Technical Report

RAL-TR-98-013

Efficient X-UV Harmonic Generation from Solid targets by the TITANIA KrF laser

TMR Large-Scale Facilities Access Programme

S Moustaizis et al

January 1998

COUNCIL FOR THE CENTRAL LABORATORY OF THE RESEARCH COUNCILS

Efficient X-UV Harmonic Generation from Solid targets by the TITANIA KrF laser

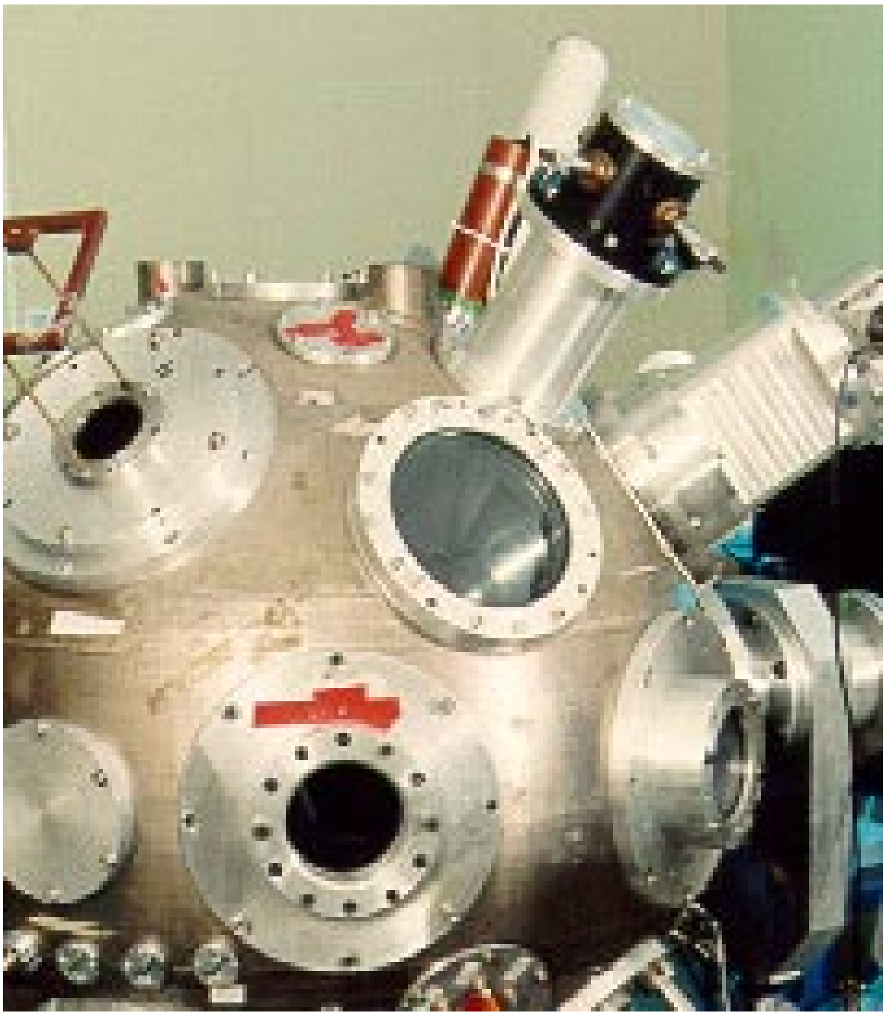
S Moustaizis et al

IESL - Forth, Heraklion, Greece

An experiment performed with funding from the

TMR Large-Scale Facilities Access Programme

Contract No. ERBFMGECT950053



Efficient X-UV Harmonic Generation from Solid targets by the TITANIA KrF laser

An experiment performed with funding from the
TMR Large-Scale Facilities Access Programme

Access to Lasers at the Central Laser Facility
Rutherford Appleton Laboratory
Contract No. ERBFMGECT950053

S Moustaizis
IESL - Forth, Heraklion, Greece

PA Norreys and D Neely
CLF, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon OX11 0QX, UK

