UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION
INTERNATIONAL ATOMIC ENERGY AGENCY
INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
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H4.SMR/984-20

Winter College on Quantum Optics: Novel Radiation Sources

3-21 March 1997

The generation of ultra-high harmonics of laser radiation

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COHERENT ULTRA-BRIGHT XUV LASERS AND HARMONICS

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Over the past decade considerable advances have been made in the generation of coherent XUV radiation. Progress has been made on three main fronts: the development of both collisional and recombination XUV lasers, the generation of high odd-order harmonics from the interaction of intense sub-picosecond lasers with noble gases, and the generation of both odd and even harmonics from ultra-intense sub-picosecond laser interactions with ponderomotively-steepened high-density plasmas.

1 Introduction

The extension of laser action into the x-ray region has been an area of increasingly successful experimental investigation for over a decade, and the subject of theoretical inquiry for more than twice that length of time. The frequency scaling of spontaneous and stimulated emission rates dictates that direct XUV laser action can only be achieved at extremely high power densities: thus the focused output of high power optical lasers has been the main method used to pump x-ray lasers.

In addition to the direct manufacture of x-ray laser media, significant advances have been made over the past few years in the production of coherent XUV radiation by the generation of ultra-high harmonics of high intensity, sub-picosecond laser pulses. Two methods of such harmonic generation have been explored. Firstly, the interaction of intense laser pulses with gaseous targets (primarily noble gases). In the intense laser field, comparable to the Coulomb field in the atom, electrons can tunnel through the Coulomb barrier, giving rise to a large non-linear susceptibility. Symmetry of the atomic potential dictates that only odd-order harmonics are generated. Such harmonics are typically generated with sub-picosecond laser pulses at irradiances of between 10^{14} and 10^{16} Wcm^{-2}. Note that atomic unit of field (i.e. the field experienced by the electron in the first Bohr orbit of hydrogen) is 5.142 x $10^{11} Vm^{-1}$, corresponding to a laser intensity of 3.52 x 10^{16} Wcm^{-2}. Secondly, if an extremely intense pulse (10^{17} - 10^{19} Wcm^{-2}) is incident onto a solid target, both odd and even harmonics can be produced. These harmonics are associated with the electron current being dragged back and forth across the density step. At the lower end of the irradiance range indicated above, and for ultrashort (say, 100-fsec) high contrast-ratio laser pulses, the relevant density step is that of the solid-vacuum interface itself, whereas for higher irradiances and longer pulselengths (of order a picosecond or longer), the density step is produced by ponderomotive steepening of the pre-plasma, formed before or during the leading edge of the pulse.

The three methods of XUV generation outlined above produce sources with widely different divergences and coherence properties. In this paper comparison of the present status of these source characteristics will be made, and a preliminary assessment of their applicability for other areas of science outlined.

2 X-ray Lasers

The first XUV laser scheme to achieve high gains, produced using the Nova laser facility at Lawrence Livermore National Laboratory, relied on collisional excitation of the Ne-like Se ion.[1] Production of the high density ($10^{21} cm^{-3}$) high temperature laser medium was achieved by irradiation of a thin film target by nanosecond pulses of optical (0.53-μm) light at intensities of order 5 x 10^{13} Wcm^{-2}. Soon after, laser schemes that relied on achieving population inversion by preferential recombination of electrons into upper states of ions in adiabatically cooling plasmas were demonstrated.[2,3] Since this ground breaking work the field has advanced on several fronts: the reduction of laser wavelength towards the so-called water-window at 44-Å;[4] the development of oscillator-amplifier modes of operation and the improvement in laser efficiency by the reduction of the effects of refraction due to electron density gradients within the gain region;[5–7] and the demonstration of "table-top" schemes utilising picosecond lasers[8] or capillary discharges.[9]

The majority of work in this area has concentrated on the collisionally pumped schemes, with Neon-like ions being used for wavelengths in excess of about 100-Å, and Nickel-like ions below this wavelength. For many years the output of these Neon-like systems was poorly understood: in particular, detailed modeling predicted higher gain on the $J = 0 - 1$ transition compared to that on the $J = 2 - 1$ doublet. This anomaly has recently been resolved. It has been found that the reduction of the $J = 0 - 1$ output was due to severe refraction of x-rays out of the lasing medium due to the steep electron-density gradient close to the surface of the target. These refraction effects are more severe for the $J = 0 - 1$ transition, as its gain region is at high densities, and correspondingly higher refractive index gradients. Reduction of

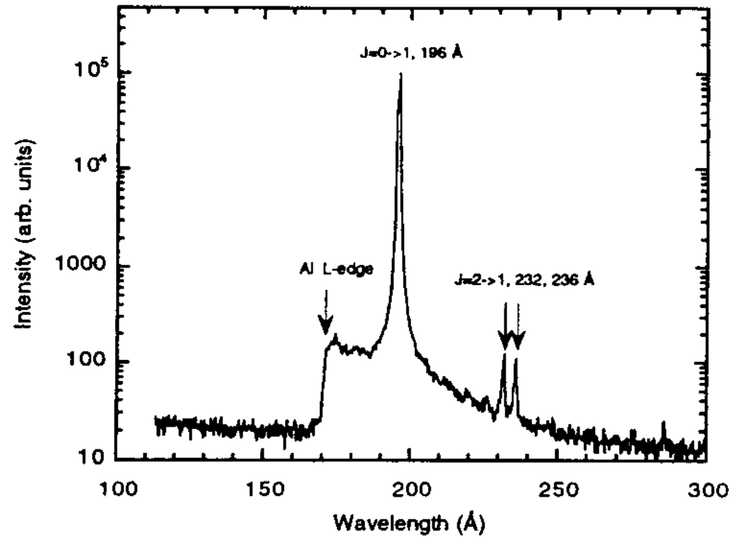

Figure 1: Spectrum of a Neon-like Germanium laser operated with a prepulse, illustrating the domination of the 19.6-nm line.

the density gradients, and consequent alleviation of refraction problems, has been achieved by operating the optical drive laser with a small prepulse (10^{-4}-10^{-1} in power) a few nanoseconds before the main pulse. The prepulse forms a relatively long scalelength plasma, which is then heated by the main pulse. This reduction in density gradient has been shown to increase the power of the $J = 0 - 1$ to over two orders of magnitude that of the $J = 2 - 1$ line, with a significant reduction in pulse length.[5-7] Fig. 1 shows the output spectrum of a Neon-like Germanium laser operated with such a prepulse.[10]

Use of this prepulse technique has greatly reduced the energy of the optical laser needed to produce saturated laser output. To increase the brightness of the source further it will be necessary to reduce the divergence of the beam towards the diffraction limit. At present, typical beam divergences from the 50-μm diameter laser are of order 10 to 25-mrad, and are not significantly better than calculated divergences based simply on the aspect ratio of the lasing medium - i.e. the beam is far from coherent, although improvements in target geometries are improving this situation.

3 Harmonics from gaseous targets

In a nanosecond high-power laser pulse, as used for the most of the XUV laser production described above, the oscillatory motion of the electrons in the laser-field is randomised by collisions with ions, thus heating the electrons to high temperatures (typically up to a keV). The impact of the electrons with the ions is the mechanism by which further ionization proceeds, and thus, though by no means in thermal equilibrium, the ionization stage reached is characteristic of the electron

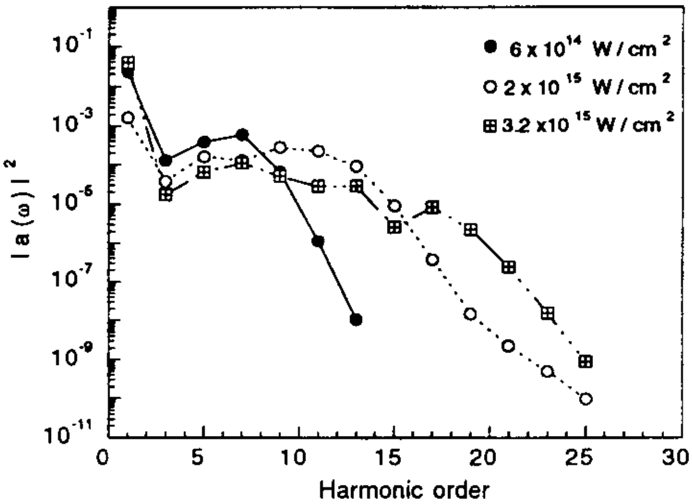

Figure 2: Simulations of the harmonic spectra of Helium for a short 248-nm pulse with a linear turn on, taken from the work of Sanpera et al (Ref. 11)

temperature. In contrast, if a very intense short pulse interacts with a target of sufficiently low density (e.g. a gas), then there is insufficient time for electron-ion collisions to occur. Under these circumstances the only mechanism available for ionization is direct ionization due to the laser field itself - i.e. at high fields the electron quantum-mechanically tunnels out of the Coulomb potential and continues to oscillate freely in the laser field: this is known as optical ionization. If the laser pulselength is sufficiently short, electrons in neutral atoms and low ionization stages can experience very high laser fields before significant optical ionization to the next ion stage occurs. This allows us to study the highly non-linear response of such neutrals and singly-ionized ions to extreme laser fields comparable to the Coulomb field of the atom, and far beyond the point where perturbation theory is valid.

The probability of the electron tunneling through the Coulomb barrier is a highly non-linear function of the laser intensity. Furthermore, after tunneling, the amplitude of the electron oscillation in the laser field is large - typically of order 10 - 100 bohr radii. Thus the susceptibility of the atom is extremely large and non-linear, and harmonics are produced. A typical harmonic spectrum, in this case simulated by a time-dependent solution to the Schrödinger equation in the single active electron approximation, is shown in Fig. 2, taken from the work of Sanpera et al.[11] Only odd orders are produced, due to the symmetric nature of the atomic potential. The generic features of harmonic spectra are a rapid decrease in production efficiency over the first few (say 5-th to 7-th) harmonics, followed by a plateau in the response, with a relatively sharp cut-off. The cut-off has been shown to occur at a photon energy $\hbar\omega$ corresponding to approximately

$$\hbar\omega = I_p + 3.17U_p \qquad (1)$$

where I_p is the ionization potential of the atom, and U_p is the ponderomotive energy of the electron quivering in the laser field, given by

$$U_p = \frac{e^2 E_0^2}{4m\omega_0^2} \quad (2)$$

where E_0 is the electric field of the laser, and ω_0 its freqency. This dependence is shown in Fig. 2, where the cut-off energy is shown to increase as the laser intensity increases.

