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Frequency Doubling of Copper Lasers Using Temperature-Tuned ADP

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Lawrence
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Frequency Doubling of Copper Lasers Using Temperature-Tuned ADP

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1. Introduction

The ability to generate high average power UV at 255 nm by frequency doubling the green line (510.6 nm) of copper lasers would greatly extend the utility of copper lasers. Material processing and microlithography are two areas of interest. The frequency-doubled copper laser could replace the KrF excimer laser, which has a similar wavelength (248 nm), in some applications. The frequency-doubled copper laser has a narrow linewidth and excellent beam quality at a competitive cost. Other attractive features are high reliability, low operating costs, and the absence of toxic gases. This paper will report recent progress in high-efficiency, high-average-power harmonic generation of the copper laser green line using noncritical phase matching in ADP. Frequency doubling of the yellow line (578.2 nm) and sum-frequency mixing of the two lines are also of interest. These processes, however, cannot be phase-matched in ADP and, therefore, will not be discussed here. The results reported and the issues identified here would be important in these other processes and also in many other frequency conversion schemes in the UV such as 4ω conversion of Nd^{3+} :YAG lasers.

The first report of harmonic generation of copper laser radiation by Isaev, et al.¹, used ADP as the nonlinear medium. They found that 90° phase matching occurred over a broad temperature region near -30°C . A conversion efficiency of 8.9% was achieved. The average power was limited to 22 mW by thermally-induced dephasing due to absorption of the fundamental and/or the harmonic. Since then, several groups have reported harmonic generation of copper laser radiation using KDP² or BBO.³⁻⁶ Only the 578.2 nm line can be phase matched at room temperature in KDP. BBO has attracted interest because of its large nonlinear coefficient, good transparency in the UV, and high damage threshold. Nevertheless, because it must be angle tuned and has high birefringence, severe restrictions are imposed on the copper laser beam quality and alignment.

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In temperature-tuned, 90° phase matching, the dependence of the phase mismatch on the angular deviations in the input beam is of second order (noncritical). This greatly reduces the beam quality requirements on the copper laser. Recent analytical studies at LLNL⁷ have indicated that, with careful attention to thermal management, noncritically phase-matched ADP can frequency double 200 W copper laser radiation with an efficiency of at least 30%. A conversion efficiency of greater than 10% was predicted for a 20 W copper laser input. Realization of this performance requires very low absorption crystals.

In the experiments described here, a conversion efficiency of greater than 30% has been demonstrated at a drive corresponding to a 12 W copper laser. The efficiency is better than predicted, probably due to improved laser beam quality over that assumed in Ref. (7). This performance, however, could only be achieved by reducing the average power with a mechanical chopper. Results at higher average power were dominated by dephasing due to absorption-induced temperature gradients. Additionally, degradation of the crystals due to the UV exposure may be a serious practical limitation.

In Section 2, below, we describe the experimental apparatus. The minimization of thermal gradients is the primary goal of the experimental design. In Section 3, a simplified model of the doubling is described which includes the most salient features of the experiment. Section 4 summarizes the results obtained to date and compares those results with the model predictions. In Section 5, we describe the calorimetric method we have used to measure the bulk absorption of the crystals. Also discussed in that section are observations of extremely low damage thresholds and nonlinear absorption which were made during the course of the absorption measurements. The results are summarized in Section 6.

2. Frequency Doubling Experiment Design

The noncritical phase matching temperature of ADP at 511 nm is approximately -27°C . The ordinary index at the fundamental varies by about $5 \times 10^{-5}/^\circ\text{C}$. The temperature coefficient for the extraordinary index at the harmonic is only about 5% as large. The resulting temperature tuning bandwidth is only a few hundredths of a degree for crystals of about 10 cm length. Minimization of temperature variations is therefore the primary experimental design challenge.

2.1 Thermal Management

Temperature variations arise from variations in the crystal's environment or from temperature gradients within the crystal. The variations due to the environment are controlled by using a highly stable, temperature-controlled bath to regulate the crystal temperature. The temperature gradients, which are due to conduction of heat generated by optical absorption, present a more fundamental problem. The UV absorption edge of ADP is 184 nm.⁸ Although intrinsic absorption above the absorption edge is thought to be very small,⁹ impurities and imperfections can result in absorption of several percent per centimeter. Many of the common impurities increase the UV absorption most strongly although absorption at the fundamental wavelength could also occur.

Efforts to characterize and minimize absorption will be discussed later. First, we will discuss efforts to reduce the resulting temperature gradients. The gradients are minimized by making the path length along the lines of the heat flux as short as possible.¹⁰ One approach is to cascade a number of thin crystals and extract the heat along the beam propagation direction using a heat transfer fluid between the crystals. While this could achieve very short heat-conduction paths, the implementation presents a number of technological difficulties. Here, the heat-conduction path was minimized in a direction transverse to the beam propagation using a slab geometry suggested by M. Norton.⁷ The heat is removed across the narrow dimension of the slab. The narrow direction is chosen to be in the direction perpendicular to the optic axis to minimize dephasing due to diffraction. The geometry is shown in Figure 1.