DM Chambers, SG Preston and JS Wark,
Clarendon Laboratory, Oxford University, Oxford, UK

M Zepf, I Watts and AE Dangor
Imperial College, Prince Consort Road, London, UK

RS Marjoribanks
University of Toronto, Ontario, Canada

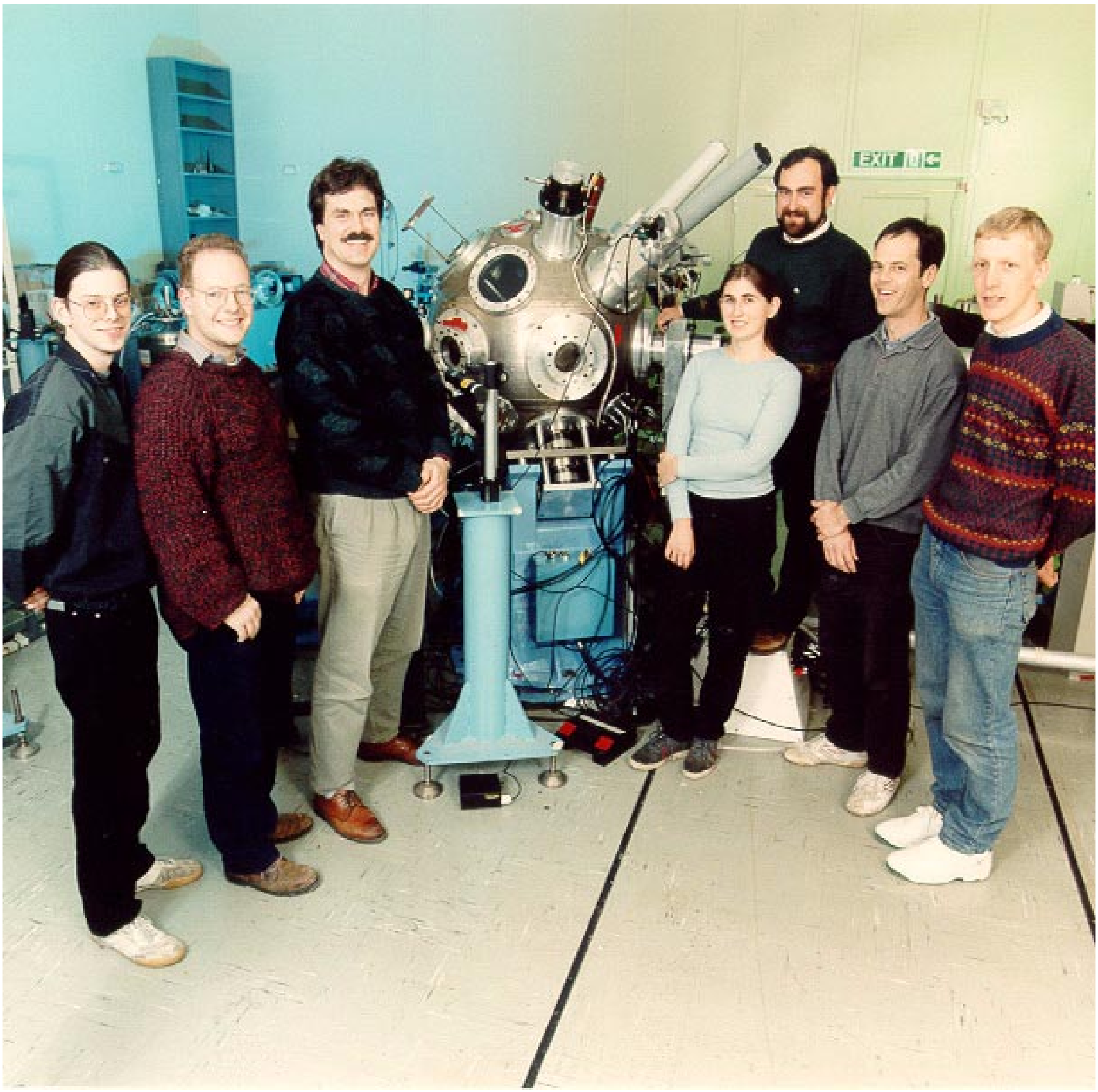
SUMMARY

This report describes the experiment entitled "Efficient X-UV Harmonic Generation, from solid targets, by the TITANIA KrF laser system" that was carried out at the Central Laser Facility from 18th Nov to 17th Jan 1997. Six weeks of experimental time was allocated between these dates. The experiment, funded by the TMR Large Facilities Access scheme was proposed by Professor Stavros Moustaizis, University of Crete. He was supported by researchers from the UK, co-ordinated by Dr Peter Norreys of the CLF, Rutherford Appleton Laboratory.

Experimental Results

- Observation of the 3rd and 4th harmonic emission from a UV laser solid interaction
- A transition between X-UV harmonic emission into a cone (whose angle was similar to that of the reflected laser beam) and isotropic emission was observed as the intensity on target was raised.
- The conversion efficiency as a function of angle peaked at 28°, which is similar to earlier resonance absorption measurements.
- Reasonable agreement with particle in cell simulations for density scalelengths of $L/\lambda = 0.8$.

The CLF makes beam time at its facilities available to European Researchers with funding from DG-XII, CEC under the Large Scale Facilities Access Scheme. For further information contact Dr. Chris Edwards at the CLF. Tel: (0)1235 445582, e-mail: c.b.edwards@rl.ac.uk



From Left to Right:

R Clarke, D Chambers, R Marjoribanks, J Knott, S Moustaizis, J Lister and E Divall

Arising Publications

One scientific paper has been accepted for publication in the scientific literature. The results have also been presented at two interaction conferences: The Conference on Lasers and Electro-Optics Pacific Rim 97, and the 27th Anomalous Absorption Conference, Vancouver, Canada, 1997.

Refereed Publications