Somewhat surprisingly, many of the salient features of the harmonic spectra can be understood in terms of the very simple semi-classical model put forward by Corkum, which is outlined below.[12] As the field of the laser oscillates, the probability of the electron tunnelling through the Coulomb barrier alters. Depending on the time of tunnelling relative to the phase of the laser, from a semi-classical point of view three scenarios present themselves. As the free electron oscillates in the laser field it can either have a time-averaged motion away from the vicinity of the parent ion, such that it never recrosses the ion, or, secondly, it can follow a trajectory which causes it to recross the ion core in the first (and perhaps several subsequent) laser cycles. In this simple semi-classical view, it is during such recollisions that the electron has a probability of returning to the ground state of the atom, emitting a harmonic photon with an energy of the ionization potential of the atom plus the kinetic energy of the electron in the oscillating field at the time of recollision. Finally, if the electron tunnels through the barrier exactly at the peak of the laser field, it will oscillate with zero time-averaged velocity, with one of its extrema being the ion core (if we neglect that fact that the free electron is not actually 'born' at the centre of the ion - i.e. we assume the amplitude of electron motion in the laser field is significantly greater than atomic dimensions - a good approximation at these laser intensities).

Corkum demonstrated that the cut off in photon energy described in equation (1) can be interpreted using the classical equations of motion of an electron. If we assume that the tunnel-ionized electron is produced at the ion core at some time in the laser cycle, and then solve the equations of motion to determine the kinetic energy of the electron upon its first recollision with the core, we find that the maximum kinetic energy an electron can achieve is $3.17U_p$, for electrons that are produced at a phase angle of 17° relative to the laser field.

From the above description it can be seen that the processes of harmonic generation and optical ionization are inextricably linked. After the electron tunnels through the Coulomb barrier it has some finite probability of recombining with the core. This probability is greatest for the first recollision: in Corkum's semi-classical model the electron wavefunction diffuses in a direction transverse to the oscillation axis, and thus the probability of recombination rapidly decreases. If the electron does not recombine with the core, it drifts away from the parent ion, and we can consider the atom ionized. Thus it can be seen that for a given atom the ponderomotive energy of the freed electron cannot increase without limit with laser intensity due to depletion of the parent atoms by ionization. Thus the magnitude of the second term in equation (1) is dictated by the maximum laser intensity that an atom can experience before significant optical ionization occurs. In quantum

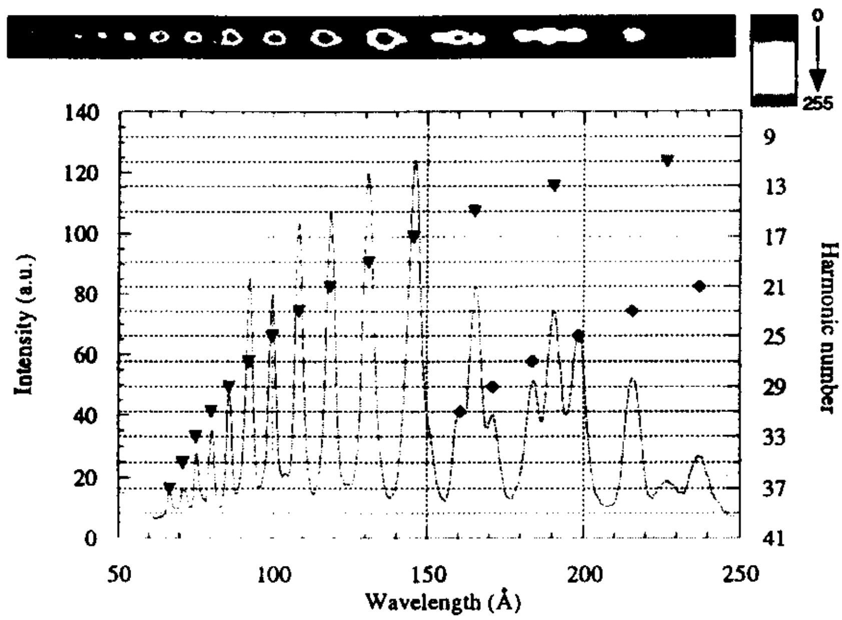

Figure 3: Harmonics generated from the interaction of a KrF (248-nm) laser with a Helium target, taken from the work of Preston et al (Ref. 24)

mechanical terms, the probability of the electron wavefunction remaining in a state corresponding to a bound level in the atom decreases with increasing laser intensity. However, those small number of neutral atoms that do survive to high intensities will generate high energy harmonics efficiently once they do eventually ionize. Furthermore, it can be seen why most work has been performed using noble gases: they can produce the highest harmonic photon energies and efficiencies as they have the highest neutral atom ionization potentials. This effects both terms on the right hand side of equation (1), in that the higher the ionization potential, the higher the laser intensity (and thus pondermotive energy) required for optical ionization.

As well as considering the response of single atoms, to find the overall conversion of laser light to XUV harmonics we must also consider phase matching effects. The fundamental laser and harmonic photon will dephase in the gaseous medium. Such effects will be far more severe in the presence of free electrons due to the far higher (negative) susceptibility at the fundamental wavelength for free electrons than the (positive) susceptibility of the neutral gas. Indeed, for neutral gases at low density the phase matching is often dominated by the geometrical phase factor of a Gaussian beam. This immediately leads to the question of the best means of maximising harmonic yield: is it better to use relatively low laser intensities, such that both the harmonic response and thus ionization of atoms is low, but the phase matching relatively good (the fundamental and harmonic propagate in a mainly neutral gas), or use high laser intensities, such that those atoms that do survive to high intensities have an extremely non-linear response, but the harmonics that are produced by each atom remain in phase with the fundamental over a significantly

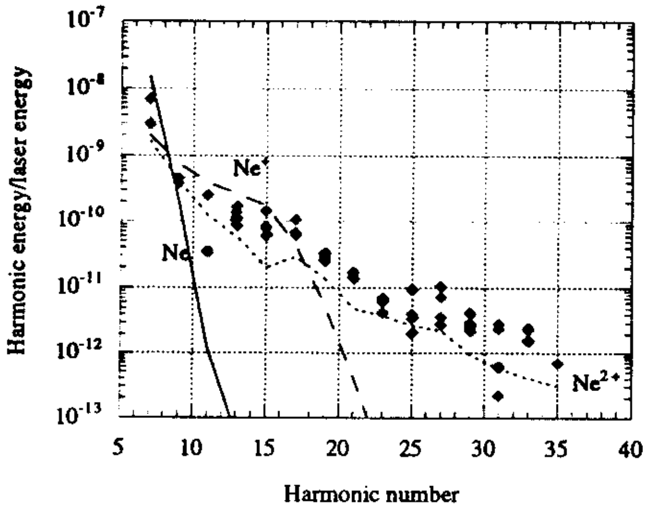

Figure 4: Comparison of experiment and theory for harmonics from Neon, taken from the work of Preston et al (Ref. 24)

shorter length, being dephased by the free electrons?

In most of the early pioneering work in the field the former conditions were used,[13–18] and detailed studies of the effects of the geometrical phase factor were performed. More recently, the scaling of conversion efficiency with atomic density has been investigated.[19] However, the work of Ditmire and co-workers established that the highest conversion efficiencies would indeed be produced at saturation intensities and above,[20] thus in the light of the above discussion, for efficient harmonic production we necessarily are operating in the presence of free electrons. Furthermore, in this regime Ditmire also showed that shorter wavelength drivers were more effective at producing harmonics, although the cutoff in photon energy given by equation (1) was necessarily reduced to the the quadratic scaling of the ponderomotive term (equation (2)) with fundamental wavelength. The increased efficiency of shorter wavelength drivers is attributable to two effects. Firstly, for a high harmonic of a given photon energy the dephasing length between fundamental and harmonic scales as ω_0^2, and thus the scaling of harmonic yield due to phase effects scales as ω_0^4. The second effect can be understood once again in terms of Corkum's semi-classical model: the electron that has tunneled through the Coulomb barrier returns to the core more rapidly (in approximately one half of a laser cycle) for a short wavelength drive, thus its wavefunction has had less time to diffuse, and the cross section for harmonic production increases. This increase in conversion efficiency for shorter primary wavelengths has been verified by detailed modeling.[21]

Thus far we have been considering the response of neutral atoms. Once the neutral has been optically ionized, we would expect that harmonics could be produced from the action of the laser field on the subsequent ionization stage. As the ion is more tightly bound, we would expect a reduction in the harmonic conversion efficiency compared to the neutral due to the reduced cross section of the electron with the ion core. However, as the inherent efficiency of harmonic production with a short wavelength driver is higher, harmonics from ions may still be observed: the higher ionization potentials of the ions will increase the cut-off energy. Thus it is interesting to enquire whether higher energy harmonics can be produced using short wavelength lasers interacting with ions than with long wavelength lasers interacting with neutrals. The majority of groups working in this area have used relatively long wavelength lasers (e.g. Ti-Sapphire or Neodymium glass). The highest harmonics produced with such long-wavelength drivers are the 109-th of Ti-Sapphire (800-nm) at 74-Å[22] and the 141-st of Nd:glass (1053-nm) at 75-Å.[23]

Thus far, harmonic generation from ions has not been identified in the experiments with 1.05-μm drivers. Although there is some indication of ion response with 0.53-μm light, it is with 0.248-μm radiation that the effect of ions has been definitively recorded. In Fig. 3 we show results by Preston et al, where they observed upto the 37-th harmonic of a KrF (248-nm) laser - a harmonic wavelength of 67-Å- in interactions with a Helium target, and upto 35-th harmonic in interactions with a Neon target.[24] Comparison with simulations has shown that the highest energy harmonic radiation for Neon was produced by the doubly-ionized atom (see Fig. 4). Simular conclusions of evidence of ion response have been reached by Krause et al,[25] in their analysis of the work of Sarakura.[26]

4 Harmonics from solid surfaces and ponderomotively steepened plasmas

In addition to harmonic generation from gaseous targets, there has recently been a renewal in interest in generating high order harmonic radiation from high-power laser interactions with solid targets.[27–31] Such high harmonics were first observed in nanosecond experiments using CO_2 lasers at irradiances of order 10^{15} Wcm^{-2} (where upto the 46th was observed), where the long laser wavelength (10.6-μm) ensured significant ponderomotive steepening of the plasma density profile.[32–36] Both odd and even order harmonics are generated via the relativistic current associated with the electrons being dragged back-and-forth across this asymmetric density step. Due to the λ_0^2 scaling of the ponderomotive force, we would expect to observe similar phenomena using 1.05-μm lasers at irradiances in excess of 10^{17} Wcm^{-2}.

However, it should be stressed that for these ultra-short pulses we can conceive of two ways in which the laser can be incident upon a steep density profile. If there is no significant plasma expansion during the laser pulse, then the laser effectively interacts with the target-vacuum boundary. If, however, plasma expansion does take place, then if the ponderomotive force is sufficiently great it may be possible for significant ponderomotive steepening of the density profile to take place during the laser pulse. These two situations can be classed as true interaction with solids, and interaction with a ponderomotively steepened plasma.

