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M. A. Summers
J. D. Williams
B. C. Johnson
D. Eimerl

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Harmonic Generation at High Peak Power

M. A. Summers, J. D. Williams, B. C. Johnson, D. Eimerl

Lawrence Livermore National Laboratory
P.O. Box 5508, L-487, Livermore, California 94550Introduction

The technology of frequency converting intense laser light was demonstrated during the early 1960s¹. In the initial experiments, the deep red wavelength of a ruby laser produced ultraviolet light, a shorter wavelength harmonic of the fundamental laser radiation, as it was passed through a quartz crystal. Shortly afterwards the Nd:YAG infrared laser was used to produce visible or ultraviolet light as it passed through a crystal of potassium dihydrogen phosphate (KDP).^{2,3} Today frequency conversion is a well characterized and widely used method of producing intense, coherent light at wavelengths unavailable from the source laser medium.

This frequency conversion capability substantially increased the research flexibility of well behaved laser media like Nd:YLF-Nd:glass. In fact, the potential for high peak power, short wavelength capability was one reason for selecting the Nd:glass laser system over the much longer wavelength CO₂ laser for Inertial Confinement Fusion (ICF) research at Livermore since the benefits of target irradiation with short wavelengths had been anticipated.⁴

Through the 1970s the frequency conversion technique was applied to high power Nd:glass laser systems used for ICF research in this country and abroad. The team at KMS Fusion, Inc. of Ann Arbor, Michigan was the

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first to conduct ICF plasma experiments using the second harmonic (2ω) of Nd:glass which is in the green part of the visible spectrum.⁵ They used a KDP crystal with a clear aperture of approximately 10 cm in diameter. At Ecole Polytechnique in France, ICF plasma characteristics were measured at the second (2ω) and fourth harmonic (4ω) of Nd:glass, again using KDP,⁶ while the University of Rochester, LLE, characterized the plasma conditions at the third harmonic (3ω) using KDP.^{7,8}

Here at Livermore, the Argus laser system was frequency converted to study plasma conditions at 2ω , 3ω , and 4ω .^{9,10} The real payoff of short wavelength was demonstrated on the Novette laser system where, at the multi-terawatt, multi-kilojoule level, higher fuel densities were achieved by irradiating targets using visible (2ω) light compared with the infrared fundamental (1ω) wavelength.¹¹

By 1980, the frequency conversion technology as applied to ICF laser systems was well established. The non-linear material of choice, KDP, had been used to demonstrate conversion efficiencies greater than 50% on high power laser systems at apertures as large as 15 cm in diameter. The challenge for the next five years would be to scale up this technology to the Nova class laser system where meaningful ICF target implosions could be achieved.

Crystal Array Design

Construction of the Nova scale (100TW, 100kJ, 1 μ m) frequency conversion subsystem required development of the technology associated with large aperture crystal arrays. Single crystals of KDP large enough

to convert the Nova 74 cm diameter beam were not available and did not appear practical in time to meet the scheduled project completion milestone.

Building an array of KDP crystals for frequency conversion is made difficult because of the precise angle tuned phase-matching tolerances required between the Nova 1 ω pump beam and the KDP crystallographic axis. In previous work using a single KDP crystal, the conversion efficiency would be maximized by manually tuning the crystal orientation; however, with an array of crystals, each must be aligned to the beam individually. An approach using individual angle adjustments, although feasible, limited the array architectural options and was of questionable practicality on such a large and complex system as the Nova laser. Without individual crystal alignment adjustments, each crystal must be precisely fabricated and mounted so that when the array is aligned to the Nova beam each crystal is within ± 100 μ rad of the optimum orientation. This fabrication tolerance was found to be beyond conventional polishing capabilities. However, single point diamond machining of KDP appeared to be capable of meeting the tolerance and was developed at Livermore to its present state of the art.

The machining and crystal orientation of KDP was remarkably successful. Not only could the surface normal to crystal axis orientation tolerance be achieved using the crystal orientation measurement system (COMS) and single point diamond machining, but the final optical finish was also generated. The Nova KDP crystals are oriented to better than ± 10 μ rad, have better than $\lambda/4$ transmitted wavefront distortion and

a residual surface roughness of less than 200 Å peak to valley.¹²

Figure 1 shows a Nova KDP crystal during the machining process.

