Section 3 TECHNOLOGICAL DEVELOPMENTS

3.A Monolithic Cell for Frequency Conversion

The origination¹ at LLE of the "polarization-mismatch" scheme for highly efficient energy conversion of Nd:glass laser radiation to its third harmonic ($3\omega = 351$ nm), and the subsequent demonstration of the process using two bare crystals of type-II KDP,² have provided the inertial-fusion program with an attractive approach towards improving the performance of laser-fusion targets. An overall energy-conversion efficiency of 80%, from 1ω ($\lambda = 1054$ nm) to 3ω ($\lambda = 351$ nm), was predicted and demonstrated for Nd:glass systems using real-beam temporal and spatial intensity profiles. The actual deliverable 3ω energy, however, has fallen below 80% because of problems with optical materials. Nonlinear loss processes at the second harmonic ($\lambda = 527$ nm)³ and photochemically induced degradation at the third harmonic⁴ have impeded the use of index-matching liquids for the recovery of energy losses from Fresnel reflections at the conversioncrystal surfaces.

An elegant conversion-cell design, which makes use of the polarization-mismatch tripling scheme, was developed by Summers *et al.* at Livermore.⁵ Two type-II KDP crystals are sandwiched in ''tandem'' in a single ''monolithic'' cell, separated from each other and from the cell windows by thin layers of index-matching fluid used to eliminate reflections from all internal surfaces.

We have adapted this design for use on our laser systems. A prototype cell has been successfully tested in our UV damage-testing facility,⁶⁷ and larger-aperture cells of the same design will be used for the conversion of the first six beams of OMEGA to 3ω in 1983. We have devoted considerable attention to long-term performance. We have tested a number of index-matching liquids and have selected Koolase,⁸ which we have found to perform well over several months of operation (over 400 shots); in particular, it exhibits good photochemical stability. Our design has also shown excellent alignment stability.

Monolithic-Cell Design

The polarization-mismatch concept for frequency conversion is shown schematically in Fig. 24. Both the KDP second-harmonic generator (SHG) and the KDP third-harmonic generator (THG) are type-II-cut, such that the z-crystallographic axis (the crystal optic axis) makes an angle of approximately 59° with the polished optical surface normal of each crystal. Laser radiation at 1ω , incident on the SHG, is linearly polarized at 35° to the o-direction of the doubler. Provided that the intensity of the incident laser radiation and the thickness of the SHG are appropriately matched,^{1,2} equal numbers of 1ω and 2ω photons emerge from the SHG, which is angle-tuned for phase matching. These photons are subsequently mixed in the THG to produce 3ω radiation. As indicated in Fig. 24, the THG is rotated with respect to the SHG by 90° about the system optical axis, and must therefore be angle-tuned in a plane orthogonal to that used to tune the SHG. Both crystals are of the same thickness in order to ensure optimum performance.⁹ The orthogonality of the SHG and THG crystals permits the design of a single frequency-conversion cell containing both crystals.



Fig. 24

Polarization-mismatch frequency conversion scheme. The incident 1ω radiation is polarized at 35° to the o-direction of the SHG to ensure that equal numbers of 1ω and 2ω photons emerge from the doubling stage. Both crystals are tuned about their o-directions to achieve phase-matching.

Fig. 25

Monolithic frequency-conversion cell. The SHG and THG crystals are held between a common pair of AR-coated, fused-silica end windows. ''Koolase'' index-matching liquid eliminates the six internal reflections of the cell, and all materials have been chosen for their chemical compatibility with this liquid. Liquid-layer thicknesses are maintained at 150 µm by glass spacers. Angle tuning for proper phase matching is then accomplished in a standard gimbal mount.