Recently von der Linde et al reported the observation of the 15th harmonic from a 130-fs laser-solid interaction using a Ti:Sapphire laser at 800-nm with intensities

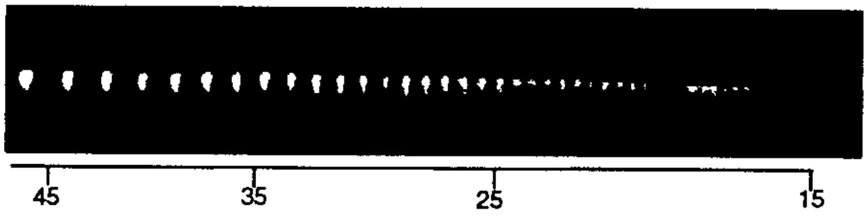

Figure 5: A spectrum of harmonics from ponderomotively steepened plasmas taken from the work of Norreys et al (Ref. 38)

upto 10^{17}Wcm^{-2}.[31] They interpreted their results as an interaction with a vacuum-solid step, conclude that the harmonics were produced in a specularly reflected narrow beam, and reported conversion efficiencies of order 10^{-8} to 10^{-9}.

For the situation of pondermotively steepened plasmas, Gibbon has recently performed PIC code simulations of harmonic generation for sub-picosecond pulses.[37] He concludes that for $I\lambda^2 > 10^{19}$ Wμm^{2}cm^{-2}, and modest shelf densities of order $N_e/N_{critical} = 10$, upto 60 harmonics can be generated with power conversion efficiencies of 10^{-6}. Importantly, Gibbon's simulations predict that the harmonic order is simply determined by $I\lambda^2$, thus short wavelength lasers should produce shorter absolute wavelengths for a given value of $I\lambda^2$. Thus short wavelength, intense lasers may eventually provide a route to shorter wavelength, higher conversion efficiency harmonics than have hitherto been generated.

The most spectacular work to date in this area has been performed by Norreys and co-workers.[38] They observed upto the 68th harmonic of 1.05-μm light in first order diffraction, with indications of 75th in second order with laser intensities on target upto 10^{19}Wcm^{-2}, and with energy conversion efficiencies estimated at ranging from 10^{-4} to 10^{-6}. The experiment was performed using the Chirped Pulse Amplification beam line on the VULCAN laser at the Central Laser Facility of the Rutherford Appleton Laboratory.[39] The laser produced pulses of 2.5 picoseconds duration and energies of around 20 J on target. The contrast ratio was measured to be better than 10^{-6} using a third order auto-correlator. A single shot auto-correlator allowed individual pulse lengths to be measured. The laser beam was focused onto the target by an f/4.2, 44 cm focal length off-axis parabolic mirror.

Fig. 5 shows a spectrum taken when 20.7 J of p-polarised laser energy in 2.6-psec was incident on a target consisting of 2-μm CH coating onto a metal sandwich target (25-μm Mo on 50-μm Pd). The maximum entropy deconvolved x-ray penumbral images established that the spot diameter was $\sim 9 - \mu$m full width half maximum (FWHM), yielding an intensity on target of 9×10^{18}Wcm^{-2}.

The harmonics were found to be emitted into a wide angular range, to be independent of additional prepulse, and to be insensitive to the polarisation of the incident beam. With the level of prepulse inherent in this laser, we would expect significant pre-plasma to be formed. These effects - no observable difference in harmonic generation between s and p polarisations, the very large angular distribution and the relative insensitivity to prepulse levels - suggest that the critical density surface is rippled during the interaction, as this blurs the distinction between s and p polarisation. The development of a Rayleigh–Taylor like instability at the critical surface has been observed in 2.5 dimensional PIC simulations when a high intensity, picosecond laser pulse interacts with a pre-formed plasma.[40,41]

5 Comparison of XUV sources

In comparing these three different XUV sources, we must first decide upon an appropriate figure of merit: this will generally be the spectral brightness - i.e. the power per unit area, per unit solid angle, per unit frequency interval. Accurate comparisons of the spectral brightness of the various sources is difficult, as in many cases the coherence of the XUV radiation has not been measured. In the case of XUV lasers, some spatial coherence measurements have been made,[42] but these were generally without the prepulses that have recently been shown to vastly improve the gain length per unit energy input by negating some of the deleterious effects of refraction (see section (2)) - although some work has started in this area.[43] By the definitions within the Van Cittert Zernicke theorem the best spatial coherence was found to be approximately equivalent to a 15-μm diameter incoherent source. This spatial coherence is not particularly good, when one considers that it is only a few times smaller than the laser aperture, and for the Neon-like Ge laser represents a system which is a few hundred times the diffraction limit! Despite this far from optimal coherence, the spectral brightness of such lasers is still remarkably impressive. For these collisional x-ray lasers, the fractional linewidth is of order 10^{-4}, which is mainly determined by thermal Doppler broadening. A comparison of the various sources, compiled by M.H. Key,[44] can be seen in Fig. 6, where we compare the lower bounds of the spectral brightnesses.

Again, for the harmonics generated from gaseous targets, information on the spatial coherence is sparse. Some coherence measurements have been made for the situation where little ionization takes place.[45] However, to our knowledge, no such equivalent measurements have been published for those situations where the laser intensity was close to, or exceeded, the saturation intensity, such as in the work of Ditmire and Preston cited above.[20,24] We would expect the coherence to be worse in these latter cases, as the electron-density, and hence refractive index, in the gas will depend on the laser intensity, thus degrading the beam quality. Thus, despite the fact that these measurements have yielded the greatest conversion efficiencies, it is not known for certain how the spectral brightnesses compare. In Fig.6 we have used the measured cone angle of the incident laser to define the brightness. Thus the figures given should be treated as a lower bound on the true spectral brightness. In general, due to pulselength considerations, the fractional linewidth of the harmonics from gaseous targets is of order 10^{-3}. If the gas ionizes during the pulse, some further spectral broadening and blue shifting can occur due to the time-dependent ionization (and hence time-dependent refractive index).

For the harmonics produced from pondermotively-steepened plasmas the situation is different again. In this case, the divergence of the source is large, as is the

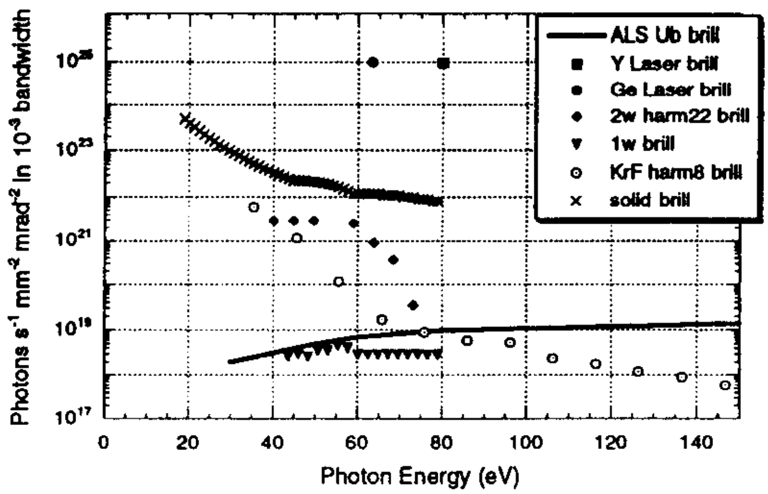

Figure 6: Comparison of the spectral brightness of various sources from the work of Key (Ref. 44). The KrF data is taken from Ref. 24, and the 1ω and 2ω data refers to the 1.05-μm and 0.53-μm data from Ref. 20.

fractional linewidth - which is of order 10^{-2}. This large linewidth is thought to be due to self phase modulation of the primary laser pulse as it traverses the plasma before reaching the critical density surface. There will also be some degree of spectral broadening due to the Doppler effect, as the critical surface is accelerated towards the target surface, with a peak velocity approaching 0.02 of the speed of light at an irradiance of 10^{19}Wcm^{-2}.[46] It should be noted that the spectral brightness is for most of these sources, at present, a few orders of magnitude greater than those available from synchrotron sources.

One particularly interesting feature to note from Fig.(6) is the superior instantaneous spectral brightness of harmonics from ponderomotively-steepened solids compared to harmonics from interactions with gaseous targets: the high conversion efficiency and small source size (the spot size of the high harmonics has been measured to be of order 2-μm) more than compensates for the high divergence and bandwidth of the source. With the large spectral coverage that these harmonics afford compared to x-ray lasers, they may prove to be a highly useful source for applications, such as non-linear optics in the XUV.

6 Applications

The two main areas in which x-ray lasers have currently been applied are interferometry of high-density (i.e. in excess of 10^{20}cm^{-3}) laser-plasmas[47] and radiography of laser accelerated foils.[48] The brightness of the Yttrium x-ray laser beam is equivalent to that from a several GeV blackbody - making it ideal for the probing of hot (keV) laser-produced plasmas. Furthermore, in such plasmas absorption and refraction render conventional optical techniques unsuitable.

Recent advances in multilayer mirror technology now allow XUV mirrors to be manufactured with reflectivities as high as 0.65, with a high degree of uniformity.[49] Furthermore, beamsplitters have been also been developed, with transmission and reflection coefficients of 0.15 and 0.2 at the Yttrium x-ray laser wavelength (155Å).[47] Using such optics, da Silva and co-workers have constructed an XUV Mach-Zehnder interferometer, and used it to diagnose the density profiles of laser-produced plasmas of relevance to laser fusion.[47]

The high brightness of the x-ray laser beam also allows it to be used to detect small thickness modulations in high opacity foils. If a beam is passed through a rippled foil, the intensity modulation due to the small variations in thickness is proportional to the thickness modulation and the product of the absorption coefficient and the thickness. Using such a technique Key and co-workers have measured the imprint pattern of a laser on a laser-accelerated thin silicon foil.[48,50] These measurements are crucial for direct drive laser-fusion research, as they help determine the degree of uniformity of illumination necessary for maintainance of the integrity of implosion of the fusion target.

High order harmonic radiation has also been used recently to diagnose high density plasmas.[51] The plasmas of interest, with densities in excess of $10^{23}cm^{-3}$ were themselves generated with a sub-picosecond laser pulse and thus, due to their highly transient nature, could not be probed with a conventional x-ray laser, which has a typical pulse length greater than 50-psec. Harmonics have also been used to measure the radiative lifetime of the 1s2p ^1P state of Helium.[52] It is also extremely encouraging to note that high harmonic radiation is now being used as a tool in condensed matter physics, with studies of antibonding states on the Ge(111):As surface being reported,[53] and more recently the application to high resolution atomic core level spectroscopy, which can be used to study chemistry at surfaces.[54]

At this early stage, it is not clear what additional specific applications such sources may have. However, for harmonics in particular, the cost of the necessary optical laser systems required to generated such high brightness XUV radiation has fallen dramatically over the past few years, so that such sources can truly be described as table-top systems of moderate cost. This wider availability will aid in the realisation of the potential of these high brightness XUV sources.

7 Conclusions

In summary, we have discussed recent improvements in the efficiency of x-ray lasers by use of the prepulse technique. The conversion of laser light into high energy harmonics has been presented, with specific results of harmonic generation from ions using a KrF laser given. Finally, harmonic generation from the interaction of a short intense pulse with a ponderomotively-steepened plasma has been demonstrated.