‘Feasibility study of high harmonic generation from short wavelength lasers interacting with solid targets’
D.M.Chambers, P.A.Norreys, A.E.Dangor, R.S.Majoribanks, S.Moustaizis, D.Neely, S.G.Preston, J.S.Wark, I.Watts and M.Zepf
Optics Communications (in press)

Conference Presentations

The 27th Anomalous Absorption Conference, Vancouver, Canada 1st - 5th June 1997.
“Generation of harmonic radiation from the interaction of terawatt lasers with solid targets”
D.M. Chambers, A.E. Dangor, M.H. Key, R.S. Marjoribanks, S.Moustaizis, D. Neely, P.A. Norreys, S.G. Preston, J.S. Wark, I. Watts, M.Zepf.

CLEO / Pacific Rim conference 14 -18 July 1997 in Makuhari, Tokyo, Japan.
"X-UV harmonic generation by ultra-intense (sub)picosecond laser pulse irradiation of solid targets"
P.A.Norreys, D.Neely, M.Zepf, F.N.Beg, P.Lee, A.E.Dangor, D.Chambers, J.S.Wark, S.Moustaizis, R.Majoribanks.

Internal Reports

“Experimental study to test the viability of high harmonic generation from solid targets with short wavelength lasers”.
D. M. Chambers, P. Norreys, A. E. Dangor, R. S. Marjoribanks, S. Moustaizis, D. Neely, S. G. Preston, J. S. Wark, I. Watts, M. Zepf
Central Laser Facility Annual Report 1996-97, RAL-TR-97-045

EXPERIMENTAL INTRODUCTION

There is currently great interest in the production of extreme ultraviolet (XUV) radiation by high harmonic generation. One method of generating the harmonics is from the interaction of intense sub-picosecond laser pulses with solid targets[1) 2)]. The electrons oscillate at close to relativistic velocities across the solid-vacuum boundary (for ultra-short laser pulses with negligible pre-plasma formation) or across the boundary of a ponderomotively-steepened density profile in the case of pre-plasma formation. The harmonic production mechanism can be viewed simplistically either as due to rectification of the acceleration of electrons[3)], or by treating the boundary layer as an oscillating mirror driven by the ponderomotive force[4)]. Both odd and even harmonics are produced due to the non-symmetric nature of the boundary layer.

