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Modeling of Large Aperture Third Harmonic Frequency Conversion of
High Power Nd:Glass Laser Systems

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1. ABSTRACT

To provide high-energy, high-power beams at short wavelengths for inertial-confinement-fusion experiments, we routinely convert the 1.053- μm output of the Nova, Nd:phosphate-glass, laser system to its third-harmonic wavelength. We describe performance and conversion efficiency modeling of the 3 x 3 arrays potassium-dihydrogen-phosphate crystal plates used for type I/type II phase-matched harmonic conversion of Nova 0.74-m diameter beams, and an alternate type I/type II phase-matching configuration that improves the third-harmonic conversion efficiency. These arrays provide energy conversion of up to 65% and intensity conversion to 70%.

2. INTRODUCTION

Large aperture Nd:Glass laser systems, at 1.053- μm wavelength are operated at a number of laboratories worldwide for inertial confinement fusion (ICF) research and laser-plasma interaction studies. These systems are typically converted to shorter wavelengths via harmonic frequency conversion in nonlinear crystals such as KDP (potassium-dihydrogen-phosphate). At Livermore, the 0.74-m aperture beams of the Nova Nd:Glass laser system (Ref. 1) are converted to the third harmonic wavelength (0.351 μm) using type II/type II phase matching in 3 x 3 arrays of 27 x 27-cm square by 1-cm thick KDP crystals (Ref. 2). Energy conversion from the fundamental to the third harmonic is limited on Nova to ~55% efficiency, primarily due to inhomogeneous depolarization of the 1.053- μm beam in the few percent range, to intensity-dependent nonlinear optical effects, and to light scattering losses at crystal surfaces (Ref. 3). The unusual temporal pulseshaping requirements of ICF also influence the energy conversion efficiency (Ref. 4,5). A third-harmonic conversion scheme that is less sensitive to depolarization has been devised using type I/type II phase-matching in KDP (Ref. 6). We have built a 3 x 3 array of 27 x 27-cm square by 1.2-cm thick, type I doubling crystals, followed by 1-cm thick type II tripling crystals and tested it on a Nova beamline (Ref. 6, 7). With this design we achieved over 65% energy conversion for square temporal pulses of 1 to 2.5-ns width. Aperture-averaged intensity conversion to 0.351 μm reached 70%. In addition, a record 8.1 kJ of 0.351- μm radiation was generated from a 12.5-kJ, 2.5-ns square drive pulse. We also generated 5 TW of 0.351- μm radiation in a 1-ns square pulse, and in the trailing 1-ns portion of a highly-shaped ICF pulse we achieved a peak power of 4.5 TW. Similar results were obtained at 1 ns by repolarization of the 0.74-m diameter Nova beam with a full-aperture thin-film dielectric polarizer (Ref. 7).

In this report we will discuss the design of the frequency conversion crystal arrays and compare type I/type II third harmonic conversion with the established type II/type II design. In particular we will discuss the problem of obtaining high efficiency with moderately depolarized (1 ~ 4 %) beams. The model we use to simulate frequency conversion of the Nova beam includes a depolarization distribution model for Nova (Ref. 8), a general radial-type depolarization distribution applicable to telescope-relayed laser systems, intensity-dependent polarization ellipse rotation over the system propagation paths (air or argon gas filled) (Ref. 9), B-integral dependent ellipse rotation effects (Ref. 10), birefringence effects from dielectric beam steering mirrors, beam repolarization with finite extinction ratio polarizers (Ref. 7), and consideration for intensity statistics (Ref. 11), and pulseshape effects (Ref. 4). Our intensity model includes a new type

of statistical distribution, that we have developed for apodized high power laser beams, that we are calling "super-Rician" statistics.

3. HARMONIC CONVERSION ARRAY DESIGN

Craxton and colleagues of the Laboratory for Laser Energetics (LLE) at the University of Rochester discussed a number of two-crystal schemes employing type I and type II phase-matching in KDP for generating the third harmonic (Ref. 12). Similar schemes were also proposed and tested at LLNL, by Linford and coworkers (Ref. 13). Third-harmonic generation (THG) typically requires two crystal plates in series; that is, a "doubling" crystal followed by a "tripling" crystal, in which the second harmonic (2ω) is mixed with the remnant first harmonic (1ω) to produce the third harmonic (3ω). The THG schemes attractive for ICF laser applications are: first, the polarization-mismatch scheme which uses two type II crystals (Fig. 1(a)); and second, an angle-detuning scheme which uses a type I doubling crystal followed by a type II tripling crystal (Fig. 1(b)).