The design shown in Fig. 25 incorporates the SHG and THG crystals between a common pair of fused-silica windows. These four optical elements are gently, but firmly, held together by spring-loaded plungers and swivel pads. Three sets of glass spacers separate the crystals from each other and from the end windows, and enable indexmatching liquid to coat all six internal optical surfaces. The two external surfaces of the cell possess dual-wavelength, 1 ω and 3 ω , AR coatings. The characteristics of materials chosen for the construction and testing of a 60-mm-clear-aperture device are indicated by the code in



Fig. 25. The gimbal-mounted device is shown in Fig. 26. A thermistor temperature sensor, mounted within the cell body in close proximity to the crystals, controls current to an infrared heat lamp, which stabilizes the cell temperature to ± 0.05 °C. Temperature stabilization is required to decouple the cell from temperature fluctuations that occur in the laser bay.



Fig. 26

60-mm-clear-aperture monolithic cell in a gimbal mount. The infrared heat lamp, visible in the foreground, stabilizes the temperature of the cell to 22 °C. The cell is rotated about the vertical axis for doubler alignment and the horizontal axis for tripler alignment.

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One obvious advantage of the monolithic-cell design, when compared with separate SHG and THG designs, is that it requires fewer optical elements and AR-coated surfaces. In addition, we have chosen to make the input and output windows identical to simplify spare-parts inventory. It is also convenient to use a single liquid for index matching of all internal surfaces.

Assembly and Testing

It is fundamental to the monolithic-cell configuration that the SHG and THG crystals be installed with their ordinary (o) and extraordinary (e) directions orthogonal, and that the o- and e-directions be distinguished from each other (see Fig. 24). Satisfying the former condition ensures that angle tuning one crystal in the gimbal mount is decoupled from angle tuning the second crystal. Satisfying the latter condition ensures that the SHG may be properly oriented with respect to the incoming 1ω laser radiation.

By utilizing the uniaxial birefringence of KDP, it is possible to devise a simple, inexpensive method for orienting and marking SHG and THG crystals prior to loading and filling the monolithic cell. Figure 27 demonstrates the two procedures required. Rotation of a crystal through 360° around its optical surface normal, while between crossed polarizers, produces four intensity minima in the lock-indetected HeNe signal. These minima define the ordinary-ray and extraordinary-ray directions projected onto the polished crystal surface. These directions may be marked onto the crystal barrel. By orienting one of these directions at 45° to the incident HeNe polarization direction, the intensity between crossed polarizers can then be



Fig. 27

a) Marking of the o- and e-directions is accomplished by rotating the KDP crystal 360° about its center while situated in a HeNe laser beam between crossed polarizers. The desired directions correspond to the four intensity minima observed.

b) The o- and e-directions are distinguished from each other by tilting the crystal about an axis of rotation coinciding with a set of barrel marks. A rapidly varying series of intensity maxima and minima result if the crystal is being tilted about its o-direction. measured as a function of angle of incidence. If the axis of tilt and the o-direction coincide, a rapidly varying series of intensity maxima and minima, separated by about 1 mrad (depending on the crystal thickness), can be observed; this occurs because the angle between the propagation direction and the crystal optic axis is varied. (The actual values of the maxima and minima do not reach 100% and 0% in Fig. 27 because of the divergence of the HeNe laser beam.) If the axis of tilt and the e-direction coincide, very little intensity change over tens of mrad is observed. Both conditions are depicted in Fig. 27. This orientation technique can define the o- and e-directions of an SHG or THG crystal to within 60 seconds of arc, and is presently more sensitive than our ability to mark and mechanically orient circular crystals in a cell.

The 60-mm-clear-aperture monolithic conversion cell, shown in Fig. 26, has been operating in our UV thin-film, damage-testing facility^{6,7} since July 1982. Compared with the separate SHG/THG, two-cell method used prior to this date, the monolithic cell exhibits greatly improved long-term pointing stability and has generated a cleaner 3ω beam.