The Nova array developed into an extremely simple device with the flexibility to efficiently produce 2ω or 3ω light. Historically, 3ω light was generated in a two step process. The first KDP crystal converted some of the 1ω to 2ω light with an optimum conversion efficiency of approximately $2/3$. A second KDP crystal, fabricated differently, is used to mix the 2ω with residual 1ω to produce 3ω light with intrinsic conversion efficiencies approaching 90%. Unfortunately, this optimum 3ω crystal array design does not efficiently produce 2ω light over the same range of pump intensities.

Methods of efficiently producing 3ω light had been thoroughly analyzed⁷ at the University of Rochester, LLE. One method was developed by them for their ultraviolet experiments. The team at Livermore recognized that another similar system could efficiently and conveniently produce 2ω light as well as 3ω light.¹³ The Nova crystal array was designed to use this modified two step process, in which both KDP crystals are cut at an angle halfway between the 2ω and 3ω phase-matching orientations and mounted in one monolithic assembly. Now the same "two crystal" architecture can efficiently produce 3ω or, by a 10° rotation and $1/4^\circ$ tilt adjustment, efficiently produce 2ω light using the Quadrature architecture. This is a remarkably attractive architecture given the large quantity of KDP that might have been required to field an optimum 2ω design plus a separate optimum 3ω design and the difficulty of changing arrays because of their inaccessibility once installed on the Nova target chamber.

The final step in the Nova crystal array design was the development of an antireflection coating for KDP. The uncoated KDP crystals mounted in a $2\omega/3\omega$ architecture would lose approximately 16% in performance due to the Fresnel reflections at each crystal surface. Historically, this loss was reduced by immersing the crystals in index-matching fluid. In fact, the first 2ω array built for the Novette laser at Livermore was a fluid filled array. This design suffered from particulate induced fluid deposition in the presence of a high intensity light. The required thin fluid gaps and fluid contamination conspired to make this approach undesirable.¹⁴

During the initial Nova design, as a backup design to the low loss but damage prone fluid filled cells, a bare crystal, deep-web "egg-crate" design was built and successfully tested on a 4ω converter for Novette target experiments. In the event that fluid filled arrays could not be made to work, the backup design would use uncoated KDP crystals and accept a 16% lower performance.

During this time, however, a sol-gel antireflection coating was developed at Livermore which could be applied to KDP with spectral characteristics of a classic quarter wave AR design and which had a high damage threshold.¹⁵ This porous SiO_2 , sol-gel coating could be easily applied by a spinning technique similar to the photoresist deposition technique used in the semiconductor technology.¹² Figure 2 shows the coating tank used to coat all of the Nova KDP crystals. This sol-gel coating, with the spectral characteristics optimized for the position in the array, was applied to each surface prior to installation in the deep-web egg crate. The final assembly is shown in Fig. 3.

Crystal Array Performance

Initial performance tests of the crystal arrays on the Nova laser system were disappointing. Above 1.5 GW/cm^2 beam power, the conversion efficiency from 1ω to 3ω departed radically from the theoretical estimates and measurements made on small-scale experiments conducted prior to the Novette/Nova trials. The 2ω performance also departed at the same pump intensity. In fact, closer examination of the early Novette 2ω , 3ω , and 4ω experiments revealed that in all cases the measured conversion efficiency departed from our theory when the pump intensities exceeded $\sim 1.5 \text{ GW/cm}^2$.

In small scale, well characterized experiments conducted on Nova production crystals, 2ω and 3ω results were obtained which were in close agreement with our theoretical calculations up to intensities as high as 4 GW/cm^2 . It was apparent that some beam parameter important to the frequency conversion process was strongly intensity dependent. The difficulty in identifying the source of low conversion efficiency was that virtually all important beam parameters are intensity dependent in high power solid state laser systems like Nova.

After investigating nonlinear propagation which affects the phase and intensity of the pump light, stimulated Raman scattering which shifts the wavelength, and polarization ellipse rotation which affects the $1\omega^o-1\omega^e$ mix ratio, we identified the problem as an intensity-dependent depolarization of the 1ω pump beam.^{16,17,18} It is due to a combination of stress-induced depolarization in optical

components and to non-linear propagation phenomena called polarization ellipse rotation causing the mix ratio to be intensity-dependent.