The first experimental work in this area was carried out using CO_2 lasers (10.6 µm) in the late 1970's by Burnett et al.[5)] . The highest harmonic eventually produced was the 46th harmonic in 1981 by Carman et al.[6)], though this relates to a harmonic wavelength of only 230 nm, far from the XUV region. Recently there has been a resurgence in this work due to rapidly increasing optical laser intensities [1, 7)] made possible by the development of chirped-pulse-amplification (CPA) technology[8)]. Importantly, simulations performed by Paul Gibbon show that both the order and efficiency of harmonic production is only dependent on $I\lambda^2$ (where I is the intensity and λ the fundamental laser wavelength) and that over 60 harmonics can be produced with efficiencies over 10^{-7} at 10^{19} Wcm^{-2} µm^{2} [3)]. Experimental work carried out by Norreys *et al*. has demonstrated the production of the 75th harmonic at 14 nm from the Vulcan 1.053 µm Nd:Glass laser in good agreement with particle-in-cell (PIC) code simulations[1)].

If Gibbon's predictions are correct, this method of harmonic generation may provide a means of production of high brightness coherent XUV radiation in the so-called water window, between 4.4 and 2.3 nm. For a KrF-pumped Raman laser, starting with a wavelength of 268 nm, the water window is reached at the 62nd harmonic. Using Gibbon's $I\lambda^2$ scaling[3)], and the results of Norreys *et al.*[1)] we could expect this harmonic to be obtained with an intensity of approximately 1.6×10^{20}Wcm^{-2}. This should be within reach of the forthcoming upgrade to the Titania KrF-pumped Raman laser[9)].

It is therefore timely to investigate the production of harmonics from the interaction of short-pulse UV laser radiation with solid targets. In the experiments described here, with the Titania CPA laser, the intensity on target was limited to 2×10^{18} Wcm^{-2}, and thus wavelengths of extremely high order are not expected. However, the experiments are still of significant interest, as they allow us to make comparisons with both the experimental results obtained using longer wavelength lasers, and with code calculations, thereby contributing to our overall understanding of the harmonic production process.

EXPERIMENTAL ARRANGEMENT

The experiment was performed using the TITANIA KrF laser[10)] in CPA configuration to give 1 ps pulses at 248.6 nm. Energies on target varied between 150 and 200 mJ, focused with an f/3.3 off-axis parabola to give intensities up to 2×10^{18} Wcm^{-2} at the target in a ~5× diffraction limited spot of 4 µm at full width half maximum. The targets consisted of optically polished slabs of fused silica, the beam was p-polarised with respect to the target.

The harmonic radiation was collected with a slitless flat field grazing incidence XUV spectrometer (based around a Hitachi 1200 l/mm grating at 4°), utilising a grazing incidence gold coated focusing mirror for increased collection efficiency. The grating and mirror were orientated so that their grazing incidence reflections gave perpendicular astigmatic line images of the focus of the laser at the detector plane. The XUV radiation was detected with a Galileo double micro channel plate (MCP), coupled to a Photonic Science DarkStar intensified charge-coupled-device (CCD) camera via a 4:1 reducing fibre optic coupler. The response of the MCP and CCD camera were calibrated in a separate experiment with respect to Illford Q-Plate film[11)]. The acceptance solid angle of the grating and gold mirror was 5.76×10^{-6} sr. A beam block was inserted in the zero order position to eliminate scattered fundamental and XUV radiation. The spectrometer viewed the interaction region at an angle of 56º to the incident laser beam.

RESULTS AND DISCUSSION

Figure 1 shows a typical line-out of the single-shot data showing the 3rd and 4th harmonics and rising plasma emission to shorter wavelengths. This data was obtained with the spectrometer viewing in specular reflection, i.e. an angle of incidence of the laser of 28º. The 4th harmonic signal was the highest order detectable, and only seen at the higher laser intensities.