Figure 1. Schematic of geometry used to minimize temperature gradients in noncritical phase matching experiments. The heat is extracted along the shortest dimension of the crystal. For a uniform heat input, this results in a parabolic temperature distribution transverse to the input beam propagation.

Based on the preliminary analysis by Norton, a crystal cross section of 5 x 10 mm was chosen. The optic axis was parallel to the longer dimension. Lengths varied from 75 mm to 100 mm. The large surface area faces were cooled by contact with a copper heat sink which was in turn cooled by a flowing heat-transfer fluid. The heat sink and the crystal were enclosed in a dry nitrogen filled chamber to prevent condensation. The holder was mounted in a 6" Aerotech mirror mount to provide for angular adjustments. The crystal holder assembly is shown in Figure 2.

Modeling results showed that reduction of the contact resistance at the crystal/copper interface is necessary for optimal performance. Several methods of reducing contact resistance were tried. Best results were obtained with a composite heat transfer material (Q-PAD II by Bergquist) consisting of a .001" aluminum foil coated on both sides with .0025" of thermally conductive Sil-Pad rubber. Q-PAD is not very pliable, limiting the gaps that it can fill. Therefore, the crystal surfaces were polished and the copper block was lapped to further improve the contact. Thermally conductive grease was also tried but had several problems. After several hours of operation, the grease dries out, perhaps due to an interaction with the crystal. Also, the crystal is very difficult to remove without damage, and care must be used to avoid getting grease on the crystal face. Another option is a silver-copper filled silicone elastomer (Consil-C by Tecknit). This material is thicker than Q-PAD and thus has a higher thermal resistance. Nevertheless, since it is much more pliable, it could reduce the contact resistance by filling in the gaps. Results with this material so far have been disappointing, but since the contact resistance cannot be measured directly, other causes cannot be ruled out.

Figure 2. Photograph of the crystal mounting assembly.

The long, thin sides of the crystal were not in contact with the copper heat sink. Heat transfer at these edges was by conduction and convection into the dry nitrogen atmosphere. The heat flow across these surfaces was expected to be small compared with that in the narrow direction. In some experiments, however, variations in the UV output perpendicular to these edges suggested these surfaces may not be adiabatic. In the future, the boundary conditions on these edges will be better controlled.

The heat sink temperature was regulated using a NESLAB low temperature bath. The heat transfer fluid (Dow SylTherm XLT) was chosen for its low viscosity and low density at the working temperature. The four gallon reservoir was connected to the crystal holder assembly through about 2.5' of .5" Teflon lines. A platinum RTD sensor approximately 10 cm upstream from the heat sink provided the temperature control signal. The RTD output is recorded on a chart recorder. With care, the chiller was able to regulate to $.02^{\circ}\text{C}$ for sustained periods. However, the chart recorder output occasionally drifted and exhibited increased fluctuations, especially when the copper laser amplifier was turned on. Whether this is due to temperature fluctuations or only noise pickup in the chart recorder is not known. The crystal mount temperature may differ from the setpoint by a few hundredths of a degree due to heat loss from the lines.

2.2 ADP Crystals

Four ADP crystals have been used. Two were grown in China. Two were grown by Cleveland Crystals in the United States. As stated above, the cross sections of the crystals are 5×10 mm. The American crystals are 100 mm long. The Chinese crystals are 75 mm long. We have recently received crystals from Quantum Technologies which have not yet been used.

The vendors were asked to supply crystals with absorption at 255 nm as low as possible. Cleveland Crystals supplied data on transmission at the fourth harmonic of the Nd:YAG laser (266 nm) as a function of pulse energy for the crystals that they supplied. Nonlinear absorption was evident in the data.⁹ However, extrapolation of the data to the small pulse energies used here suggests negligible nonlinear absorption. The linear absorption coefficient was $.01 \text{ cm}^{-1}$. No data was supplied with the Chinese crystals.

We made transmission measurements at 255 nm using a copper laser doubled in a BBO crystal. Fluctuations in the UV amplitude limited the accuracy of this measurement. Nevertheless, the differences between the crystals from the two manufacturers were measurable. The measured absorption of the Chinese crystal was $.078 \text{ cm}^{-1}$ with an uncertainty of $\sim .013 \text{ cm}^{-1}$. The measured absorption of $9 \times 10^{-4} \text{ cm}^{-1}$ in the American crystal was below the minimum measurable absorption ($\sim .01 \text{ cm}^{-1}$). This measurement cannot distinguish scattering and surface losses from bulk absorption. The absorption coefficients given here were obtained assuming no scattering and only Fresnel losses at the surfaces. These coefficients, therefore, are of limited use in predicting the performance of the crystals. To address these uncertainties, calorimetric measurements, discussed in more detail below, have been used to measure bulk absorption.