The spectral brightness of these three distinct types of source has been compared. At present, x-ray lasers are the brightest source in the XUV, followed by harmonics from ponderomotively-steepened plasmas. The latter source having the advantage of some degree of tunability. Harmonics from gaseous targets are also of import, as they can be generated using table-top equipment, and such sources are starting to find application in other areas of research, such as condensed matter physics. We believe that these short pulse, bright XUV sources will become a useful, complementary source to more conventional sources such as synchrotrons.

Acknowledgments

Much of the work described in this article was jointly funded by the United Kingdom Engineering and Physical Sciences Research Council and the European Communities Large Facilities Access Programme. The authors gratefully acknowledge the support of laser operations, target preparation and engineering support groups of the Central Laser Facility of the Rutherford Appleton Laboratory.

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Coherent High-Harmonic XUV radiation

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University of Oxford.

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Presented at the Trieste College,
March 1997

Introduction to 3 Lectures

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- In the last few years it has been shown that high brightness XUV radiation can be generated by making very high harmonics of an intense, shortpulse (generally sub-picosecond) laser. By this method optical light can be directly converted into XUV radiation with wavelengths of a few tens of angstroms. In these lectures I will briefly discuss
- (i) The short pulse lasers (very briefly!)
- (ii) Harmonics from gaseous targets
- (iii) Harmonics from solid, or ponderomotively-steepend plasmas.
- Experimental results and very simple theory will be outlined for the two methods of generating harmonics.

Motivations for producing high order harmonics

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- 101st harmonic of 1-μm light is ~100Å - in the XUV.
- Fundamental physics of atoms and ions in intense laser fields.
- Holography of living cells?
- Photoionization cross section measurements (P. Balcou et al, Optics Letters, to be published).
- Photoemission studies of semiconductors (Haight and Peale, Rev. Sci. Instrum. 65, 1853 (1994)).
- Non-linear optics in the XUV
- ???

The "Water Window"

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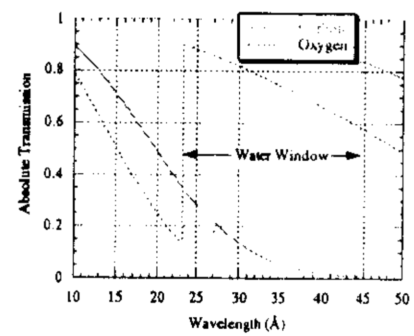
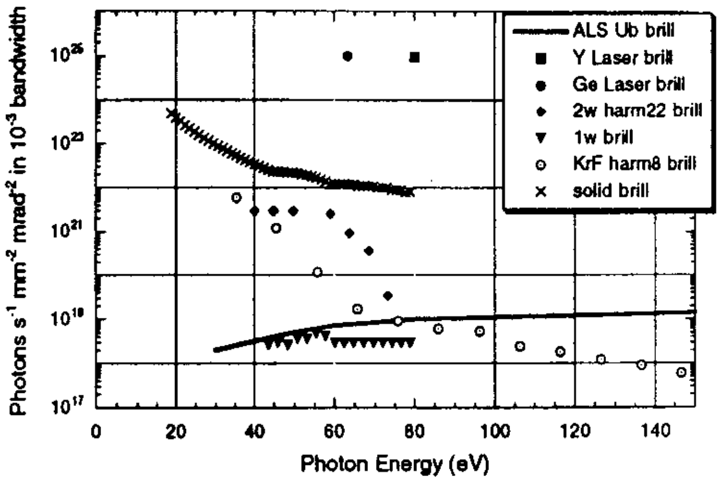


Brightness of various XUV sources



Parameters of harmonic XUV sources

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- At present wavelengths range from optical down to about 67Å for "harmonics from gases" and 150Å for "harmonics from solids"
- Pulse length is close to that of the drive laser - i.e. from tens of femtoseconds to a couple of picoseconds.
- For harmonics from gases, the conversion efficiency is of order 10^{-6} for lowish energy harmonics (355Å) to about 10^{-10} for high energy harmonics. Thus at 355Å we get about 1-μJ of light per harmonic. The divergence of the harmonics is roughly the cone angle of the incident laser. The harmonic bandwidth is of order 10^{-3}.
- For harmonics from "solids", the conversion efficiency is always above about 10^{-6}, giving several mJ of energy per harmonic. For the XUV harmonics produced to date, they diverge into all angles.

Laser Parameters needed for harmonics

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- Both harmonics from "solids" and from gases, require ultrashort laser pulses, typically less than 1 picosecond.
- Harmonics from gases are produced at laser irradiances ranging from about 10^{14} Wcm^{-2} to 10^{17} Wcm^{-2}.
- Harmonics from "solids" are produced at laser irradiances from $10^{17}Wcm^{-2}$ to the present limit, at in excess of 10^{19} Wcm^{-2}.

The Chirped-Pulse-Amplification (CPA) concept

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- The power deliverable by a laser system is generally limited by damage that would occur in the system due to self focussing of the laser beam caused by non-linear effects.
- This problem can be overcome by taking a short (<1psec) seed pulse - stretching it in time to a few hundred picoseconds - amplfying it at a relatively low power - and then recompressing it in time.
- The temporal stretching and compression is generally performed with parallel compression gratings.

CPA has revolutionised high power laser physics

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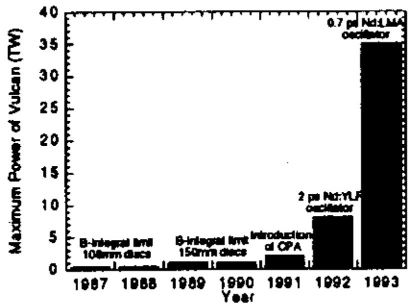

Schematic layout of the Sprite Laser

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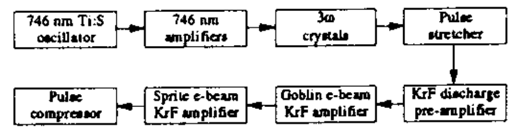

- Note the characteristic CPA configuration of "stretch, amplify, compress"

The Sprite pulse stretcher

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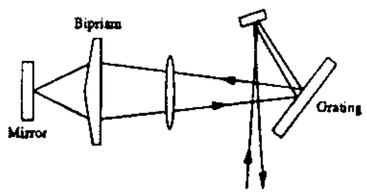

Stretched pulse length:

$$\tau = \frac{2(f-l)(m\sigma)^2 \lambda \cdot \Delta\lambda}{c\cos^2\theta_d}$$

- Note the light diffracts from the grating twice - in doing so it is "chirped".
- After being stretched in time it is amplified before temporal recompression.

The pulse compressor

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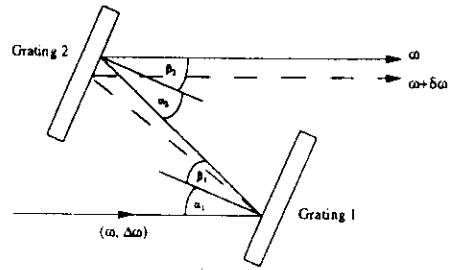

- The pulse is recompressed using a pair of parallel gratings

"Vulcan" target chamber (outside)

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Harmonic Generation

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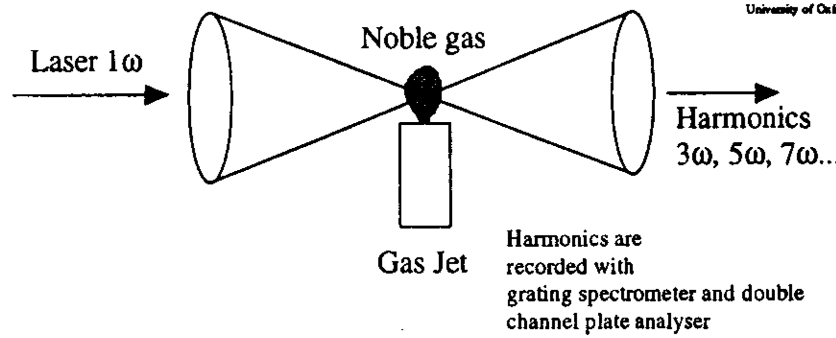

- The thickness of the gas from the gas-jet is chosen so that the fundamental and harmonics do not get too out of phase due to the differing refractive indices. (We cant yet phase match as done for low harmonic generation in crystals).

Observation of 37th harmonic of 248.6nm (67.2Å) in He+

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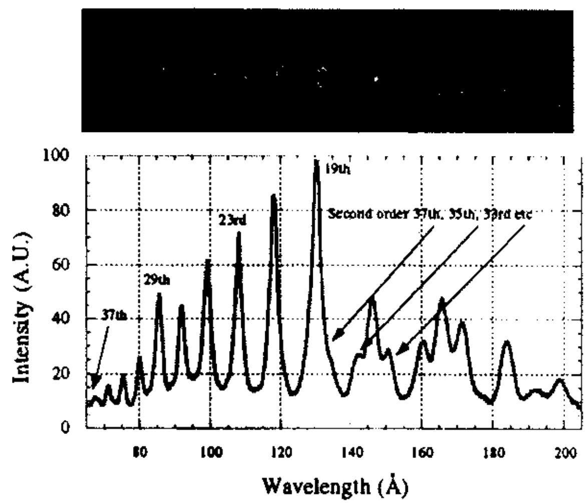

Schematic of Results

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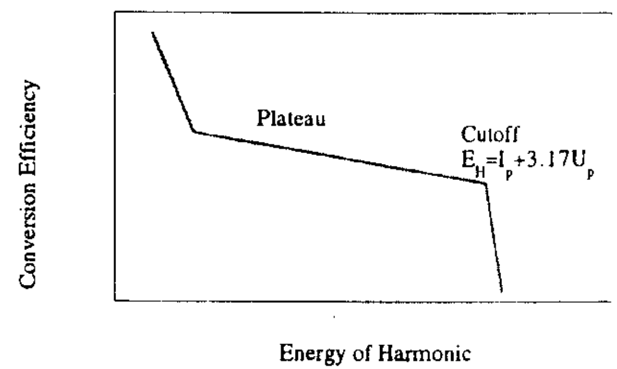

- Early results and time-dependent solutions of Schrödinger's equation indicated that the highest harmonic had an energy of $I_p+3.17U_p$, where I_p is the ionization potential, and U_p the oscillatory or 'quiver' energy of the electron in the laser field.
- $U_p = q^2E_0^2/(4m\omega^2)$, therefore most groups use long wavelength lasers to get high energy harmonics.

Harmonic generation

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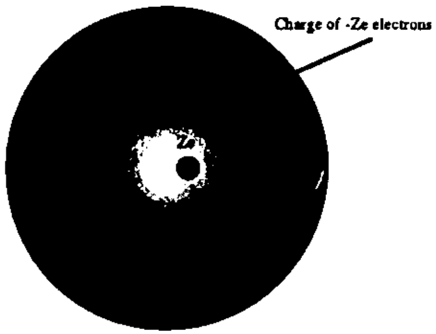

$E = E_L \sin \omega t$

$$P = \varepsilon_0 \chi E = \varepsilon_0 \left\{ \chi_1 E_L \sin \omega t + \chi_2 E_L^2 \sin^2 \omega t + \chi_3 E_L^3 \sin^3 \omega t + ... \right\}$$

$$= \varepsilon_0 \left\{ \chi_1 E_L \sin \omega t + \frac{\chi_2}{2} E_L^2 (1 - \cos 2\omega t) + \frac{\chi_3}{4} E_L^3 (3 \sin \omega t - \sin 3\omega t) + ... \right\}$$

In a centrosymmetric system we only find odd order terms.

