices for SHG of low-power sources because of their low d_{eff} and large ρ , which severely limits l_a .

Better operation on the ${}^4F_{3/2}$ - ${}^4I_{9/2}$ transition should be possible. With the recent availability of much higher-power diode lasers,²⁸ the 946-nm laser could be operated farther above threshold and with higher output coupling to get higher slope efficiency, and the rod length could be increased to absorb a larger fraction of the pump light. Eventually it may be possible to have higher slope efficiency for this device than for a 1.06- μm Nd³⁺ laser. Other host materials, such as other garnets and perovskites such as YAlO₃, may be of interest. Both classes of material have a relatively large splitting for the ground-state manifold, which reduces the lower laser level population. However, spectroscopic data such as absorption in the diode-laser pump bands, stimulated emission cross section, and energy levels of the ground-state manifold need to be known before judgments can be formed on the desirability of a given host.

In summary, continuous-wave diode-laser-pumped operation of a 946-nm Nd:YAG laser at room temperature has been demonstrated with low thresholds and 16% slope efficiency near threshold. Slope efficiency as great as 34% in this device is projected for operation well above threshold. A number of SHG crystals have been evaluated for SHG of this laser; KNbO₃ and periodically poled LiNbO₃ are the most promising.

This research was supported by NASA. T. Y. Fan would like to thank the IBM Corporation for supporting him with an IBM Graduate Fellowship.

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