Fig. 1(a). Type II/type II crystal scheme and polarization directions for THG

Fig. 1(b). Type I/type II crystal scheme and polarization directions for THG

For optimum THG in either scheme, the doubling crystal must convert the fundamental radiation to the second harmonic with ~66.7% efficiency to provide a 1:1 "mix" of 2ω to 1ω photons in the tripling crystal for optimum sum-frequency-mixing. The corresponding 2:1 ratio of 2ω to 1ω intensity is termed the " $2\omega/1\omega$ mix ratio". In the type II/type II scheme (Fig. 1a) the $2\omega/1\omega$ mix ratio is controlled by orienting the electric field polarization of the 1ω beam at a specific angle (~35°) with respect to the ordinary (o)-axis of the doubler, thus controlling the type II doubling efficiency. In the type I/type II scheme (Fig. 1(b)) the $2\omega/1\omega$ mix ratio is controlled by angle-tuning (i.e., tilt-adjusting) the extraordinary (e)-axis of the first crystal relative to the phase-matching direction, thus controlling the type I doubling efficiency. Second-harmonic generation (SHG) may also be done with these configurations using one or both of the crystals. For example, at room temperature, the type II phase-match angles in KDP for doubling and tripling 1.053- μm radiation are separated by < 6-mrad, enabling one with a small adjustment in tilt angle, to phase-match for either SHG or THG. In the type I/type II arrangement the type I crystal is always used for SHG; although one can rotate the crystal pair about the beam direction, and angle-tune the type II crystal for additional SHG.

In the configurations above, note that the crystals are oriented with their ordinary (o)-axes orthogonal to one another (i.e., in "quadrature" (ref. 14)). Neither arrangement requires a waveplate or polarization component between crystals. The two crystals are tilt-adjusted for phase-matching about orthogonal (o)-axes, so doubler and tripler tuning are nominally independent.

For efficient type II/type II THG, the crystal combination is rotated about the beam direction to position the (o)-axis of the doubler at 35.3° to the 1ω electric field vector, and tilt-adjusted about the ordinary axes to achieve 2ω phase-matching in the doubler and 3ω -matching in the mixer. With the 1ω electric-field-polarization angle at 35.3° , the ratio of 1ω intensity along the (o)-axis to 1ω intensity along the (e)-axis is 2:1 (i.e., the square of the cotangent of the angle) which provides the near optimum $2\omega/1\omega$ mix ratio at the tripling crystal. This ratio of $1\omega(o)$ to $1\omega(e)$ intensity incident on the doubler, termed the " 1ω mix ratio", is critical for type II/type II tripling. Figure 2(a) plots type II/type II tripling efficiency versus the 1ω mix ratio for several 1ω drive intensities. The calculations assume flat-in-space-and-time beams and misalignment from exact phase matching of $50 \mu\text{rad}$ (internal angle). Crystal absorption loss and surface losses (1.5% per surface) are included. At high drive intensity ($>2 \text{ GW/cm}^2$) the acceptable 1ω mix ratio is restricted to values between $1.7 \sim 2.6$. As the 1ω intensity increases this range significantly narrows.

Fig. 2(a). 3ω conversion efficiency vs. 1ω mix ratio as a function of 1ω intensity: 0.5(a), 1.0(b), 2.0(c), 3.0(d), and 4.0(e) GW/cm^2 .

Fig. 2(b). 1ω mix ratio vs. ellipse orientation angle to (o)-axis, as a function of ellipticity: 0.0(a), 0.1(b), 0.16(c), 0.23(d), 0.33(e), 0.5(f).

To examine the effect of elliptical polarization on the 1ω mix ratio, Fig. 2(b) plots this 1ω mix ratio as a function of orientation angle of the major axis of the polarization ellipse to the (o)-axis, for electric-field ellipticities ranging from linear ($\epsilon = 0$) to elliptical ($\epsilon = 0.5$). In the reference frame of the ellipse, $\epsilon = \sqrt{(p/1-p)}$, where p is the depolarized light fraction. Fig. 2(a) shows that in the type II/type II tripling scheme, the input 1ω polarization angle (or major axis of the electric-field ellipse) must be controlled to better than a degree or two. As Fig. 2(b) shows, an increase in ellipticity (i.e. depolarization) at a fixed orientation angle causes the 1ω mix ratio to decrease, typically causing a drop-off in conversion efficiency, particularly for intensities $>3 \text{ GW/cm}^2$.