How large are the non-linear susceptibilities? (for 'weak' light)

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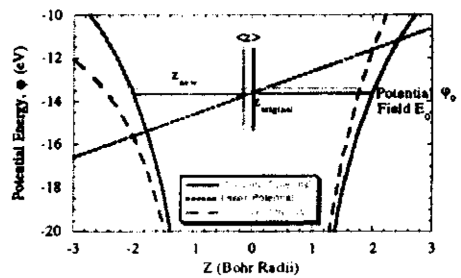

From a simple Taylor expansion we obtain:

$$< z > \approx -\left\{ \left| \frac{dz}{d\phi} \right|_{\phi_0} (E_L a_0) + \frac{1}{3!} \left| \frac{d^3 z}{d\phi^3} \right|_{\phi_0} (E_L a_0)^3 + ... \right\} \approx a_0 \left\{ \left(\frac{E_L}{E_0} \right) + \left(\frac{E_L}{E_0} \right)^3 + \left(\frac{E_L}{E_0} \right)^5 + ... \right\}$$

As $\chi = \frac{N_{atom} e < z >}{\varepsilon_0 E_L}$, $\chi_n \approx \frac{N_{atom} e a_0}{\varepsilon_0 E_0} \left(\frac{E_L}{E_0} \right)^{n-1}$

Breakdown of the perturbative approach for 'strong' light

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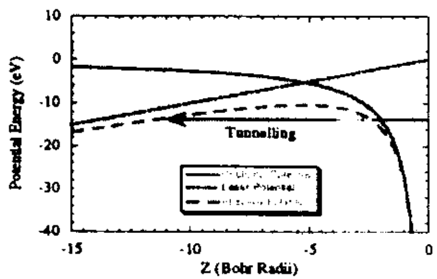

- At these high intensities the simple perturbative approach is no longer valid.
- This is because the electron can quantum mechanically tunnel out of the Coulomb field, and travel far from the atom.
- For a few oscillations the electron is still 'bound' to the nucleus.
- A small change in electric field can produce a VERY large change in polarisation - the process is *highly* non-linear.
- A classical non-linear oscillator driven hard exhibits high harmonics.

Classical non-linear oscillator exhibits harmonics

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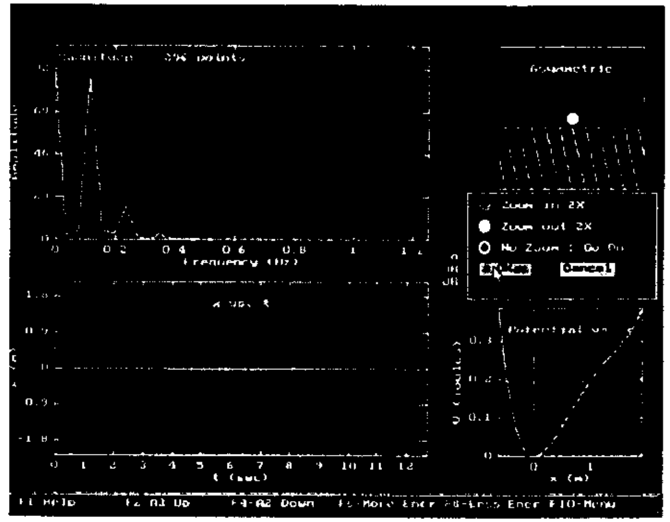

Intensities required

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- The atomic unit of field, E_0, is 5.14×10^{11} Vm^{-1}.
- For the laser field to equal this value we need an irradiance of 3×10^{16} Wcm^{-2}. (simply from the Poynting vector)
- This is equivalent to 10^{12}W (1 TW) in a 30 micron spot.
- 1 TW is about the output of the whole of the national grid!
- We are using some of the most powerful lasers in the world.

Ionization proceeds by the electron tunnelling out of the Coulomb field

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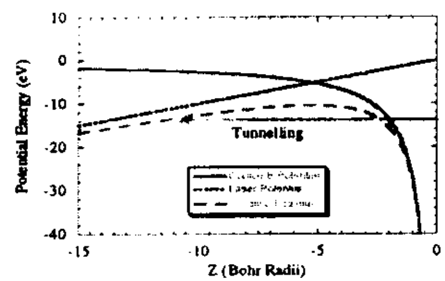

$$R_{Hydrogen} = 4\omega_0 \left(\frac{E_0}{E}\right)^{1/2} \exp\left(-\frac{2E_0}{3E}\right)$$

E is the electric field of the laser

E_0 the atomic unit of field (5.14×10^{11} Vm^{-1})

ω_0 the atomic unit of frequency (4.16×10^{16} s^{-1})

Ionization and harmonic production are linked

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- In a strong laser field the electron can tunnel through the Coulomb barrier.
- As the laser field reverses (as it is oscillating), the electron can return to the core.
- If the electron returns to the ground state, a photon (the harmonic photon) is emitted. However, this is most likely on the first recollision, and the probability drops in time due to wavefunction diffusion.
- If no harmonic is produced, the atom is optically ionised.
- The high non-linearity of the process means we wish the atom to experience the highest possible field - therefore we require short pulse lasers, so that the atom has not completely ionised by the time the field in the pulse reaches its peak.

Ionization rates for Helium

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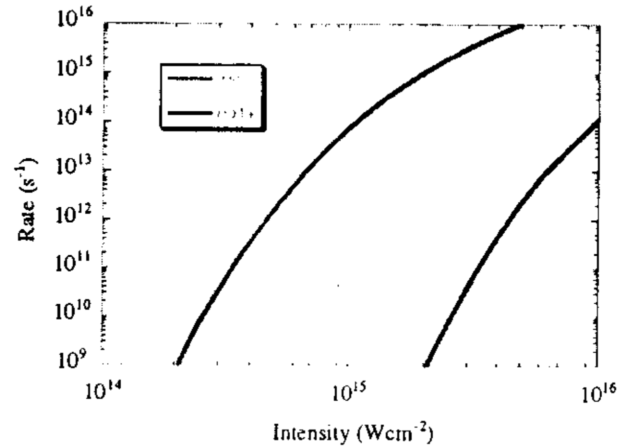

Ionization rates for Neon

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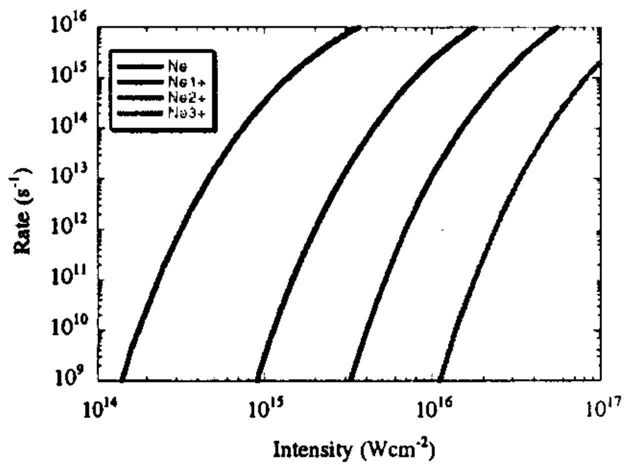

The need for sub-picosecond pulses and gaseous targets

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- Neutral atoms are more efficient than ions at generating harmonics, but the orders of the harmonics they can produce are lower.
- There are two main reasons why we require ultrashort pulses:-.
- (i) We want the atom or ion of interest to experience the highest possible field before being completely ionized (as the process is highly non-linear the strength of the harmonics is still a high power of the field - ~ 5 - 7).
- (ii) We want any ionization that does occur to do so due to these high fields (optical ionization), NOT due to free electrons in the system colliding with atoms and ions - .i.e we want a pulse shorter than an electron-ion collision time. This also sets an upper limit on the material densities we can use (we operate with gas targets).

The electron wavefunction diffuses in time - thus the recollision cross section scales as λ^{-2}

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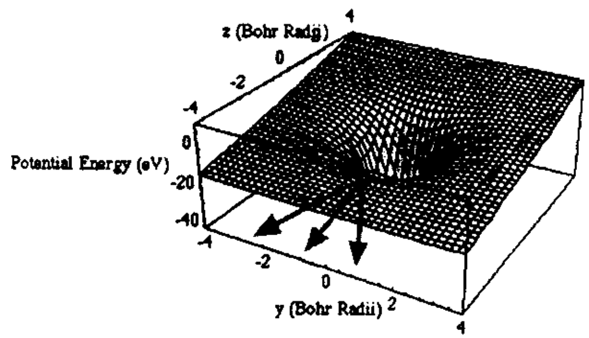

- After tunnelling the wavefunction is localised in the direction perpendicular to the laser field, and diffuses ~linearly in time.

Approaches to understanding harmonics from gases

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- We need to understand how a single atom responds to the laser field, as well as the effects of the medium as a whole.
- Note we cannot solve the problem analytically as we have an extremely non-linear problem far outside of the perturbative limit. In general, we most use a computational approach.
- That said, a very simple semi-classical model presented by Paul Corkum can give good physical insight, and explains many of the salient features seen in both simulations and experiments.

Full simulations

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In atomic units we must solve

$$i\frac{\partial\Psi}{\partial t} = -\frac{1}{2}\frac{\partial^2\Psi}{\partial x^2} + V(x)\Psi + E_0 x\sin(\omega t)$$

- Most simulations use the single-active-electron (SAE) approach - one electron moves in a potential representing the nucleus, the rest of the electrons, and the time-dependent laser field. Solved computationally using the Crank-Nicholson method.
- The harmonics are calculated by taking the fourier transform of the dipole moment.
- Simulations can be performed in both 1 and 3 dimensions.