Absorption at the fundamental wavelength could also be a problem although most of the suspected contaminants absorb more strongly at shorter wavelengths. Empirical evidence from the doubling experiments also suggests that absorption of the fundamental is not significant at the power levels used up until now. When the crystal is slightly colder than the 90° phase matching temperature, a change of angle can phase match the interaction. When this is done, the UV is initially bright and uniform. Only after a few seconds do thermal effects appear causing a hole in the middle of the beam or, at higher powers, nearly complete extinction of the UV. If the fundamental absorption were significant, the temperature-induced dephasing would occur as soon as the UV was generated by angle tuning. The calorimetric absorption measurements, discussed below, later confirmed absorption in the green of less than $0.1\%/ \text{cm}$.

A visual examination of the crystals between crossed polarizers was also made. The isophote pattern in the Chinese crystals appeared noticeably sharper than in the American crystals. This qualitative comparison may indicate that the Chinese crystal has better homogeneity or lower strain resulting in less wander of the optic axis through the crystal.

2.3 Optical Setup

The idealized slab geometry is approximated by using an elliptical beam profile (Figure 3). The circular laser output is made elliptical by an anamorphic beam-reduction telescope. L1 is a spherical lens with a 1m focal length. L2 and L3 are both plano-concave cylinder lenses. L2 has a focal length of -99 mm for rays in the plane of the figure. L3 collimates the rays out of the plane of the paper. Several values of the focal length of L3 were used.

Figure 3. Optical layout of ADP experiment.

The input copper laser beam is nominally circular with a diameter of 8 cm. There is a shadow at the bottom of the beam due to the molten copper at the bottom of the tube. There is a 5 mm diameter round shadow in the center of the beam due to the hole in the scraper mirror of the unstable resonator. The laser is polarized out of the plane of the figure.

The laser oscillator for these experiments had a 40 mm bore plasma tube. The resonator was an injection-seeded, unstable resonator. The laser differs only slightly from those routinely used in the AVLIS program.¹¹ The magnification of the master oscillator is increased to -50 in order to improve the beam quality and beam paths in the oscillator are enclosed to reduce the effects of air turbulence. This laser has a total output of approximately 30 W of which about 18 W is in the 510.6 line. The repetition rate of the laser is 4350 sec^{-1} . The pulse width is about 50 nsec. The beam divergence is 2-3 times diffraction limited based on the size of the far-field spot.

The output of the oscillator was magnified by 2 times and then passed through an amplifier with a bore of 80 mm. For most of the experiments, the amplifier discharge was off. Clipping of the oscillator beam at the amplifier tube walls and losses in the telescope and windows reduced the available green power, with the amplifier off, to 11 W or less. With the amplifier on, the available power was about 100 W, but experiments reported here only used a maximum of 60 W. An electronic shutter (UniBlitz VS14, Vincent Assoc.) in front of the crystal reduced the average power in many of the experiments. Since the peak power in the copper laser pulse is unchanged, this can be used to investigate the expected improvement from reduced material absorption.

A right angle, fused silica prism separated the UV and fundamental beams emerging from the crystal. Powers were measured with a Gentec power meter. The throughput of the prism and the exit window of the crystal holder were measured at 255 nm using the beam generated in the BBO crystal. The powers reported here are corrected for these losses as well as for the Fresnel loss at the crystal faces. Thus, the quoted conversion efficiency is always the internal efficiency. The fundamental powers were measured with the shutter open. The actual average power must be multiplied by the shutter duty factor.

A Gentec holographic beam splitter sampled the output beam for additional diagnostics. The third order UV was used to image the crystal output onto a CCD camera. The negative third order was used to obtain a temporal pulse profile. The holographic beam splitter was AR coated for 532 nm and had significant loss for the UV. This loss was taken into account in power measurements.

3. Model

To better understand the behavior, and to provide guidance for optimizing the performance, a model has been developed to predict the effect of thermally-induced dephasing. The model assumes an ideal slab geometry. No variation of the temperature or the beam profile along the direction of the optic axis is included. This model gives insight into important aspects of the problem although quantitative agreement with experiment is not satisfactory. Some of the assumptions will be relaxed in a more complete model currently under development. Before discussing the results and their comparison with the experiments, an outline of the model and its assumptions will be discussed.

3.1 Basic Assumptions

The model treats the wave propagation using the coupled wave equations.¹³ The assumptions involved in reducing the Maxwell wave equations to the coupled wave equations are discussed in Ref. (13). One central assumption is that the fundamental and harmonic are described by uniform plane waves with slowly varying amplitude and phase. In order to allow for temperature-induced amplitude and phase variations in the transverse direction, the model used here divides the crystal into subapertures over which the uniform plane wave assumption is presumed valid. In the slab geometry, these subapertures are thin slices. This approach is plausible when the Fresnel number for an individual slice is large compared to unity. Beam distortion as a result of temperature-induced refractive index gradients is also ignored in this approach. With this approximation, the transverse coordinate, x , becomes a slowly varying parameter. The equations which we integrate then are:

$$\begin{aligned}\frac{\partial A_1(x,z)}{\partial z} &= -\frac{1}{2}\alpha_1 A_1 - i\kappa A_1 A_2^* e^{-i\int_0^z \Delta k dz} \\ \frac{\partial A_2(x,z)}{\partial z} &= -\frac{1}{2}\alpha_2 A_2 + i\kappa A_1^2 e^{+i\int_0^z \Delta k dz}\end{aligned}\tag{1}$$