Figure 3 illustrates the relationship between the polarization ellipse and the 1ω mix ratio. As drawn in the reference frame of the crystal (o) and (e)-axes, the corner of the rectangle that circumscribes the ellipse defines an effective polarization angle, α . The 1ω mix ratio is simply the square of the cotangent of α .

In crystal frame (E_x, E_y):

Ellipticity:

$$\epsilon = \pm E_y / E_x = \tan(\chi)$$

$$\eta = E_y / E_x = \tan(\alpha)$$

Mix ratio:

$$E_x^2 / E_y^2 = \cotan^2(\alpha)$$

Polarization angle:

$$\alpha = 0.5 \cos^{-1}(\cos(2\psi) \cos(2\chi))$$

Fig. 3. Diagram of polarization ellipse in crystal reference frame.

As seen from the figure above, increasing the ellipticity, increases the polarization angle α , and decreases the 1ω mix ratio. At a nominal ellipse orientation of $\psi \sim 35.3^\circ$ the changes in polarization angle for the range of ellipticities of interest ($0 < \epsilon < .3$) is less than two degrees. While the effect of the decrease in 1ω mix ratio (from 2.0 to 1.77) on THG is not insignificant at intensities $>4 \text{ GW/cm}^2$, the predicted reduction in efficiency at moderate intensities is insufficient to describe the drop from 70% to 55% that we have observed in Nova experiments. When the orientation of the ellipse can change as a function of intensity due to nonlinear optical effects in the laser system or propagation paths, then the 1ω mix ratio can change dramatically, causing serious loss in THG efficiency (Ref. 3). The ellipse orientation change is proportional to ϵ , and the accumulated B-integral over the propagation path (Ref. 10); and therefore, reducing the sources of optical birefringence in the laser amplifier chain and transport path are critical to the laser system design.

Notice from Fig. 2(b), that at $\psi = 45^\circ$ the 1ω mix ratio is not sensitive to ellipticity. An alternate type II/type II scheme can be devised where the $2\omega/1\omega$ mix ratio at the tripler is controlled by angle-tuning rather than polarization-tuning. As we can see from Fig. 2(a), for 1ω mix ratios near unity, this scheme cannot achieve a conversion efficiency above ~42%. Intensity-dependent ellipse rotation effects will reduce this further. Our experience on Nova has shown that the sensitivity of type II/type II tripling to the state of the polarization is a serious disadvantage, even for depolarization of only a few percent (Ref. 2). The type I/type I and type II/type I tripling schemes discussed by Craxton (Ref. 12) are also polarization-tuned, and as such are subject to the same polarization sensitivity as the type II/type II scheme used at LLE and LLNL.

In the type I/type II scheme (Fig. 1(b)), the depolarized component of the 1ω beam, oriented along the (e)-axis of the doubler, is not phase matched in either crystal and is effectively rejected. The $2\omega/1\omega$ mix ratio at the tripler is controlled by angle tuning the doubling crystal about its phase-matching direction (Ref. 6). The tolerance on this angle becomes tighter at high 1ω drive intensities. Figure 4(a) plots the THG efficiency versus the tuning angle from exact

type I phase matching for several 1ω drive intensities. The calculations assume an ideally polarized beam, flat-in-space-and-time. At high 1ω intensity the 3ω efficiency is optimum for doubler detunings of between 175 and 225 μrad (internal angles). Below 2 GW/cm^2 , however, the doubler tuning range is very broad ($\sim \pm 275 \mu\text{rad}$), which makes the type I/type II scheme very attractive at moderate intensities. For comparison, the angular sensitivity of the tripling crystal (identical in both schemes) is shown in Fig. 4(b). At moderate 1ω drive intensities, the tripler tuning sensitivity is comparable to that of the type I doubler. As the 1ω drive increases the tolerance for angular misalignment from phase matching decreases; but not as severely as the tolerance of the type I doubler for intensities beyond $> 2.5 \text{ GW}/\text{cm}^2$.

Fig. 4(a). Type I/type II THG efficiency vs. angular detuning of the type I doubler, as a function of 1ω intensity: 0.5(a), 1.0(b), 2.0(c), 3.0(d), 4.0(e) GW/cm^2 .

Fig. 4(b). THG efficiency vs. angular detuning of the type II tripler, as a function of 1ω intensity: 0.5(a), 1.0(b), 2.0(c), 3.0(d), 4.0(e) GW/cm^2 .