Index-Matching Liquid

Koolase is the refractive index-matching liquid chosen for our monolithic conversion cell. This proprietary azeotrope is composed of esters, heterocyclic, and primary hydroxyl compounds.⁸ It has a viscosity of 2 cst. at 21°C, a refractive index of 1.44 at λ = 337 nm,⁴

and negligible optical absorption at $\lambda = 527$ nm and $\lambda = 1054$ nm. The linear absorption coefficient for Koolase at $\lambda = 351$ nm can be as small as 0.37 cm⁻¹, but this may vary depending on the batch. This fluid was developed as a non-toxic, non-hygroscopic, laser-flash-tube coolant, and is therefore photochemically stable under intense UV irradiation. We have determined that Koolase is compatible with KDP and a number of other materials. Table 1 gives the results of a thirteenmonth passive soak test, conducted at room temperature. Particulates may be removed from the 2.0- μ m-filtered, as-purchased liquid, by additional filtering through a Nalge 0.20- μ m nylon-membrane

Table 1

Compatibility of various materials with filter using a glass syringe.

COMPATIBLE	NOT COMPATIBLE	WHY
• KDP	 Transparent mylar 	Swelling
 Fused silica 	 Dyed mylar 	Bleeding
• Indium	 Vinyl tubing 	Increased UV absorption
Gold	 Tygon tubing 	Increased UV absorption
• 6061 Aluminum	 Gum rubber tubing 	Increased UV absorption
 Black anodized aluminum 	 Polyflo tubing 	Increased UV absorption
 316 stainless steel 	 Silicon tubing 	Increased UV absorption
 Polypropylene 	 Buna N O-ring 	Increased UV absorption
 Nylon membrane filter 	 Neoprene O-ring 	Bleeding
• Viton	 Polyurethane O-ring 	Increased UV absorption
Red silicon O-ring	 Black delrin 	Increased UV absorption
White silicon O-ring	• Lexan	Increased UV absorption
White delrin	 Teflon TFE 	Increased UV absorption
Teflon PFA	Copper	Chemical reaction
Teflon FEP		
• RTV 108		

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In both the 60-mm-aperture monolithic cell and a separate 140-mm THG cell installed on the GDL facility, Koolase has shown no long-term photochemically induced degradation to itself or to the KDP crystal surface over the past three to six months. During this time period, 350 laser shots were taken at an average 3ω fluence of 0.2 J/cm², and 100 shots at 0.5 J/cm², all with a pulse width of 1 ns. The passive absorption losses for 1ω , 2ω , and 3ω laser radiation in the three 150-µm-thick fluid gaps of the monolithic cell have remained below 1% total during this time period.

Halocarbon oil,^{3,10} another index-matching fluid that has been found effective and stable in a 140-mm SHG cell at $\lambda = 527$ nm, has not exhibited adequate photochemical stability in a 140-mm THG cell. Figures 28a and 28b indicate that, after two months and 200 shots of 351-nm radiation at 0.5 J/cm², Halocarbon oil degraded within the cell and caused chemically induced etching of the polished KDP crystal surface.

Summary

We have described the design, construction, and performance of a monolithic cell for frequency conversion of moderate-intensity, Nd:glass laser radiation from $\lambda = 1054$ nm to $\lambda = 351$ nm. This design



Fig. 28

Incompatibility of Halocarbon oil with KDP in the presence of 3ω laser radiation. Photochemically induced degradation to Halocarbon oil after 2 months and 200 shots of 3ω radiation at 0.5 J/cm² resulted in chemical attack to a 140-mm THG-cell output surface. Optical scatter off the roughened and etched surface is evident in (a). A spectral scan comparing new and used oil [see (b)] shows that this liquid lacks chemical stability in the presence of 3ω laser radiation. takes advantage of the highly energy-efficient polarization-mismatch conversion scheme, wherein two type II KDP crystals of equal thickness are orthogonally oriented between a common pair of cell windows. Long-term pointing stability and 3ω beam quality are enhanced with the monolithic-cell design. Long-term photochemical stability problems, associated with the use of index-matching liquid, have been solved by the choice of Koolase.

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