In the above equations, the subscript 1 refers to the fundamental and 2 refers to the harmonic. The A 's are proportional to the electric fields and the coupling constant is proportional to the effective nonlinear coefficient d_{eff} . The transverse coordinate, x , appearing in these equations is treated only as a parameter. The phase-mismatch term is a result of the difference between the local temperature and the exact phase-matching temperature. That is,

$$\Delta\kappa = \frac{2\pi}{\lambda} (T(x,z) - T_0) \frac{dn}{dT}\tag{2}$$

Here $T(x,z)$ is the local temperature, T_0 is the phase-matching temperature and dn/dT is the variation with temperature of the difference in refractive indices of the fundamental and harmonic.

The first terms on the right of equations (1) represent absorption of the incoming and generated beams. Only linear absorption has been considered so far. Because of the long pulse length and low peak power, two-photon absorption¹¹ is negligible. However,

long-lived defects formed by exposure to UV have been observed to cause linear absorption over a broad spectral range covering both the fundamental and the harmonic.¹⁹ Under some conditions, this defect formation is observed to be linear in the UV power. This process could lead to a parametric dependence of the absorption of both the fundamental and harmonic on the UV intensity. This has not yet been included.

In the simple model described here, the heat flow is one-dimensional - normal to the large area faces of the crystal as shown in Figure 1. Two-dimensional heat flow will be included in the model in the future. The model does not assume uniform temperature in the propagation direction. A temperature gradient along the propagation direction results from the variation of absorbed power with z , but heat flow due to this temperature variation is neglected. Contact resistance at the boundary is included. The temperature distribution generates the phase mismatch in the coupled-wave equations via equation (2). This mismatch is assumed to result from the temperature variation of the refractive index neglecting thermal-stress-induced birefringence.

3.2 Input Parameters

The material parameters needed for the model generally have not been measured at -30°C . Some of the parameters characterizing experimental conditions also are difficult to obtain. In this section, the important parameters will be discussed and the best estimates of the values will be given.

The refractive indices determined from the Sellmeier equation with temperature-dependent indices, as given in Ghosh and Bhar²³, gives excellent agreement with the measured noncritical phase-matching temperature for the copper laser line and also the 514 nm Ar laser line. Therefore, the value of dn/dT obtained from this reference should be sufficiently accurate.

There are many of measurements of the nonlinear constant d_{36} .^{15,16} Many of these are at 1.06 μm , although there are a number of measurements in the visible also. Unfortunately, the available measurements differ widely. In some cases, this may be due to different measurement methods. In other cases, there may be variations simply due to the definition of the nonlinear coefficient. In the results quoted here, a value of 0.56 pm/V has been used. This is at the high end of reported values but gives better agreement with the low average power results found in this work. All the reported measurements are at room temperature, though it is not likely that the nonlinear coefficient changes significantly with temperature.

Thermal parameters have not been extensively studied. There is only one reported measurement of thermal conductivity and it is at 315 K.¹⁷ For this work, I have used the value found in that reference for conductivity parallel to the optic axis. The absorption, especially at the harmonic wavelength, varies greatly from sample to sample. The measured values discussed above therefore were used with some adjustments to improve agreement with observations. There is no available information on the variation of absorption with temperature. As already mentioned, the absorption determined from transmission measurements may not be due entirely to bulk absorption.

The contact resistance is difficult to estimate. Parameters, such as surface roughness, are not known well enough in this experiment to make use of available theories. In some cases, departure from flatness, either by the crystal or the copper heat sink, are believed to have produced gaps leading to increased contact resistance. The approach taken here was to estimate a reasonable range for contact resistance and then vary the value within that range to obtain best results.

As will be seen below, this model is not adequate at present to quantitatively predict experimental results. This is due both to the simplicity of the model and to incomplete knowledge of the required input parameters.

4. Results

4.1 Oscillator Power

Most of the experiments reported here used only the oscillator power. When only the oscillator was used, the laser power available at the crystal input was about 12 W. The limits on high-average-power doubling due to thermal dephasing were evident even at this power. The effect of lowered thermal dephasing was investigated by varying the duty cycle of the UniBlitz shutter. Best conversion efficiencies are achieved when the UniBlitz shutter duty cycle is small (<25%). Since the duty cycle affects the power absorbed in the crystal but not the peak power of the input pulse, the effect is equivalent to reducing the absorption of the crystal (the repetition rate of the shutter is faster than the thermal time constants of the crystal).