The type II/type II and type I/type II schemes can both provide high SHG and THG conversion efficiency over a selected range of drive intensities which can be fixed with appropriate choices of crystal thicknesses. On Nova the intensities in shaped ICF pulses incident on the conversion arrays can reach up to $3.5 \text{ GW}/\text{cm}^2$. To maximize the energy delivered to targets requires that the crystal thicknesses be chosen to optimize conversion efficiency over as large a range as possible, up to these peak intensities. We calculated the THG efficiency at specific intensity values in the proposed operating range, generating contours of constant conversion efficiency, similar to those shown in Figures 5 and 6 below, for type II/type II and type I/type II schemes as functions of first (L1) and second (L2) crystal thicknesses. The calculations assume ideal linearly-polarized, flat-in-space-and-time beams. To account for angular misalignment, we assumed errors of $\pm 50 \mu\text{rad}$ for type II crystals, and averaged results obtained with 150 and 250 μrad detuning for the angle-tuned type I crystals. In general, selecting the thinnest crystals that give the desired THG performance will bias the operation range towards higher intensities, and reduce sensitivity to angular misalignment. Similar plots for SHG indicate that thickening the crystals will improve the second-harmonic performance, while significantly increasing the angular sensitivity for tripling. On Nova we use 1.0-cm thick type II doublers and triplers, optimized for tripling efficiency, and 1.2-cm thick type I doublers, here again optimized for type I/type II tripling. For ideal flat-in-space-and-time beams, Figs. 5 and 6 indicate that peak THG efficiency can exceed 82% for type II/type II THG, and reach 75% for type I/type II THG at intensities above $2 \text{ GW}/\text{cm}^2$. This is rarely achieved in practice, however (Ref. 12).

Fig. 5(a)

Fig. 5(b)

Fig. 5(c)

Fig. 5. Contours of constant THG efficiency for type II/type II THG vs. thickness (cm) for first (L1) and second (L2) crystals at 1ω intensities: 1.0(a), 2.0(b), 3.0(c) GW/cm^2 .

Fig. 6(a)

Fig. 6(b)

Fig. 6(c)

Fig. 6. Contours of constant THG efficiency for type I/type II THG vs. thickness (cm) for first (L1) and second (L2) crystals at 1ω intensities: 1.0(a), 2.0(b), 3.0(c) GW/cm^2 .

A well engineered frequency conversion design will be one in which the angular sensitivity is consistent with the accuracy with which the crystals can be aligned to the laser beam (Ref. 15). On Nova, a total of eighteen crystals, arranged back-to-back in a 3×3 array, are used to frequency convert each 0.74-m beam. The largest crystal plates are 27-cm square, chosen to be within the size of available KDP boules, and to avoid potential transverse stimulated-Raman-scattering losses in KDP at 3ω (Ref. 16). The crystals are mounted in a mechanical structure, called an "eggcrate", which holds each set of crystals parallel to form one monolithic set of input and output crystals. The crystals are registered mechanically to the two outer surfaces of the eggcrate which are precision diamond-turned to $< 10\text{-}\mu\text{m}$ flatness, holding the crystals parallel to within $\pm 30\text{ }\mu\text{rad}$ (referenced to internal crystal angles). The Nova crystal plates themselves are individually machined with diamond-bit tools to provide surfaces that are flat and parallel ($< 5\text{-}\mu\text{rad}$ wedge), and to precisely orient the surface normal with respect to the 2ω and 3ω phase-match angles. To insure this precision, each crystal is measured on a crystal orientation measurement system at several stages during the machining process. The angular errors budgeted for the Nova KDP arrays (in internal angle) are $\pm 33\text{ }\mu\text{rad}$ for orienting the surface normal with respect to the crystallographic axes, $\pm 33\text{ }\mu\text{rad}$ for crystal to crystal alignment in the array, and $\pm 33\text{ }\mu\text{rad}$ for alignment of the array to

the Nova beam (Ref. 17). Because of increased angular sensitivity of the type I/type II scheme at high power, we worked for a tolerance of $\pm 15 \mu\text{rad}$ for orienting the surface normal of the type I crystals. For the modeling presented in the next section, the individual errors are assumed to sum in a root-mean-square fashion, resulting in maximum alignment errors of $\pm 55 \mu\text{rad}$ and $\pm 45 \mu\text{rad}$ (referenced to internal crystal angles) for type II and type I crystals, respectively.