Typical Simulation Output

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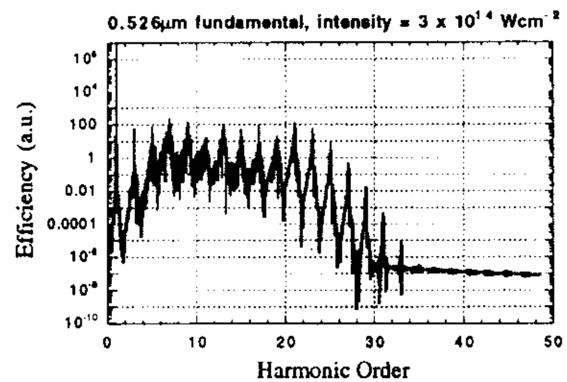

The plateau increases with irradiance

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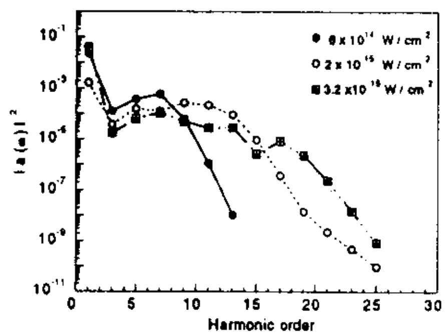

The "cutoff" is found to occur at $I_p + 3.17U$. Where I_p is the ionization potential and U is the quiver energy of the electron in the laser field

The "quiver" or ponderomotive energy

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$$\frac{d^2z}{dt^2} = \frac{qE_0Cos(\omega t)}{m}$$

$$\frac{dz}{dt} = v = \frac{qE_0Sin(\omega t)}{m\omega}$$

$$\therefore U = \frac{m\bar{v}^2}{2} = \frac{q^2E_0^2}{4m\omega^2}$$

The Keldysh Parameter

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The Keldysh parameter for an atom of binding energy I_p in a field of ponderomotive energy U is defined as:-

$$\gamma = \left[\frac{I_p}{2U}\right]^{1/2}$$

For a binding energy I_p, in a field E_0, the electron must tunnel through a barrier of width order:-

$$l = I_p / qE_0$$

with a velocity of order:- $v = \sqrt{\frac{2I_p}{m}}$, i.e. it tunnels through in a time:-

$$\tau = \frac{l}{v} = \frac{\sqrt{m}}{qE_0}\sqrt{\frac{I_p}{2}}$$

which we can compare to the laser frequency by writing:-

$$\omega\tau = \frac{\omega\sqrt{m}}{qE_0}\sqrt{\frac{I_p}{2}} = \frac{1}{2}\gamma$$

i.e. the tunnelling picture is valid at high intensities.

Harmonics as a function of wavelength of the primary radiation

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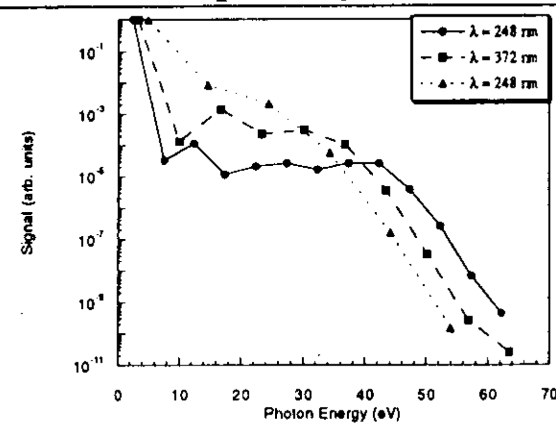

- Shorter wavelength drivers are more efficient, but seem to cut off at a lower energy harmonic for a given laser intensity

Summary of Lecture 1

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- CPA lasers have made possible laser powers hitherto unaccessible. This is achieved by stretching a pulse in time, amplifying it, then recompressing it by means of parallel diffraction gratings.
- XUV harmonics can be generated by interactions of these laser pulses with gases or solids. The mechanism of production is totally different for the two cases.
- The brightness of the harmonics is greater than the output of a synchrotron but, as yet, not as bright as an XUV laser.
- Examples of full simulations have been given for harmonics from gases - next lecture we will consider a simple semi-classical model that explains a lot of the characteristic behaviour.

A simple semi classical model

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- A simple semi-classical approach can explain *some* of the physics and provide rough scaling laws.
- Calculate instantaneous ionization rate at a given phase in the sinusoidal laser field using simple QM tunnelling theory.
- Assume that at this phase a free electron is born at zero velocity at $z=0$.
- Calculate the subsequent motion of the particle classically.
- Calculate the energy of the particle on recollision with the core.
- A detailed prediction of spectra is only possible through calculating the Fourier transform of the dipole moment from Schrödinger's equation.

Due to Paul Corkum - Phys Rev Lett 71, 1995, 1993

Equations of motion of a free electron (1)

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$$\frac{d^2z}{dt^2} = \frac{qE_0 Cos(\omega t)}{m}$$

$$\frac{dz}{dt} = v = \frac{qE_0 Sin(\omega t)}{m\omega} + v_0$$

$$z = -\frac{qE_0 Cos(\omega t)}{m\omega^2} + v_0 t + z_0$$

Equations of motion of a free electron (2)

Clarendon Laboratory, University of Oxford

At $t = t_0$ let $z = 0$, $v = 0$ then

$$v = \frac{qE_0}{m\omega}\{Sin(\omega t) - Sin(\omega t_0)\}$$

$$z = -\frac{qE_0}{m\omega^2}\{Cos(\omega t) + \omega t Sin(\omega t_0) - Cos(\omega t_0) - \omega t_0 Sin(\omega t_0)\}$$

Solve for v when the electron returns to the core ($z = 0$). The energy at impact is $mv^2/2$.

Ionization probability and energy at first recollision as a function of phase angle

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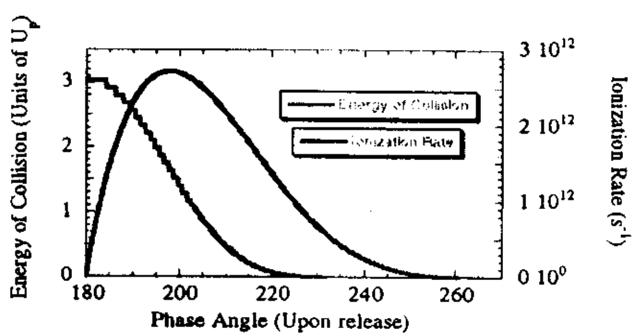

- The peak energy at recollision is 3.17 U_p at a phase angle of 197^0.
- The tunnelling rate has been calculated assuming an intensity of 10^{14}Wcm^{-2}.

The wavefunction diffuses

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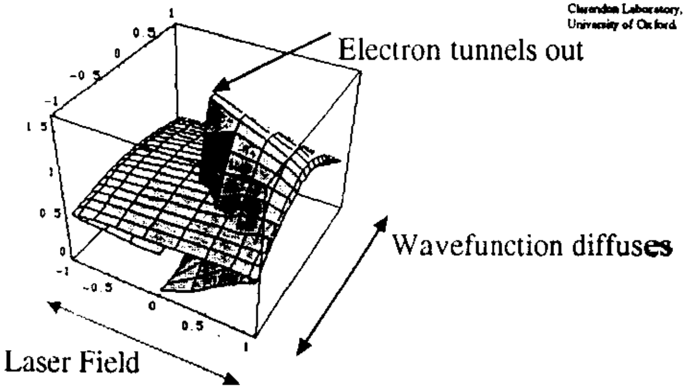

- For a few oscillations there is a probability of returning to the ground state and emitting harmonics - after this time we can think of the electron as being ionized.

Diffusion of the wavefunction

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- The free electron wavefunction diffuses approximately linearly in time in the directions perpendicular to the laser field.
- It takes of order half a laser period for a recollision, thus the cross section for harmonic generation scales roughly as λ^{-2}.
- The original wavefunction is confined to a distance of order $a_0/2$, and thus diffuses at a rate of roughly 2Å / fsec.
- For 248-nm light half a laser period is about 0.4-fsec, therefore the wavefunction approximately triples in radius upon recollision.

Wavelength Dependence of Harmonic Generation (1)

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- Conventional wisdom says that long-wavelength lasers produce the highest energy harmonics.
- This is because the cut-off has been shown to occur at $I_p + 3.17U_p$, where I_p is the ionization potential, and U_p the oscillatory or 'quiver' energy of the electron in the laser field.
- $U_p = q^2E_0^2/(4m\omega^2)$, therefore most groups use long wavelength lasers to get high energy harmonics.

Wavelength Dependence of Harmonic Generation (2)

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- However, why dont we utilise the higher I_p of ions, rather than neutrals, to produce high energy harmonics?
- This is difficult with long wavelength lasers due to both the dephasing effect of free electrons, and a significantly lower conversion efficiency.
- These problems are not so severe for short wavelength lasers.
- Can a short wavelength (248-nm) laser produce higher energy harmonics from ions than a long wavelength (800 - 1053-nm) laser can from neutrals?

A schematic of the predictions

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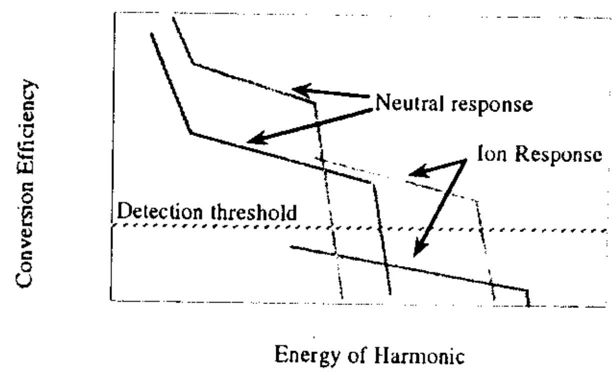

- Could higher energy harmonics be produced using a short wavelength laser operating with ions than from a long wavelength laser operating with neutrals?

Harmonic Generation and Plasmas (2)

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- The efficiency of harmonic production is limited by the fact that the fundamental of the laser and the harmonics get out of phase with each other due to the wavelength dependent refractive index of the plasma.

The phase mismatch is due to the plasma refractive index

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$$\mu = \sqrt{1 - \frac{n_e e^2 \lambda_0^2}{\varepsilon_0 m 4\pi^2 c^2}}$$

$$\mu(q\lambda_0) \approx 1$$

$$\mu(\lambda_0) \approx 1 - \frac{n_e e^2 \lambda_0^2}{\varepsilon_0 m 8\pi^2 c^2}$$

$$\therefore \mu(q\lambda_0) - \mu(\lambda_0) \propto \lambda_0^2$$

$\therefore$ Coherence length scales as $1/\lambda_0^2$

Intensity scales as $1/\lambda_0^4$

Laser Parameters

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- SPRITE laser at the Central Laser Facility of RAL.
- KrF - wavelength of 248.6-nm.
- 350-fsec pulselength.
- 150-mJ per pulse.
- 8-cm diameter beam, f/10 off-axis parabolic reflecting optics.
- ~6 X diffraction limit.
- Peak focused intensity of 10^{17} Wcm^{-2}.