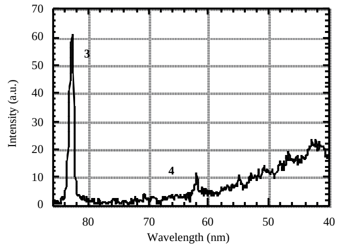


Figure 1. Typical lineout of the data taken with a laser intensity of $2{\times}10^{18}$ Wcm^{-2} , at an incidence angle of 28º, with the spectrometer in the specular reflection direction.

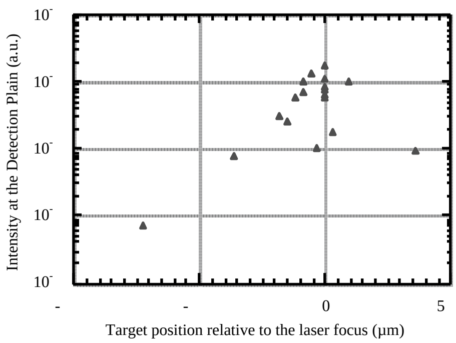


Figure 2. Third harmonic intensity at the detector, as a function of target position relative to the laser focus

The dependence of the collected third harmonic energy with incident laser intensity was investigated by moving the target through the laser focus. The results are shown in figures 2 and 3. The intensity at focus was calculated assuming a five times diffraction limited spot, consistent with equivalent-plane far field images taken from leakage through the penultimate turning mirror before the target. Intensities for all the other data points have been calculated assuming the beam to be in the geometrical limit, being at least three Rayleigh ranges from focus. As can be seen in figure 3 when the collected energy is now plotted against $I\lambda^2$ an interesting feature develops around 10^{16} $Wcm^{-2}\mu m^2$. For the reasons outlined below we identify this feature as the transition from specular to diffuse emission of the harmonic radiation.

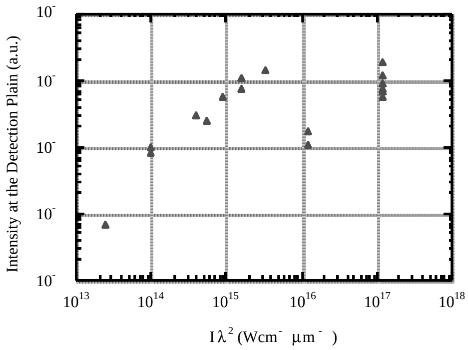


Figure 3. Third harmonic intensity at the detector as a function of $I\lambda^2$

In previous experiments, using Ti:Sapphire lasers (800 nm) with pulse lengths of between 60 and 150 fs giving intensities up to 1×10^{18} Wcm^{-2}, it was assumed (though not experimentally verified) that the harmonic emission was specular - i.e. confined to the cone angle of the reflected laser pulse[7,12)] . This assumption was made because the component of the wavevector parallel to the surface is conserved in coherent non-linear processes at the steep solid-vacuum interface. However, in contrast, both the work of Norreys *et al.* using picosecond pulses at close to 10^{19} Wcm^{-2}, [1)], and the early work utilising nanosecond CO_2 radiation[5,6)] , demonstrated diffuse harmonic emission. This broad angular emission has been attributed to rippling of the critical density surface by Raleigh Taylor like instabilities as the expanding plasma is pushed back by the ponderomotive force[13)] .

Importantly, in their recent studies of second harmonic radiation of clean, pre-pulse free,1µm, 1ps, Nd:Glass CPA laser pulses, Marjoribanks *et al.*[14)] , demonstrated that the second harmonic emission was specular over incident values of $I\lambda^2$ ranging from 10^{14} $Wcm^{-2}\mu m^2$ to the mid 10^{16} $Wcm^{-2}\mu m^2$. However, with the removal of a saturable absorber (introducing a pre-pulse 1.5 ns before the main pulse at $\sim10^{-4}$ of I_{peak}) they observed a transition from specular to diffuse emission as the irradiance was increased from approximately 10^{15} $Wcm^{-2}\mu m^2$ to 10^{16} $Wcm^{-2}\mu m^2$. All of the above observations are consistent with the view that if the laser interacts with a pure solid-vacuum interface, the harmonic emission is specular, but if it interacts with a plasma (with the interface being formed via ponderomotive steepening) at sufficiently high intensity, diffuse emission occurs due to an intensity-dependent instability at the critical surface.