Figure 4 shows the depolarization pattern observed on a Nova 74cm diameter beam by rotating the frequency conversion array so that it converted to 2ω only the depolarized part of the beam. Clearly a large fraction of the aperture is depolarized. The depolarization is caused by stress in the split disk amplifiers and vacuum loaded spatial filter lenses. The amount of depolarization at the crystal array is less than 3% in energy averaged over space and time and is intensity dependent. Polarization ellipse rotation rates as high as $5^\circ/\text{GW}/\text{cm}^2$ were observed in the strongly depolarized parts of the beam.¹⁹

The solution is to add a polarizer to the end of the laser chain to prevent the "contamination" of the beam by weakly depolarized light. The conversion efficiency increased significantly as shown in Fig. 5. Residual alignment errors introduced by the prototype 74cm aperture polarizer are believed to be the source of the slightly low conversion efficiency and can be removed by improving the optical quality of the polarizer substrate. However, using the polarizer, 3ω conversion efficiencies approaching 70% have been demonstrated at the Nova scale.

Conclusion

The development over the last five years of large aperture frequency conversion arrays for the Nova laser has propelled this technology to a new level of performance. Frequency conversion now incorporates state-of-the-art crystal architectures, crystal orientation accuracy,

optical finishing techniques, AR coatings, and mounting techniques. Just one beam of the Nova laser has already produced record levels of 351nm ultraviolet light (>5kJ, 1ns). All ten Nova beams together can produce >50kJ of ultraviolet light at 1ns and even more energy at longer pulse widths. Using this crystal array technology, there is no fundamental limitation on continued scaling of the aperture and therefore the output energy and power of future laser systems.

Figures

1. A Nova 27x27x1 cm KDP crystal is immersed in an oil shower for temperature stability during the machining process. The single point machining process is used to orientate the surface normal and internal crystal structure as well as generate the final optical finish.
2. A crystal coating tank is used to apply the sol-gel antireflection coating on the Nova KDP crystals. The crystal is mounted on a spinning plate located in the center of the tank and the coating solution (0.75% SiO_2 in ethanol) is applied using a syringe mounted above while the crystal is spinning at about 380rpm.
3. The 74 cm clear aperture, KDP crystal array converts the Nova infrared light to visible or ultraviolet light with high conversion efficiency. Either wavelength can be selected by simple tilt and rotation adjustments of the entire array. Individual crystals are mounted on both sides of a deep-web "egg crate" which has been single point machined flat so that the array performs like one monolithic crystal.
4. These Nova 2ω burn patterns show the extent of depolarization in a Nova beam. The crystal array is aligned for peak conversion efficiency in the left image. Assuming linearly polarized 1ω input, the crystal array was rotated 45° about the beam axis in the right image, so that only the depolarized component of the beam can be converted to 2ω and burn the film.
5. The conversion efficiency observed on Nova beam number 10 before and after the addition of a 74cm polarizer. The polarizer eliminates depolarization contamination of the 3ω conversion and allows Nova to approach the high performance seen in small-scale studies.

References

1. P. A. Franken, et al., Phys. Rev. Lett., 7, 118 (1961).
2. J. A. Giordmaine, "Mixing of Light Beams in Crystals," Phys. Rev. Lett., 8, 19 (1962).
3. P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage, "Effects of Dispersion and Focusing on the Production of Optical Harmonics," Phys. Rev. Lett., 8, 21 (1962).
4. J. Nuckolls, L. Wood, A. Thiessen, and G. Zimmerman, "Laser Compression of Matter to Super-High Densities: Thermonuclear (CTR) Applications," Nature, Vol. 239, September 15, 1972.
5. KMS Fusion, Inc., "1977 Annual Report on Laser Fusion Research," KMSF-U762, pp. 2-17.
6. F. Amiranoff, et al., of GILM Ecole Polytechnique, "Interaction Experiments at Various Laser Wavelengths," presented at the Twenty-first Annual APS Meeting Division of Plasma Physics, Boston, Mass., Nov. 12-16, 1979.
7. R. S. Craxton, "Theory of High Efficiency Third Harmonic Generation of High Power Nd:Glass Laser Radiation," Optics Communications, Vol. 34, No. 3 (September 1980).
8. W. Seka, et al., "Demonstration of High Efficiency Third Harmonic Conversion of High Power Nd:Glass Laser Radiation," Optics Communications, Vol. 34, No. 3 (September 1980).
9. G. J. Linford, et al., "Large aperture harmonic conversion experiments at Lawrence Livermore National Laboratory," Applied Optics, Vol. 21, No. 20 (October 15, 1982).
10. G. J. Linford, W. Seka, et al., "Large aperture harmonic conversion experiments at LLNL: comments," Applied Optics, Vol. 22, No. 13 (July 1, 1983).