A final finishing cut on the diamond-turning machine reduces the crystal surface roughness to ~ 30 angstroms RMS. This improves the optical transmission of the crystal by reducing surface scatter and by providing a high-quality surface on which to apply an antireflective (AR) coating. The development of sol-gel AR coatings for KDP (Ref. 18) has eliminated the need for large optical windows and index-matching fluid, greatly simplifying the design of large area crystal arrays. For Nova arrays, a $1\omega/2\omega$ compromise coating with a nominal transmission of 99.1% at 1.053 and $0.526 \mu\text{m}$ is applied to the first three crystal surfaces. A $2\omega/3\omega$ compromise coating with a nominal transmission of 99.6% at 0.526 and $0.351 \mu\text{m}$ is applied on the final surface. The crystal arrays for the our tests were constructed from standard Nova hardware and spare or used crystals, with the exception of the type I crystals which were fabricated specifically for these tests. The crystals used in the type II/type II array had been in prior use on Nova over three years, and transmission was below specification by 1-3 % percent because of environmental degradation of the crystal surfaces. Measured surface losses were taken into account for our calculations (presented below) that are compared against data.

4. DESCRIPTION OF MEASUREMENTS

To test these arrays in the full 0.74-m-aperture beam at the maximum energy and power available from a rebuilt Nova beamline (Ref. 5) we constructed a diagnostic station configured to measure the energy, pulseshape, and beam parameters of the frequency-converted beam following the crystal array (Ref. 7). Measurements of the 1ω beam input to the array was provided by the Nova output sensor, thus completing the information needed to directly assess frequency conversion efficiency. The crystal array at the diagnostic station was located 68 m from the end of the laser-amplifier chain, 8-m further than the longest propagation path to the Nova Ten Beam target chamber. Most of the measurements were made using flat-in-time pulses of 1-ns and 2.5-ns duration, with incident 1ω energies up to 8.5 kJ and 12.5 kJ, respectively. The diagnostic station used full-aperture, fused-silica beamsplitters situated at low angles of incidence to sample and attenuate the high-power frequency-converted beam and direct it along various paths. A Galilean telescope in each beam path reduced the size of the beam for calorimetry, temporal pulseshape measurement, and near-field or far-field photography. We estimate that the 1ω energy was measured to within 4.4% precision, based on the repeatability of calibrations, and to an accuracy of 7%. The measurement of the harmonic energy was precise to within 4.5% and accurate to within 9%. The temporal shapes of the 1ω and the 2 or 3ω pulse were measured with ~ 20 -ps resolution with standard LLNL streak cameras.

A small portion of the 1ω beam incident on the array was directed to a 1-meter spectrometer to monitor for the presence of frequency-shifted spectral components produced by stimulated-rotational-Raman scattering (SRRS) of the 1ω beam in the airpath to the diagnostic station. Spectral components at the wavelengths expected from SRRS in N_2 were observed on 1-ns shots with intensities exceeding 2.2 GW/cm^2 . The corresponding IL product of 14.9 TW/cm^2 is in agreement with the published SRRS threshold values in air at this pulse length (Ref. 18). After this observation, we isolated a 31-m section of airpath using fused-silica windows and back-filled it with argon, decreasing the effective airpath from 68 to ~ 45 meters (31 m of air plus an additional 8 meter contribution from the windows), and increasing the SRRS threshold by several orders of magnitude. No SRRS lines were observed with this shorter air-propagation path.

5. MEASUREMENTS AND CALCULATIONS OF CONVERSION EFFICIENCIES

We summarize the type II/type II THG measurements without beam repolarization in Fig. 7, which shows the conversion efficiency, defined as the ratio of 3ω fluence generated by the crystals to the 1ω fluence incident on the crystals, as a function of the incident 1ω intensity. The measured efficiency peaked at $\sim 56\%$ at a drive intensity of 1.5 GW/cm^2 and remained approximately constant, but with some small decrease as the intensity increased. Our model for Nova frequency conversion is compared with the data in the figures. The three calculated bands shown in Fig. 7 are with successively more detailed beam characteristics in the model.

Fig. 7. Plot of measured type II/type II THG vs. 1ω drive intensity for 1-ns (■,□) and 2.5-ns (○) square-in-time pulses. 1-ns pulses (■) were obtained with 31 m of the 68-m air-path replaced with argon. Bands are calculated with different beam characteristics in model. Upper band assumes perfect polarization, but includes beam statistics (see text). Middle band (dark) assumes 31 m of argon in path, lower band (light) assumes 68 m of air-path. Beam depolarization of 1.67% is assumed for middle and lower bands, and also includes 1ω beam statistics (see text).