Schematic of Experimental Set-up

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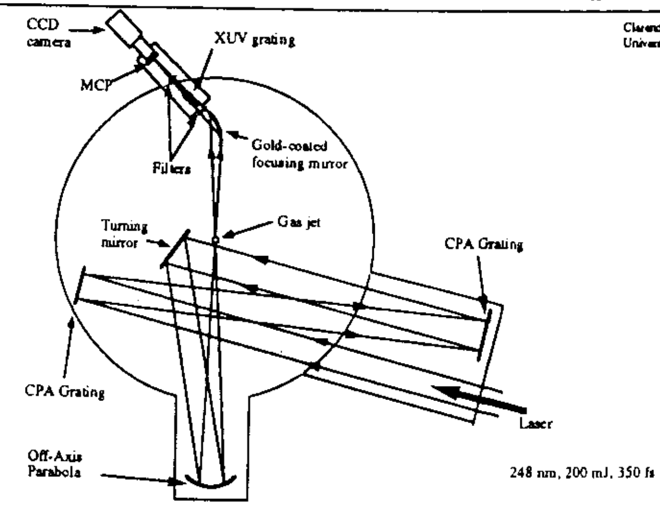

- Target chamber lay-out on the Sprite KrF laser at the Rutherford Appleton Laboratory

"Sprite" Target Chamber (inside)

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The XUV spectrometer (Plan view)

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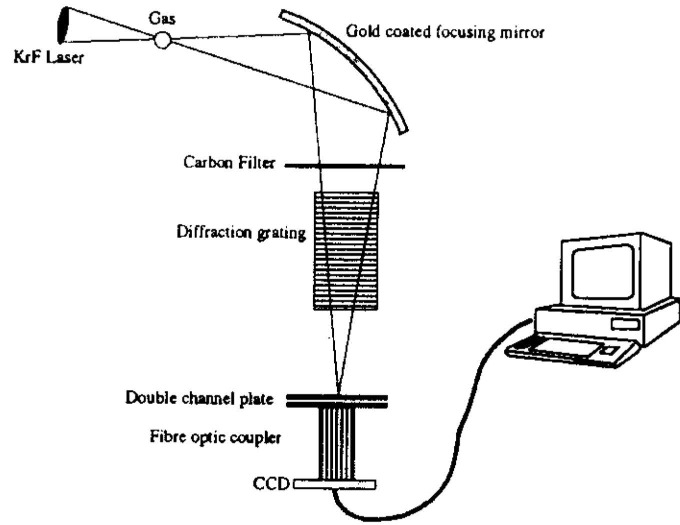
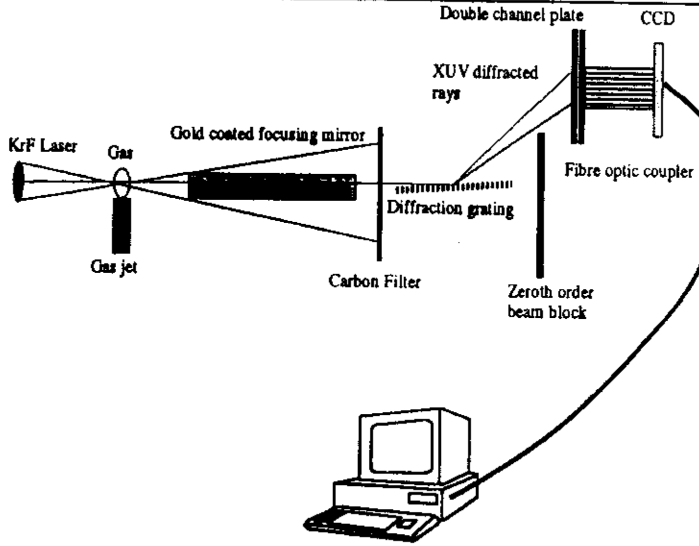


The XUV spectrometer (Side view)



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Observation of 37th harmonic of 248.6nm (67.2Å) in He+

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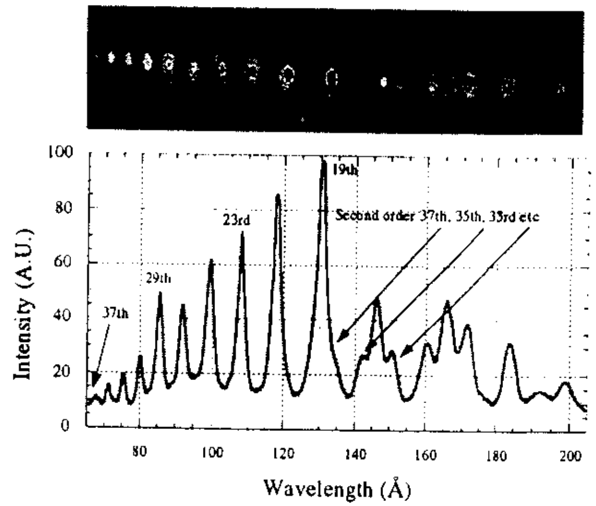

33rd harmonic in Neon - evidence of response of doubly-ionized Neon

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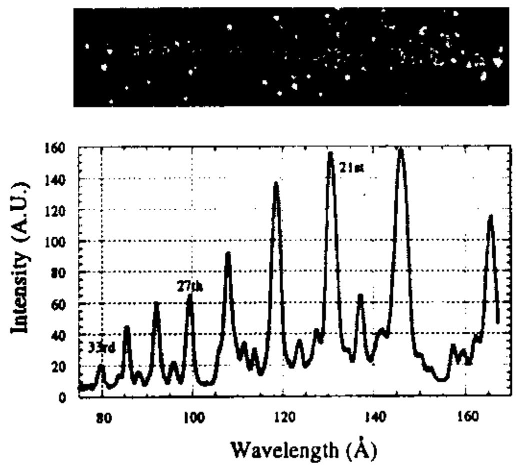

Conversion efficiencies - Helium

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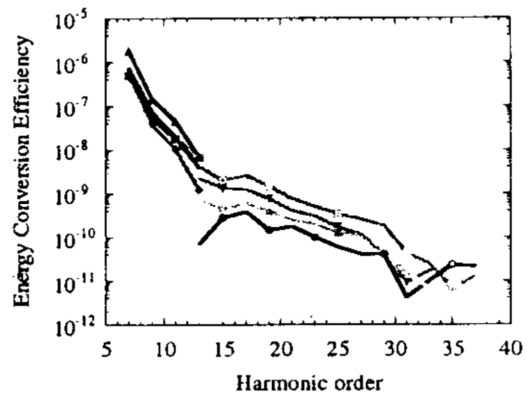

- A preliminary calculation of conversion efficiencies shows that we have a peak power of order 1-MW in 7th harmonic (355Å).

Conversion efficiencies - Neon

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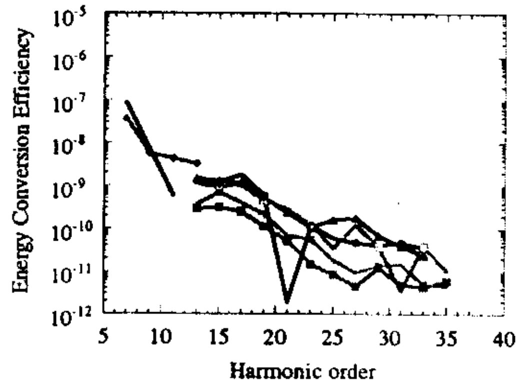

Predictions of the theoretical model

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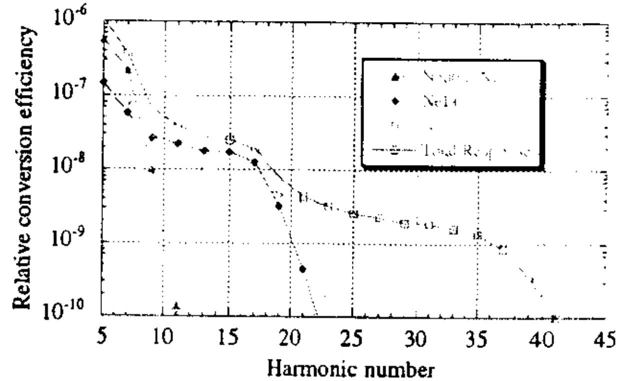

- Harmonics above 21st are due to doubly ionized Neon.

Why generate high harmonics from plasmas using KrF lasers?

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- Ions can generate shorter wavelength harmonics, as the maximum possible photon energy scales as $h\nu_{max} = Ip + 3.17Up$
(where Ip is the ionisation potential and Up the ponderomotive potential)
- The short optical cycle of KrF lasers leads to less wavepacket dilation
-> Recombination probability scales as $1/\lambda^2$
-> much stronger ionic response
- Smaller phase mismatch through refractive index between UV lasers and harmonics -> greater efficiency

Simulated single atom harmonic efficiency as a function of irradiance

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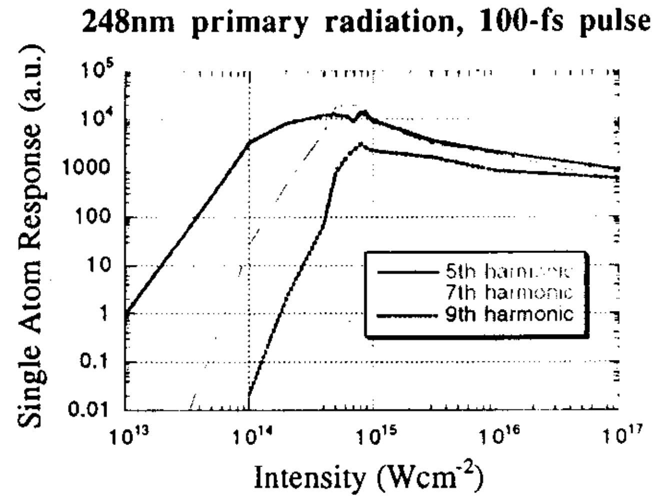

Simulated population of neutrals at peak of pulse

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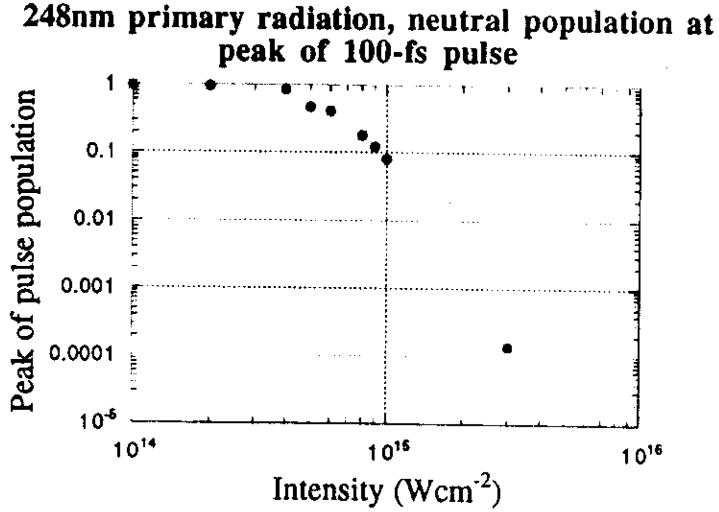

Ionisation at harmonic production time is constant

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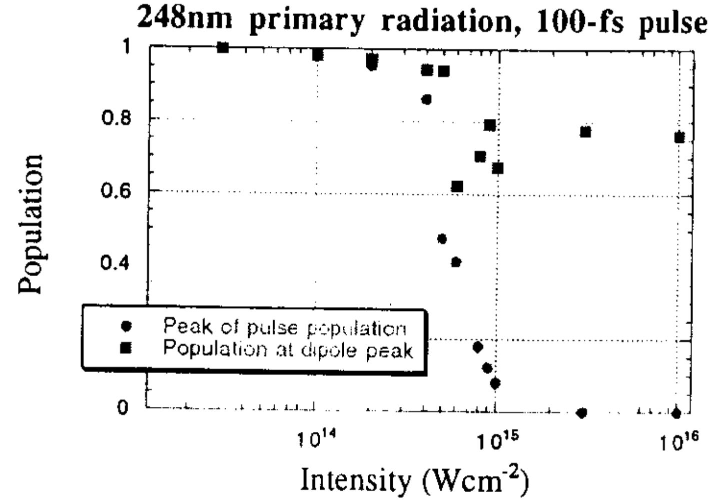