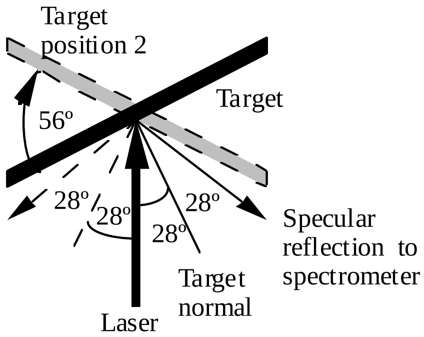


Figure 4. Schematic of the targets angular position with respect to the incident laser beam. The black target and solid lines represent the normal target position 1, with the spectrometer in the specular reflection direction. The grey target and dashed lines represent target position 2 with the angle of incidence, at 28°, and the spectrometer position remaining the same, but with the specular reflection direction being 112° from the previous direction.

In order to demonstrate that the emission at the higher intensities was indeed diffuse in this regime, the target was rotated, such that the laser was incident on target at the complementary position, i.e. keeping the incidence angle at 28°, but now with the spectrometer viewing far from the specular direction at an angle of 84° to the target normal, 112° from the specular reflection direction, as shown as position 2 in figure 4. In this new spectrometer position, strong 3rd harmonic emission was still detected at tight focus, of a comparable intensity to that observed in the specular direction. Further credence to this interpretation is afforded by plotting the third harmonic conversion efficiency as a function of irradiance, whilst assuming that the emission is into 2π for values of $I\lambda^2$ in excess of 10^{16}Wcm$^{-2}\mu$m^2, but into an f/3.3 cone for irradiances below this value. The results are shown in figure 5. The consistent fit of all the data plotted gives weight to the interpretation that between 2×10^{15}Wcm$^{-2}\mu$m^{2}and 2×10^{16}Wcm$^{-2}\mu$m^2 the angularly-integrated conversion efficiency continues to rise, but the collected energy temporarily decreases (see figure 3) due to the transition from specular to diffuse emission. Similar conclusions have been reached previously by Marjoribanks and co-workers[14)] .

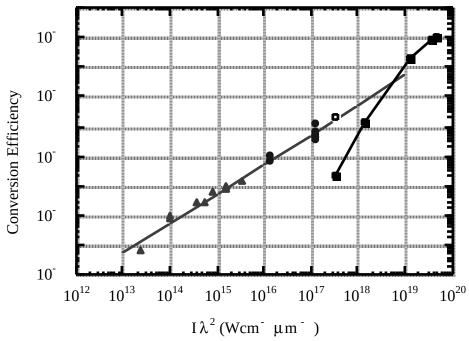


Figure 5. Conversion efficiency into the third harmonic versus incident laser irradiance. Triangles represent emission into the laser cone angle only, the circles emission into 2π steradians. The straight line fit represents 5.2 x $10^{-22} \times I\lambda^2$ The solid black squares and fit represent the PIC code simulation results of Lichters et al. for a step-like density function. The open square represents the PIC code result for a density scale length of 0.8 λ, to match the experimental conditions. Lichters results are taken from figures 2 and 9 in reference 15).

In addition to studying the intensity dependence of the harmonic emission at a given angle of incidence described above, we also investigated the influence of incidence angle of the laser on the harmonic emission. For these studies the target was rotated, leaving the spectrometer position fixed. In all cases the target was placed in the focal plane of the laser beam, and thus the incident intensities were of order 1×10^{18}Wcm^{-2}. The results are shown in figure 6, where we have assumed diffuse emission for all angles of incidence in calculating the conversion efficiency. It can immediately be seen that there is an extremely strong dependence of the emission on the angle of incidence of the laser, with the peak conversion occurring for angles of incidence around 30º. At an incidence angle of the laser of 56º and with the spectrometer in the specular reflection direction no harmonic emission was detected. Thus there is a strong angular dependence of the harmonics, showing a reduction in conversion efficiency of greater than 3 orders of magnitude from a change in incidence angle from 28º to 56º.