The upper-most band is the range of efficiencies calculated when we include measured surface scatter-losses of the crystal array and representative distributions of intensities approximating the measured spatial intensity statistics of the incident 1ω beam. We developed a modified form of the Rician intensity distribution for laser intensity statistics, applicable to apodized high power beams, and include it as part of the conversion efficiency model. To validate the model we photo-densitometered and analyzed photographs of output Nova beams from shots over a wide range of intensities. The observed time-averaged statistical distributions of spatial-intensity were carefully fit by averaging a "flat beam" Rician density function for laser intensity noise (Ref. 11) with a "supergaussian" spatial envelope function. A Rician parameter of 0.01 representing the ratio of the noise intensity variance to the envelope intensity, and supergaussian envelope functions of from 4th to 10th power best fit the data, requiring that we plot the conversion theory as bands. The measured temporal pulseshapes were best approximated by an 8th power supergaussian profile. The conversion efficiency predicted by this model (upper band, Fig. 7) is substantially higher than that measured, indicating that beam statistics alone is insufficient for modeling real system performance.

We attribute the poorer than predicted conversion efficiency to a small nonuniform electric-field depolarization (~1.7% of the beam energy) of the Nova beam and to intensity (or B-integral) dependent polarization effects (Refs. 10). We have surmised from collateral experiments that a small amount of stress birefringence in the spatial-filter lenses, resulting from their use as vacuum windows, combined with residual unannealed and thermally-induced stresses in the laser disks in the output amplifier stages is the cause for depolarization of portions of the normally horizontally-polarized beam (Ref. 8). Measurements of the Stokes parameters made in the depolarized region of an output beam (Ref. 10) and photographs of the depolarized fraction of the 1ω beam showed that the polarization varies across the aperture and changes with beam intensity. The depolarized light fraction was measured directly using calorimetry and estimated by using the "2 ω null technique" (Ref. 8). Figure 8 is a photograph of the depolarized light from the 1ω Nova beam used in our experiments, obtained by reflection of the out-of-plane electric field component of the beam from a large glass plate placed at Brewster's angle in the beam path. Densitometry analysis of the photo showed that small regions of the beam were observed to have as much as 8% depolarization, but more typically, 50% of the beam area was at a depolarization of 1% or less. The aperture-average value for our test beamline was 1.67%.

Fig. 8(a). Photograph of depolarized light in full aperture Nova beam (dark on light background)
Depolarization is $\sim 1.7\%$.

Nova Depolarization Level Model

Level	Depol(%)	Ellipticity	Weight (%)
1	0	0	18.0
2	.25	.050	9.1
3	.25	-.050	9.1
4	.75	.087	6.2
5	.75	-.087	6.2
6	1.50	.123	8.8
7	1.50	-.123	8.8
8	2.50	.160	6.2
9	2.50	-.160	6.2
10	3.50	.190	4.75
11	3.50	-.190	4.75
12	4.50	.217	3.10
13	4.50	-.217	3.10
14	5.50	.241	1.70
15	5.50	-.241	1.70
16	7.00	.274	1.15
17	7.00	-.274	1.15

In our current Nova model, we simulate the polarization of the beam at the output of the laser chain as a set of discrete elliptical polarizations states with area weight factors determined from photographs of the beam similar to the one shown in Fig. 8. At low intensity, the orientation of the major axes of the ellipses are assumed to be directed along the nominal polarization direction. Calculation of the expected conversion efficiency is done by calculating the intensity-dependent polarization-ellipse rotation (ICSP) in the airpath to the crystal array for each of 17 discrete elliptical polarization states (shown in the table above), and calculating a THG efficiency versus intensity curve for each state by numerically integrating the differential equations for frequency doubling and tripling (mixing) (Ref. 12). In this model, the orientation of the major axis of the electric-field ellipse is linearly proportional to the length of air-path, the ellipticity parameter, $\epsilon/(1+\epsilon^2)$, and the 1ω intensity. Ellipsometry equations in the code relate the state of the ellipse for a given depolarization level, at a given 1ω intensity, to the 1ω mix ratio at the input crystal. The average of the THG efficiency curves for all discrete polarization states weighted by their relative areas yields a THG efficiency curve for the distribution, neglecting intensity statistics and pulsed shape. The super-Rician distribution mentioned previously, is then mathematically applied to the "flat-in-space-and-time" efficiency curve to find the expected conversion efficiency at the average 1ω beam intensity. For non-square-in-time pulses the spatially-averaged efficiency curve can then be applied to each small time-slice of the pulse to obtain the converted pulsed shape and overall energy conversion efficiency (Ref. 4). To consider beam average depolarization values other than 1.67%, we uniformly scale up (or down) the ellipticity values in the table, maintaining the area weight factors, until we obtain the desired area average depolarization.