11. J. F. Holzrichter, E. M. Campbell, J. D. Lindl, and E. Storm,
"Research with High-Power Short-Wavelength Lasers," Science,
Vol. 229, No. 4718 (September 13, 1985).
12. "Frequency Conversion for High Power Lasers," Technology Transfer
Symposium, available from Lawrence Livermore National Laboratory.
13. D. Eimerl, et al., "Laser Fusion with Green and Blue Light," E&TR,
UCRL-52000-82-8 (August 1982).
14. M. A. Summers, B. C. Johnson, J. D. Williams, and L. G. Seppala,
"Nova Frequency and Focusing System," Laser Program Annual Report,
UCRL-50021-83 (1983) p. 2-8.
15. D. Milam and I. M. Thomas, "Sol-Gel Coatings," Laser Program Annual
Report, UCRL-50021-84 (1984) p. 6-39.
16. P. D. Maker, R. W. Terhune, and C. M. Savage, "Intensity-Dependent
Changes in the Refractive Index of Liquids," Scientific Laboratory,
Ford Motor Company, Dearborn, Michigan, Phys. Rev. Lett., Vol. 12,
No. 18 (May 4, 1964) p. 507.
17. A. Owyong, R. W. Hellwarth, and N. George, "Intensity-Induced
Changes in Optical Polarizations in Glasses," California Institute of
Technology, Pasadena, California, Phys. Rev. B, Vol. 5, No. 2
(January 15, 1972) p. 628.
18. V. N. Alekseev, D. I. Dmitriev, A. N. Zhilin, and V. N. Chernov,
"Depolarization of the exit beam of a neodymium-glass amplifier in
the case of small-scale self-focusing," Sov. J Quantum Electron.
13(4) (April 1983) p. 533.
19. M. Henesian, et al., to be published.