This optimum angle of incidence is consistent with previous measurements of energy absorbed via resonance absorption made by Borghesi and co-workers using almost identical irradiance conditions on the Rutherford KrF laser system[16)]. Their results are shown for comparison in figure 7. The resonance absorption clearly peaks around the same angle as the harmonic radiation maximises. Borghesi's measurements would indicate that at an angle of incidence of, say, 10 degrees, the absorbed fraction decreases by a factor of 2 or so from the peak. Comparison with figure 6 would indicate that this decrease in absorbed energy is insufficient on its own to account for the factor of a few drop in harmonic conversion efficiency observed in moving from an angle of incidence of 30 to 10 degrees, though considering the error bars associated with this data, further work is obviously needed to verify this conclusively.

Finally it is of interest to compare the experimental conversion efficiencies with computational predictions. Recently, Lichters et al. have shown with a fully relativistic one dimensional PIC code, including a step-like density function, that strong oscillations are generated at the critical density surface by the ponderomotive force. They proceed from this to show that the harmonic generation process can be understood in terms of the light being reflected from an oscillating mirror[4,15]. This "oscillating mirror model" was first proposed by Bulanov et al. in 1994 [17] . The predicted harmonic conversion efficiencies, from the PIC code simulations with a step-like density function, are shown alongside the experimental data in figure 5.

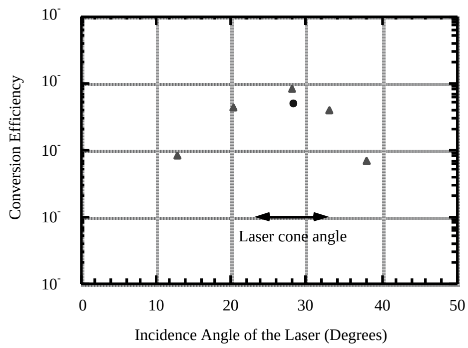


*Figure 6. Conversion efficiency into the third harmonic versus angle of incidence of the laser with respect to the target normal. The triangles represent the average of at least 2 data points, with the target in position 1. The circle represents emission collected with the target in position 2, see figure 4.*_*The black arrow represents the angular divergence of the laser*

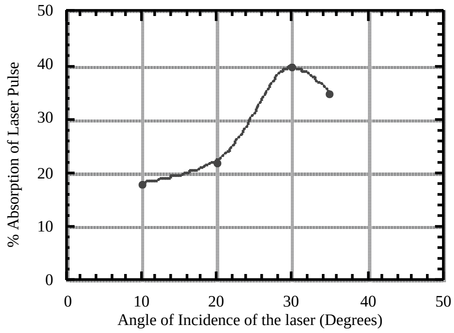


Figure 7. Percentage of the laser pulse absorbed by resonance absorption versus incidence angle of the laser, as measured by Borghesi et al. (16) .

More sophisticated PIC code simulations, using a density ramp to simulate different plasma density scalelengths, again provided by Lichters, go on to show for the lower harmonics, that an increase in conversion efficiency occurs with increasing density scalelength up to the order of a wavelength[15)] . Lichters et al. have suggested an explanation for this in terms of the oscillating mirror model, i.e. that the laser can drive larger oscillations of the critical density surface due to the weaker plasma restoring force. Obviously though, with too long a density scale length, the idea of a reflecting surface becomes somewhat blurred, and as expected the simulations show that the harmonic efficiencies are reduced with scalelengths over a wavelength. This explanation is backed up by the PIC code simulations, including step-like density functions, which also predict increased harmonic intensities with reduced plasma densities.