Figure 9(a) shows results of the Nova conversion efficiency model for beam-average depolarizations ranging from 0 – 4% at a fixed air-path length of 68 m. Super-Rician intensity statistics with a noise parameter of .01 and a 10th power spatial supergaussian parameter were used. In addition to the Nova depolarization model, we have constructed a radial birefringence model, that partitions the aperture of the beam into a large number of annular rings and radial sectors. The model assumes that the difference between radial and tangential refractive index increases quadratically with radius. Detailed ellipsometry is computed for each sector/annuli of the beam to determine the ellipticity and ellipse orientation. Results of this model, shown in Fig. 9(b), are in good agreement with the original Nova model. Less conversion efficiency loss at 2–4 % depolarization is predicted by the radial model. Nevertheless, to achieve a 3ω conversion >70% requires that the 1ω depolarization be kept less than 0.5% in either model. For acceptable performance above 2 GW/cm^2 , we require even more stringent reductions in depolarization, or a significant reduction in the air-path length from the laser amplifier output to the crystal arrays, as we will see next.

Fig. 9(a). Nova depolarization model: THG efficiency as a function of aperture-average depolarization: 0%(a), 0.25%(b), 0.5%(c), 1.0%(d), 2.0%(e), 4.0%(f)

Fig. 9(b). Radial depolarization model: THG efficiency as a function of aperture-average depolarization: 0%(a), 0.25%(b), 0.5%(c), 1.0%(d), 2.0%(e), 4.0%(f)

Figure 10(a) illustrates the effect of air-path length on 3ω conversion at a fixed depolarization of 1.67%, with the same super-Rician statistics used before. An alternative to reducing the beam depolarization is to reduce the air path from the laser system output to the crystal arrays. To achieve >70% conversion efficiency requires that the path be reduced to < 15 m at nominal Nova depolarization levels. Ellipse rotation effects in windows, and fill gases must be considered. To illustrate the ICSP effect (Ref. 9), we plot the polarization ellipse orientation angle as a function of 1ω intensity in Fig. 10(b). The ellipse rotation measurements shown were performed on Nova with a 10-cm diameter test beam with a fixed ellipticity of $\epsilon \approx 0.5$, and were conducted over a 68-m path (Ref. 9). When a substantial fraction of the beam path was filled with argon gas the ellipse rotation rate was substantially reduced.

The equation relating the orientation of the major axis of the ellipse $\psi(z,t)$ in the crystal reference frame (see Fig. 3) is given by

$$\psi(z,t) = \psi(0,t) + C \cdot z \cdot I(t) \cdot \epsilon / (1 + \epsilon^2), \quad (1)$$

where $\psi(0,t)$ is the initial orientation, z is the path length in meters, $I(t)$ is the intensity (GW/cm^2) at time t in the pulse, ϵ is the electric-field ellipticity ($\epsilon > 0$ for clockwise electric field rotation, as viewed looking into beam), and the nonlinear index coefficients (related to the third-order-susceptibility $\chi_{123}(-\omega, \omega, \omega, -\omega)$) are $C = 0.0045$ for air at 1 atm, $C = 0.00038$ for argon at 1 atm, and $C = 3.39$ for glass (BK-7) in units of radians/meter per GW/cm^2 (Ref. 9). The linear dependence of ellipse tilt angle with intensity was experimentally observed (Fig. 10(a)).

Fig. 10(a). THG efficiency as a function of air-path length, assuming a beam depolarization of 1.67%. Air-paths(m): 0(a), 15(b), 45(c), 68(d), 90(e)

Fig. 10(b). Measurements of intensity-dependent ellipse rotation for $\epsilon = 0.5$ (Ref. 9).

The Nova depolarization model predicts the lower (light) performance band in Fig. 7, which is in fair agreement with the efficiencies that we measured. Better agreement would result we believe from a more detailed description of the polarization states of the Nova beam. As a test of the model, and our assumptions concerning ellipse rotation (ICSP) over the air-path, the calculated conversion efficiency was compared to data obtained when 31 m of the air-path in the beamtubes was replaced with argon. Ellipse rotation in the thin fused-silica windows used to seal the beamtubes and from the argon gas was included in the calculation, and gives an effective air-path length of 45 m. The reduced airpath suppressed SRRS and reduced the polarization rotation; both of which improved the observed frequency conversion, shown by the intermediate (dark) band. The two data points obtained with the argon-filled beamtubes lie within the band. Further reduction of air-path length (or equivalent) would have increased the 3ω conversion even further at high drive conditions, $> 2.5 \text{ GW}/\text{cm}^2$, as indicated by calculations in Fig. 10(a).