Some difficulties with repeatability of the results were encountered. This seemed to be largely related to variations in contact resistance even when care was taken to mount the crystal identically. Changes in the crystals over the duration of these experiments may also play a role. The condition of the surfaces may have varied from one run to the next. All the crystals were repolished at least once, and this could have induced additional strains or microcracking in the crystals. The results also depended on the stability of the temperature controller which varied somewhat from day to day perhaps due to noise on the power line.

Another potential variation is the laser beam quality and other aspects of the optical alignment. Despite these variations, the data is sufficient to indicate the range of performance which is possible with this system.

The thermal gradients were manifested in several features of the SHG results. Two aspects will be discussed here. The first is the broadening of the temperature tuning curve with an increase in average power. The second is the change with average power of the UV intensity pattern emerging from the crystal.

The temperature tuning curve is the curve of the UV output power as a function of the chiller setpoint temperature. This curve is a sensitive indicator of thermal effects in the crystal. At very low average input power, the output power is not influenced by temperature gradients. The width of the curve of output power as a function of the setpoint temperature of the constant-temperature bath is determined only by the temperature variation of the refractive indices. The only material parameter which affects the peak value of this low-average-power, temperature-tuning curve is the effective nonlinear coefficient. At higher powers, the temperature-tuning curve broadens as different portions of the crystal become phase matched at different temperatures. The peak value increases more slowly than the average power resulting in decreased conversion efficiency. The peak also moves towards lower temperatures as the boundary value is lowered below the phase matching temperature to compensate for the increase in the interior temperature.

Figure 4 shows a temperature-tuning curve of one of the American crystals when the shutter duty factor was 0.083. Also shown in Figure 4 are the predictions of the model for this experiment. The good agreement in width between model and experiment indicates that the refractive indices used in the model are accurate. The peak height predicted by the model also is fairly close, indicating that the value of the nonlinear coefficient also is pretty good. Note that even at this low duty factor, the experimental temperature-tuning curve has marked asymmetry.

Figure 4. Temperature-tuning curve at very low average power for one of the American ADP crystals.

The best conversion efficiency obtained with this low chopper duty factor was 34%. This was obtained at an input power of 5.7 W using the Chinese-grown crystal. The UV power was 161 mW. When the chopper duty cycle was increased to 0.25, the conversion efficiency fell to 19%. A conversion efficiency of 26% was obtained with the crystal from Cleveland Crystals also with a duty factor of 0.083. In this case the input was 11.9 W and the UV output was 260 mW. More typically the conversion efficiency was 20-25% at low duty factors. Various experimental difficulties prevented a complete exploration of the parameter space and the repetition of many of these results. Nevertheless, there seems to be enough data to indicate the range of conversion efficiency which is achievable with this material in this geometry in the absence of strong thermal dephasing.

As the average power of the input is increased, thermal effects due to absorption begin to dominate the process. The conversion efficiency drops, the temperature tuning curve broadens, and the bath setpoint for optimum output gets lower. A temperature tuning curve at full oscillator power is shown in Figure 5. Other conditions were the same as those in Figure 4. The peak conversion efficiency for the data in Figure 5 is 7%.

Figure 5 shows that the degree to which the model underestimates the effect of thermal dephasing is much greater at higher power. Some improvement could be achieved by assuming much larger (probably too large) contact resistance. Preliminary results from a model that treats the two-dimensional heat-conduction effects indicates that the elliptical beam profile produces a broader tuning curve than that predicted by the one-dimensional mode. This doesn't seem large enough to account for the discrepancy either. Observation of the output UV near-field beam profile suggests that nonuniformities in heat-loss or absorption mechanisms also are important. All these potential causes of the discrepancy between the model and the experiment are currently being investigated.

The best conversion efficiency which has been obtained with full oscillator power is 9.0%. This was obtained with the Chinese-grown crystal using Q-PAD for heat transfer. The input was 10.7 W and the output was 0.96 W.

Figure 5. Temperature tuning curve for full oscillator power (UniBlitz shutter removed).

The near-field intensity distribution of the UV also showed strong thermal effects. At some temperature settings, the output was concentrated in only about one-third of the output aperture. At other temperature settings the output was more uniform, or concentrated, in other regions of the output aperture. The nonrepeatability discussed above was most apparent in these intensity distribution observations. Under what we believe were the best conditions, fringes appeared in the near-field output. A typical pattern is shown in Figure 6. As the temperature setpoint was varied, the fringes moved vertically across the face of the crystal. Variations of UV output across the output face are also predicted in the model, although such high-contrast fringes have not been obtained theoretically. The origin of these fringes seems to be a non-monotonic dependence of the UV power with distance along the

crystal. The thermal dephasing is large enough to result in back-conversion of UV to the fundamental. The distance over which this occurs varies across the aperture.

Figure 6. Intensity distribution of UV at exit of crystal. Several qualitatively different types of patterns were observed depending, apparently, on the thermal contact conditions at the edge of the crystal. The fringes shown here occurred under the conditions which gave the best conversion efficiency.