In order to make more consistent comparison with the Lichters simulations, an estimate of the plasma scalelength present in the experiment can be made from our knowledge of the angular dependence of the resonance absorption. The standard results of resonance absorption allows the plasma density scale length to be estimated from the formula[18)] .

$$\sin\phi = 0.8(\omega L/c)^{-1/3}$$

Where ϕ = the laser incidence angle for maximum absorption, ω = the fundamental laser frequency and L = the plasma density scale length in units of the laser wavelength. This implies the plasma density scale length to be 0.8 (±0.1) λ, i.e. 0.20 (±0.02) µm. Indeed, for this plasma density scalelength Lichters PIC code simulations predict a conversion efficiency of 2×10^{-4} at an irradiance of 3×10^{17} $Wcm^{-2}\mu m^2$. This point is also shown alongside the experimental data in figure 5. The good agreement is now striking, and gives confidence that theoretical models can provide reasonable indicators of expected conversion efficiencies.

In conclusion we have investigated the generation of the third harmonic of a picosecond KrF laser interacting with a solid target. Both the intensity and incidence angle dependence of the conversion efficiency have been explored. The conversion efficiency has been shown to vary almost linearly with $I\lambda^2$ from about 10^{-8} at 10^{13} to 10^{-4} at 10^{17} $Wcm^{-2}\mu m^2$ with evidence of angular broadening from specular to diffuse emission between 2×10^{15} $Wcm^{-2}\mu m^2$ and 2×10^{16} $Wcm^{-2}\mu m^2$. This broadening is consistent with previous studies of the second harmonic of a Nd:Glass CPA laser performed by Marjoribanks and co-workers[14)]. The conversion efficiency as a function of angle of incidence was investigated at values of $I\lambda^2$ of order 10^{17} $Wcm^{-2}\mu m^2$ showing a strong dependence on the incidence angle of the laser. Harmonic generation peaked at an angle of incidence close to 30 degrees, consistent with the angle at which maximum resonance absorption was found by Borghesi and co-workers using similar irradiance conditions. Most encouragingly, these results are in agreement with theoretical simulations using a fully relativistic, one dimensional PIC code with appropriate plasma density scale lengths [15)]. These results are therefore very encouraging for forthcoming KrF experiments with ultra high intensities, where if the theoretical predictions are correct, water window harmonics should be within reach.

The authors would like to acknowledge the help and support of the staff of the Rutherford Appleton Laboratory, UK This work was jointly funded by the United Kingdom Engineering and Physical Science Research Council (EPSRC) and the European Communities Large Access Programme. D. M. Chambers would like to acknowledge funding from the EPSRC and A.W.E., Aldermaston, UK S. G. Preston would like to acknowledge support from Merton College, Oxford, UK

REFERENCES

1) P.A. Norreys et al. Physical Review Letters, 76 (11), 1832-1835 (1996).

2) D.M. Chambers et al. Journal of Applied Physics, 81 (5), 2055-2058 (1997).

3) P. Gibbon. Physical Review Letters, 76 (1), 50-53 (1996).

4) R. Lichters et al. Physics of Plasmas, 3 (9), (1996).

5) N.H. Burnett et al. Applied Physics Letters, 31 172 (1977).

6) R.L. Carman et al. Physical Review A, 24 (5), 2649-2663 (1981).

7) S. Kohlweyer et al. Optics Communications, 117 431-438 (1995).

8) D. Strickland et al. Optics Communications, 56 219-222 (1985).

9) S. Angood et al. Rutherford Appleton Laboratory Technical Report No. RAL-TR-94-014, (1994).

10) E.J. Divall et al. Journal of Modern Optics, 43 (5), 1025-1033 (1996).

11) M. Zepf et al. Rutherford Appleton Laboratory Technical Report No. RAL-TR-95-025, 113-114 (1995).

12) D. Von der Linde et al. Physical Review A: Rapid Communication, 52 (1), R25-R27 (1995).

13) S.C. Wilks et al. Physical Review Letters, 69 (9), 1383-1386 (1992).

14) R.S. Marjoribanks et al. Bull. Am. Phys. Soc., Denver (1996).

15) R. Lichters et al., In Multiphoton Processes, IOP Publishing Ltd, 221-230 (1997).

16) M. Borghesi et al. Physical Review E, To be published. (1997).

17) S.V. Bulanov et al. Physics of Plasmas, 1 (3), 745 (1994).

18) C.E. Max. Physics of Laser Fusion, 1 Lawrence Livermore Laboratory Report (1981).