Figure 1

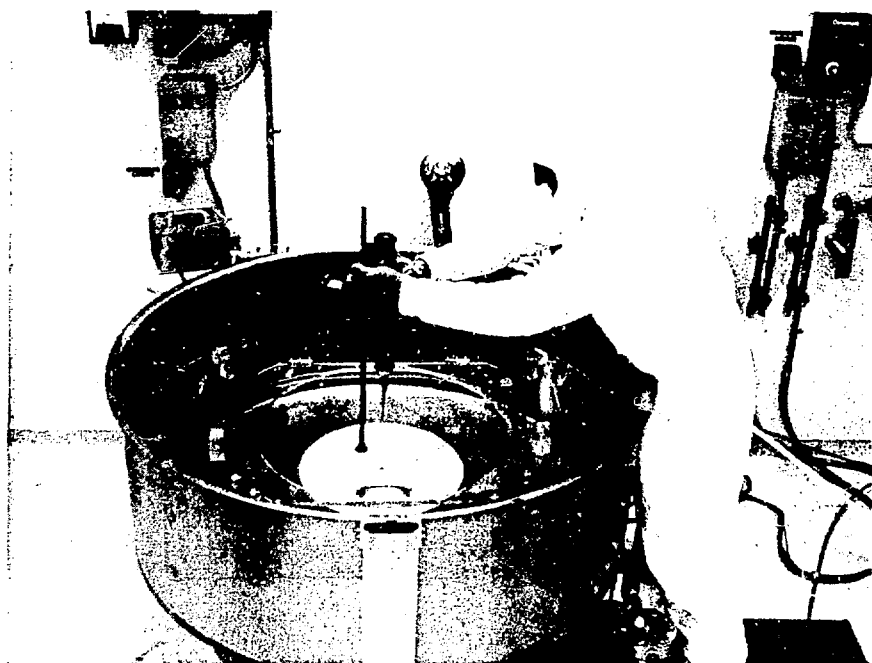


Figure 2

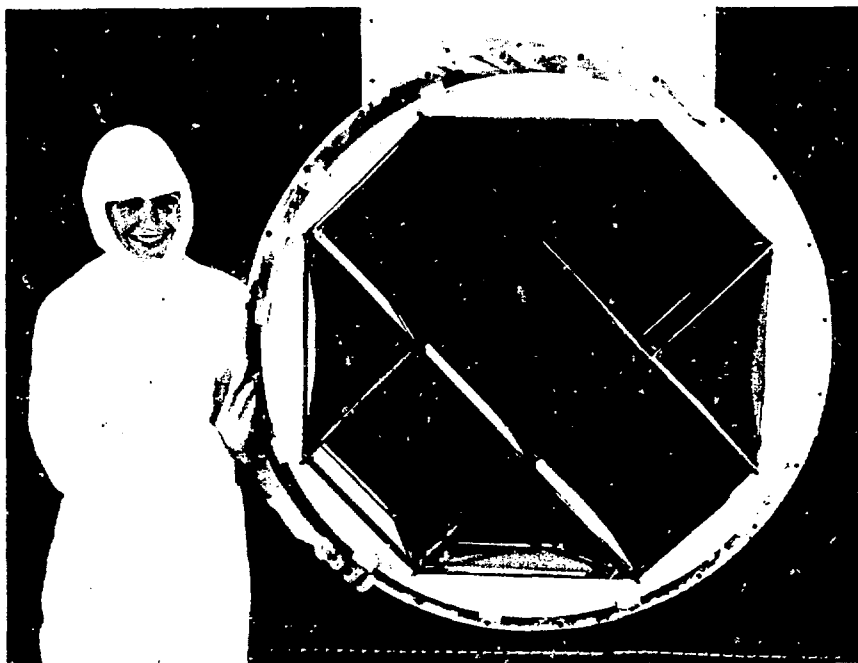
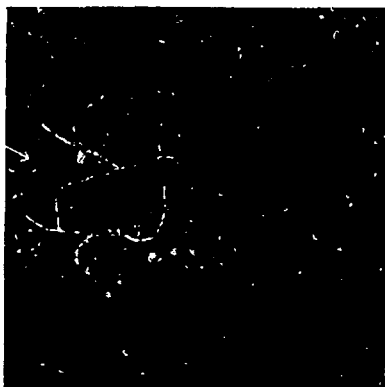


Figure 3

Normal array alignment

$\vec{E}_{1\omega}$ at 45° to "0" axis



Orientation for peak conversion efficiency

Null orientation

$\vec{E}_{1\omega}$ at 0° to "0" axis



Only depolarized light will convert in this orientation

Figure 4

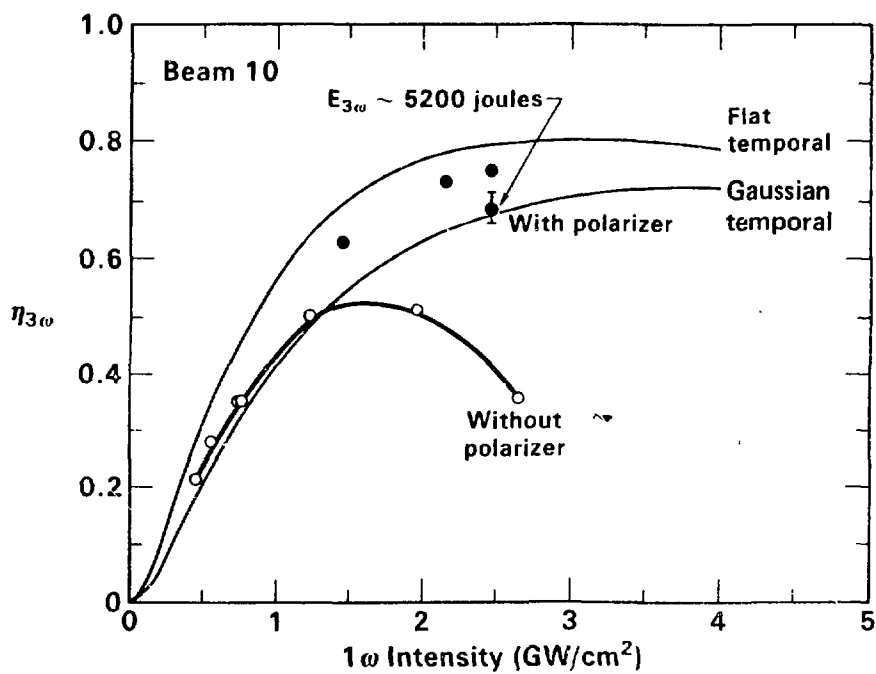


Figure 5