At the higher average powers, the thermal dephasing also resulted in instabilities. This was most noticeable as the temperature setpoint was reduced to compensate for the temperature rise due to absorption. In Figure 4, the data was taken by gradually lowering the setpoint. As the setpoint was reduced from -27.8, the UV output initially rose. Over a period of a few minutes, the near-field output exhibited a lot of fluctuation. Then, suddenly, the center of the beam went dark, followed quickly by the power across the whole aperture dropping. This instability is not taken into account in the model, which is a steady-state theory, but may be partly responsible for the inadequacies of the model.

4.2 Amplifier Power Levels

Additional difficulties were encountered at very high input powers. When amplifier power was used, measurable UV was not obtained unless very low shutter duty factors were used. Since the minimum "on" time for the shutter was limited to 5 msec, the low duty cycle necessitated a low shutter repetition rate. At this low repetition rate, the power meter was difficult to read accurately. Several times the damage threshold of the shutter was exceeded. The input power was too high to be measured directly with the Gentec power meter. The power was measured by inserting a calibrated attenuator in the beam in front of the 1 m spherical achromat.

Despite these problems, a few measurements were made. With a duty factor of 0.014 and an input power of 63 W, an output of 0.196 W was obtained with the Chinese-grown crystal. The conversion efficiency in this case was 34%. No measurements with the crystal from Cleveland Crystals have yet been made.

5. Calorimetric Absorption Measurements

In an effort to more fully understand these results, calorimetric measurements of the absorption of the ADP crystals have been made. In the course of making these measurements, we have also found evidence that damage from long-term exposure to UV, even at very low fluences, may present another fundamental limitation to the use of ADP for high-average-power, deep-UV harmonic generation.

The method used for this measurement is similar to that described by Pinnow and Rich²⁰. A schematic diagram of the apparatus is shown in Figure 7. One crystal is used as a reference to compensate for environmental temperature changes. The UV for the measurement is generated in a critically phase-matched BBO crystal. The crystal is illuminated with low power UV and peak temperature rise in the crystal is measured. The input beam is then shut off and the cooling rate of the crystal is measured. Since the heating rate at the peak temperature must equal the cooling rate, the heating rate and thus the absorption coefficient can be derived.

Figure 7. Apparatus used for calorimetric absorption measurement.

Figure 8 shows the crystal temperature and the transmitted UV power during a typical measurement. Note the decrease in transmitted power over the course of the measurement. In some cases, the UV power generated in the BBO decreased 20-25% during the course of the data run due to thermal effects in the BBO. However, the decrease in transmitted output power was invariably greater than the decrease at the input. This occurred with all samples and at all powers used. The origin of this decrease is not certain, but it may be due to surface damage. For powers greater than 40 mW, surface discoloration was observed. Microscopic examination of the surfaces was not done and, in many cases, the apparatus was not even disassembled between data runs to make a close visual inspection. Therefore, no definite statement can be made regarding damage below 40 mW. The UV beam area was approximately 0.34 mm^2 . Therefore, the peak power was only about 190 kW/cm^2 (9.5 mJ/cm^2 pulse energy) even at the highest input power used (140 mW). This is much lower than any damage thresholds I am aware of in the literature.^{14,22} The damage is perhaps due to an accumulated effect of many pulses. The crystal was typically exposed to the UV for an hour (1.6×10^7 pulses). It should also be pointed out that similar damage was observed in a sample of KDP tested in this same apparatus at comparable power levels. If we assume a damage threshold of 12 W/cm^2 (40 mW over $.34 \text{ mm}^2$) average power, we find that even in the absence of temperature gradients, the maximum output from crystals of this geometry is about 6 W.

Figure 8. Typical results of absorption measurement showing the rise in crystal temperature and the decrease of UV power transmitted through the crystal. During this run the UV input power decreased by about 30% - not enough to account for the observed drop in output.

Yet another potential explanation for the reduction in transmitted power over the course of the experiment is the formation of defects in the bulk of the crystal.¹⁹ These defects may be due to nonlinear absorption and thus would form very slowly at the low peak powers used here. However, they may also have very long lifetimes allowing them to accumulate over the course of the experiment. Except at the lowest input powers, a track of light blue fluorescence along the path of the UV was evident, indicating some excitation of the crystal. The fluorescence was not uniform along the length of the crystal but was much stronger in localized areas (especially in the Chinese-grown crystal), indicating that the excitation may be associated with impurities or dislocations in the crystal. At the highest powers used, the blue fluorescence appeared not just along the path of the UV beam but throughout the crystal. It was not steady but fluctuated. This fluctuating fluorescence may be the result of UV refracted at the surfaces which were beginning to damage. However, without further testing, other explanations cannot be ruled out. If formation of long-lived defects by UV plays a role in the absorption measurements, it is likely to be very temperature dependent and thus potentially much more important in the frequency doubling experiment at -27°C.