Improving the polarization of the 1ω beam should significantly improve the efficiency of type II/type II THG. To demonstrate this, we repolarized the full-aperture beam immediately in front of the crystal array. A large (1.45-m)

two-piece thin-film-dielectric polarizer, consisting of SiO_2 and TiO_2 on fused-silica substrates, provided a variable extinction ratio via small changes to the incident angle of the beam on the polarizer coating. We measured the conversion efficiency at extinction ratios of approximately 20:1 and >500:1, and observed a marked increase in conversion efficiency but no measurable difference between the two extinction cases. In Fig. 11 we plot the conversion efficiency after losses from repolarization are accounted for. The results show an improvement from ~56% at 2 GW/cm^2 without beam repolarization to >68% at the same intensity with repolarization; a consequence of improved THG efficiency in the depolarized regions of the beam, and by elimination of any SRRS by reducing the air-path from 68 to 45 m. Near-field photographs of 3ω beams taken before repolarization of the 1ω beam confirmed the existence of regions of poor conversion that correlate with the regions of depolarization. Repolarizing the 1ω beam significantly improved the near-field uniformity of the 3ω beam (Ref. 7). Similar results were also obtained with type I/type II conversion, shown below in Fig. 12.

Fig. 11. Plot of measured type II/type II THG after 1ω beam repolarization for 1-ns square pulses with 68 m air-path (▲), and with argon in path (Δ). Upper band (dark) assumes perfect polarization and with beam statistics modeling (see text).

Fig. 12. Plot of measured type I/type II THG vs. 1ω drive intensity for 1-ns (■, □) and 2.5-ns (○) square pulses. 1-ns pulses (■) obtained with argon in air-path. Upper band (light) assumes perfect polarization, lower band (dark) assumes 1.67% depolarization, both including beam statistics modeling.

The highest 3ω energy produced with repolarization was 5 kJ converted from 8.1 kJ in a 1-ns pulse. The increased risk of catastrophic damage to the polarizer coating at longer pulse lengths, prevented us from making measurements at 2.5 ns. The measured improvement in THG efficiency is in agreement with the trend predicted by our model but falls ~10% below the calculated efficiency. The discrepancy is likely attributable to losses larger than the assumed 4% at the polarizer and/or larger than those estimated for the KDP crystal surfaces.

The measured THG efficiency of the type I/type II array was equal to that of the type II/type II array/polarizer combination throughout the range of fluences ($<2.5 \text{ J/cm}^2$) at which the latter scheme was investigated. Without the constraint imposed by optical damage to the polarizer, we produced higher energies with the type I/type II array, with over 8.1 kJ of 3ω energy from a 12.5-kJ, 2.5-ns drive pulse. Figure 12 shows the measured conversion efficiency versus intensity for the 1-ns and 2.5-ns pulse lengths along with the modeled performance bands. The upper band is without and the lower band is with depolarization included in the model, and illustrates the insensitivity of the type I/type II scheme to small amounts of beam depolarization. The model is identical to that used for the type II/type II calculations but with

crystal constants and angular errors appropriate for type I doubling. We assumed an angle-tuning error for the type I doubler of $\pm 45 \mu\text{rad}$, and took into account the doubler-tuning angle offset of $200 \mu\text{rad}$, where the tripling efficiency is a maximum. To calculate the efficiency with the estimated errors we took doubler tuning angles of $245 \mu\text{rad}$ and $155 \mu\text{rad}$, and plot the average to represent the conversion efficiency. We attribute the rolloff in conversion efficiency above 1.8 GW/cm^2 to Raman scattering in the 68-meter airpath to the crystal array. Filling the beam tube with argon enabled us to reach an efficiency of 69% at 2.3 GW/cm^2 .

6. SUMMARY

The next generation glass laser built for ICF research is presently envisioned to require 1.5 to 2.0 MJ of short-wavelength energy delivered to a target in a nominal 3-ns-duration pulse. In order to provide the high energy at a reasonable cost, components in the laser will have to operate reliably with low-loss and high efficiency. The frequency-conversion system is particularly crucial since, for a fixed energy on target, the size of the laser amplifier scales inversely as the product of conversion efficiency times the focus-optics transmission. An overall efficiency of third harmonic on target to incident fundamental energy of greater than 65-70% is highly desirable. Results of measurements reported here, confirmed by system modeling and simulation, give some assurance that high third-harmonic conversion efficiency of large beams is possible with KDP. To achieve the desired high overall system efficiency will require high quality, well polarized beams from the laser amplifier, efficient beam transport, and an optimized conversion system.

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