The effects discussed above called into question the validity of bulk absorption coefficients determined by this method for all but the lowest input powers. At very low powers, the accuracy of the measurements suffered due to the difficulty of measuring low temperatures and low UV powers. Nevertheless, enough data was obtained to conclude that the bulk absorption coefficient for the Cleveland Crystals samples is about 1-2%/cm and that for the Chinese-grown crystals is about 3-4%/cm. These results are fairly consistent with the transmission measurements mentioned earlier. The bulk absorption for the fundamental was also measured for one of the American-grown crystals. At this wavelength there were no unusual problems and no damage despite the input power being 560 mW. The bulk absorption for the fundamental was found to be about 0.05%/cm.

6. Discussion and Future Work

Careful attention to thermal design has resulted in output powers about 50 times that achieved in the early work of Isaev.¹ The conversion efficiencies predicted by M. Norton⁷ have been exceeded in this experiment at very low average power. The higher conversion efficiencies are likely due to beam quality which is much better than the 20 x DL assumed there. At higher average power, however, the conversion efficiency is severely limited by thermal gradients introduced by UV absorption. The output power has not exceeded 1.0 W. Performance below that predicted by Norton is not surprising since the absorption coefficient is at least an order of magnitude larger than assumed in that memo and since the experiments showed evidence that nonuniformities and thermal contact resistance were limiting the output power.

If ADP is to be a useful material for high-average-power frequency doubling of copper lasers, the results presented here show that very substantial improvements will be needed. The primary obstacles to increased UV output are the large UV absorption coefficient, the difficulty of removing the heat, and the low damage threshold found in repetitively pulsed measurements.

The output power limitation due to thermal gradients will scale with the absorption coefficient for a fixed geometry. In order to obtain an output on the order of 100 W by a reduction of the absorption coefficient alone, a reduction of two orders of magnitude would be required. This is unlikely. An aggressive materials development effort could perhaps have a reasonable goal of reduction of the UV absorption coefficient by one order of magnitude. Some efforts in this direction are already underway. ICP mass spectroscopy measurements on the Chinese and American crystals used here are being done to try to identify particular impurities which may cause UV absorption. Samples of material from other vendors is also being obtained. One particularly interesting source is material grown

at Moscow State University by a "fast-growth" method.¹⁸ High purity raw materials are essential in this method due to the high levels of supersaturation employed. These crystals are likely, therefore, to have very low impurity absorption although other important parameters such as stress birefringence are potential problems. An ADP crystal grown by Zaitseva has been obtained and will soon be cut and characterized.

Since reduction of the absorption alone doesn't seem likely to be able to sufficiently improve the performance, improvements in the heat removal efficiency must also be made. The thermal contact resistance could be reduced by direct contact between the crystal and the cooling fluid. Very stable boundary conditions could be assured by using boiling heat transfer. Material compatibility between the crystal and the cooling fluid is a big potential problem with this approach. A shorter heat-conduction path could be achieved by reducing the 5 mm height of the crystal. Because of the difficulty of polishing thin crystals, 2 mm is the smallest this could be made. Issues of increased beam divergence, increased fluence, and difficulty of alignment also arise in this approach. The heat could be extracted more efficiently using thin crystals with longitudinal heat flow, but the implementation of this approach presents many problems.

The extremely low surface-damage thresholds observed need to be more fully understood before ADP can be seriously considered for reliable SHG. The damage has not been observed in SHG experiments to date. However, with even modest improvement in the absorption coefficient and/or the heat transfer, the damage threshold would become a serious limitation. A smaller crystal cross-section would further aggravate the effect. The damage threshold has not been systematically studied in this work. It was only noted as occurring during the course of the absorption measurements. The accurate determination of the threshold power levels, the exposure duration dependence, and the temperature dependence are needed to evaluate its effect on the frequency doubling performance.

Optimization of the experimental parameters will be crucial to obtaining adequate performance. Initial modeling results have suggested that shorter crystals may work better. The model is currently being upgraded to provide more reliable guidance in this optimization. The greatest modification is the inclusion of two-dimensional heat conduction. Some improvement in agreement with the experimental data has been observed as a result of accounting for the elliptical beam shape. Initial attempts to reproduce the nonuniform output beams by introducing local variations in the thermal contact resistance have not yet been successful. The effects of non-uniform input beams will also be explored. Heat conduction along the direction of light propagation could be included in the model if needed. If experiments continue to suggest that some of the results

are really unstable operating points, time dependent modeling may be needed to fully explain the experimental results.

While the problems encountered with ADP do not seem to be insurmountable (with the possible exception of the damage threshold problem?), their solution would require considerable effort. A renewed look at other nonlinear crystals is advisable. We have recently demonstrated noncritical phase matching of copper laser radiation in KDP at about -67°C . Because the temperature variation of the refractive index difference in KDP is an order of magnitude smaller than that of ADP, its temperature tuning bandwidth is larger. In addition, considerable work has been done on reduction of KDP absorption at the fourth harmonic of the Nd:YAG laser (266 nm). The damage threshold for repeated exposure to UV may be a difficulty with KDP. The Neslab chiller is currently being upgraded to operate at this temperature and KDP crystals with the same geometry as the ADP crystals discussed here are being obtained. Critical phase matching in BBO is the other viable SHG method. Continual improvements in copper laser beam quality and the use of cylindrical focusing and alternating $-z$ geometries partially alleviate the disadvantages of critical phase matching. At the lower powers used in BBO experiments to date, the drive cannot be made large enough to obtain high conversion efficiency without introducing significant angular dephasing. Experiments at higher input powers are planned. At higher powers, the thermal considerations investigated here will become important in critical phase matched SHG also.

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8. References

- (1) A. A. Isaev, G. Yu. Lemmerman, and G. L. Malafeeva, "Second Harmonic Generation from Pulsed Copper Vapor Laser Radiation," *Sov.J.Quant.Elec.* **10**, 983-985 (1981)
- (2) Yu. P. Polunin and V. O. Troitskii, "Nonlinear Frequency Conversion of Copper Vapor Laser Radiation," *Sov.J.Quant.Elec.* **17**, 1433-1434 (1987)
- (3) D. W. Coutts, M. D. Ainsworth, J. A. Piper, "Enhanced Efficiency of UV Second Harmonic Generation from Copper Vapor Lasers," *IEEE JQE* **26**, 1555-1558 (1990)
- (4) K. Kuroda, T. Omatsu, T. Shimura, M. Chihara, and I. Ogura, "Second Harmonic Generation of a Copper Vapor Laser in Barium Borate," *Opt.Comm.* **75**, 42-46 (1990)
- (5) T.G.M. Freegarde and G. A. Naylor, "Efficient High Average Power Harmonic Generation Using Copper Vapor Lasers," *CLEO paper CTHK3* (1990)
- (6) W. Molander, M. Norton, and J. Chang, "Improved Efficiency of Second-Harmonic Generation in BBO Using the 511-nm Line of an Injection-Seeded Copper Laser," *CLEO paper CTuW11* (1991)
- (7) M. Norton, "The Potential for 300-500 Watts of 255 nm Light via Second Harmonic Generation of the Copper Lasers," LLNL interdepartmental memo dated May 10, 1990; M. Norton, "Second Harmonic Conversion with ADP at 510.6 nm," LLNL interdepartmental memo dated June 14, 1990
- (8) W. L. Smith, "KDP and ADP Transmission in the Vacuum Ultraviolet," *Appl.Opt.* **16**, 1798 (1977)
- (9) The actual level of intrinsic absorption, however, is not actually known and may not be insignificant. Dr. R. S. Adhav of Quantum Technologies believes it is in the area of 0.5%/cm (private conversation).
- (10) D. Eimerl, "High Average Power Harmonic Generation," *IEEE JQE* **23**, 575-592 (1987)

- (11) J. Reintjes and R. C. Eckardt, "Two-Photon Absorption an (sic) ADP and KD*P at 266.1 nm," IEEE JQE **9**, 791-795 (1977)
- (12) B. Warner, "An Overview of Copper Laser Development for Isotope Separation," SPIE Proceedings **737**, 2-6 (1987)
- (13) A. Yariv, Quantum Electronics 3rd Ed, Wiley (1989) p.399
- (14) V. G. Dmitriev, Handbook of Nonlinear Optical Crystals, (Springer, 1991)
- (15) S. Singh "Non-Linear Optical Materials," in CRC Handbook of Lasers, R. J. Pressley, ed. pp.489-525 (CRC Press, 1971)
- (16) D. Eimerl, "Electro-Optic, Linear, and Nonlinear Optical Properties of KDP and its Isomorphs," Ferroelectrics **72**, 95-139 (1987)
- (17) K. A. McCarthy and S. S. Ballard, "New Data on the Thermal Conductivity of Optical Crystals," JOSA **41**, 1062-1063 (1951)
- (18) N. Zaitseva, "Fast Growth of KDP and DKDP Crystals from Solution," paper printed and distributed by Metastat Advance Information Services, Eugene, Oregon (1992?)
- (19) H. Lee, J. E. Davis, and R. Hughes, "Recent Results on Optically Generated Defects in KDP and KD*P".
- (20) D. A. Pinnow and T. C. Rich, "Development of a Calorimetric Method for Making Precision Optical Absorption Measurements," Appl.Opt. **12**, 984-992 (1973)
- (21) K. Kato, "Conversion of High Power Nd:YAG Laser Radiation to the UV at 2661 Å," Opt.Comm. **13**, 361-362 (1975)
- (22) S. C. Matthews and J. S. Sorce, "Fourth Harmonic Conversion of 1.06μ in BBO and KD*P," SPIE Proceedings, Vol. 1220, Nonlinear Optics, 137-147 (1990)
- (23) G. C. Ghosh and G. C. Bhar, "Temperature Dispersion in ADP, KDP, and KD*P for Nonlinear Devices," IEEE JQE **18**, 143-145 (1982)

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