There is an optimum intensity for production of a given harmonic

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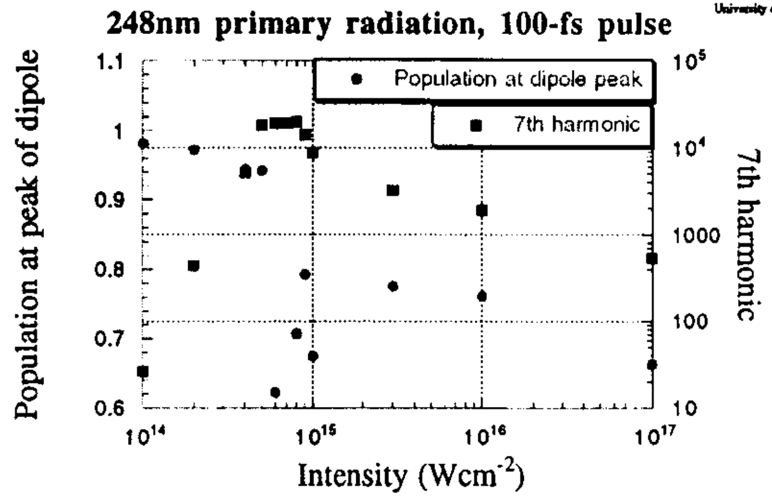

Experimental verification of optimum intensity

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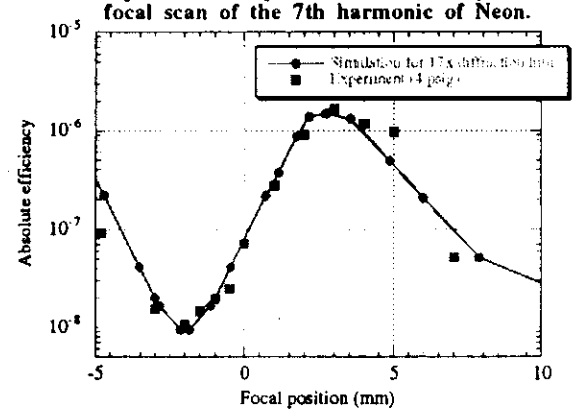

- The theoretical graph has been obtained by weighting the single atom response with the area of the beam, and allowing for the coherence length due to free electrons

Summary of harmonics from gases section

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University of Oxford

- Harmonic generation from gases can be understood in terms of a very simple semi-classical model.
- This model explains the presence of the cutoff, and predicts the correct cutoff energy and wavelength scaling.
- Short wavelength primary lasers produce harmonics more efficiently because of single atom effects (the wavefunction does not have so much time to diffuse after tunneling through the coulomb barrier) and because the harmonics take longer to get out of phase with the laser.

Generation of Harmonics from Solid Targets

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- High order harmonics can also be generated from high intensity laser-solid interactions
- Recent experiments using 2ps, 1μm light show very high conversion efficiencies ($>10^{-6}$@200Å), which scale strongly with the irradiance $I\lambda^2$ and orders upto 75th (140Å)
- Comparison with previous work with CO_2 lasers (10.6 μm) and Ti:Sapphire lasers (0.8 μm) indicate that the conversion efficiency into each order depends only on $I\lambda^2$.
- Scaling the results at 1μm to KrF suggests generation of harmonics in water window region of the spectrum (44-20Å) with conversion efficiencies $>10^{-6}$ for $I=1.5\times10^{20}$ W/cm^2

Schematic diagram of experiment

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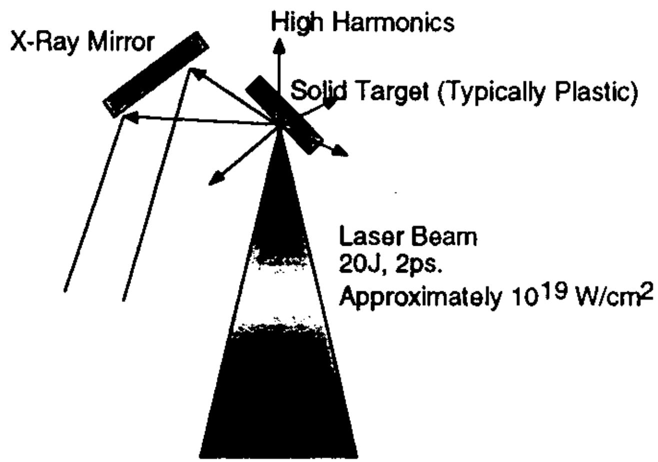

Example of the Raw data

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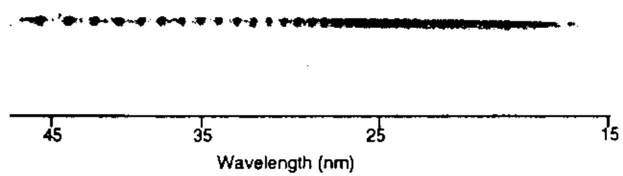

Deconvolved Lineout of a Spectrum

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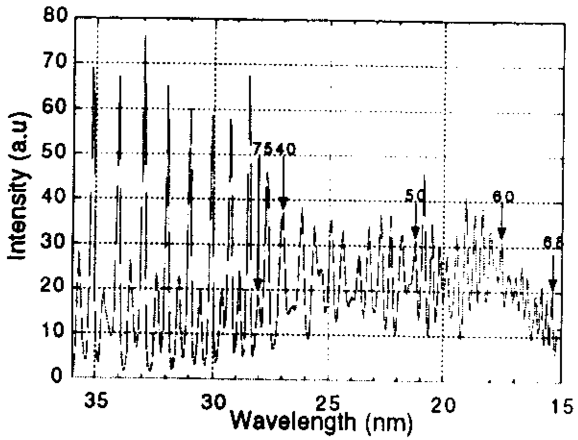

Data deconvolved using a maximum entropy routine.
Upto 75th harmonic is visible in 2nd order. The first order range was limited by the detector position. The 2nd orders were verified by filtering

Intensity Dependence of Harmonic Spectra

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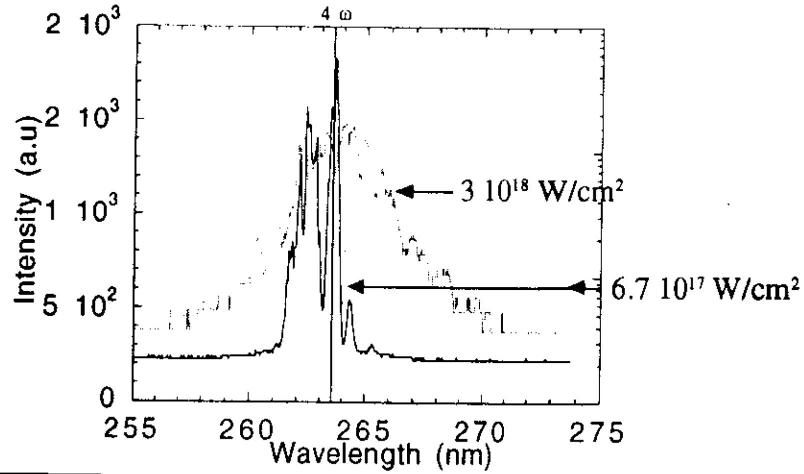

Exponent vs Irradiance

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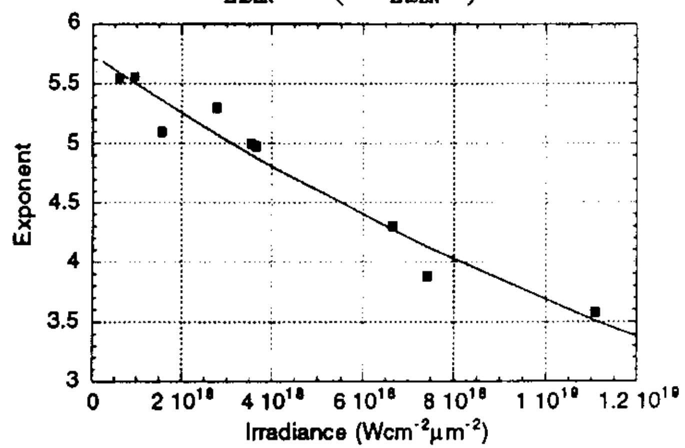
$$\frac{E_{HARMONIC}}{E_{LASER}} = \left(\frac{\omega_{HARMONIC}}{\omega_{LASER}}\right)^{-X}$$



Approaches to understanding harmonics from solids

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- Relativistic PIC codes - these are computer simulations that solve the relativistic equations of motion of electrons with self-consistent electric and magnetic fields.
- Simple analytic models. There are two simple models that help us understand these harmonics - the "trajectory model", and the moving mirror model.

Summary of Lecture 2

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- Harmonics from gases can be understood in a simple way from the semi-classical model of Corkum.
- At much higher intensities we see that we get both odd and even harmonics from interactions with solids, though we have not yet discussed mechanisms
- Next Lecture we will discuss simple models of the mechanism of harmonic generation from laser-solid interactions.

Dependence of conversion efficiency on $I\lambda^2$

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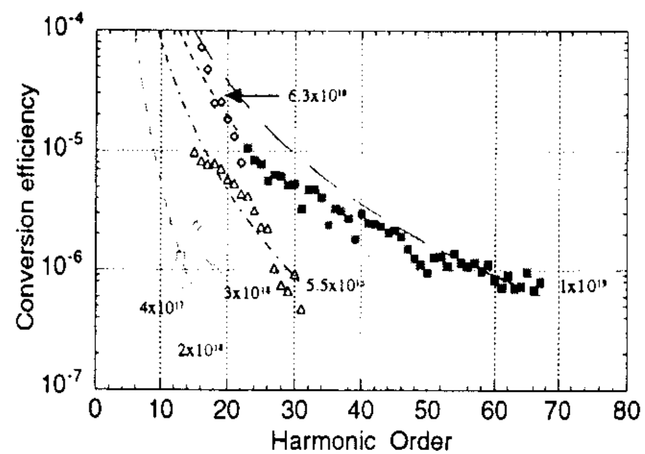

The PIC-code model

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- Solve the self-consistent equations of motion of a set of charged particles.
- The charges move in a combination of the laser field, and the field due to all the other charges.
- Generally we use a spatial scale (grid) that smooths out fine scalelength microfields - e.g. of order an electron Debye length.
- For a description of a simple PIC code see Kruer's book "The Physics of Laser-Plasma Interactions", Addison-Wesley, 1988.

Gibbon's PIC-code predictions - Trajectories

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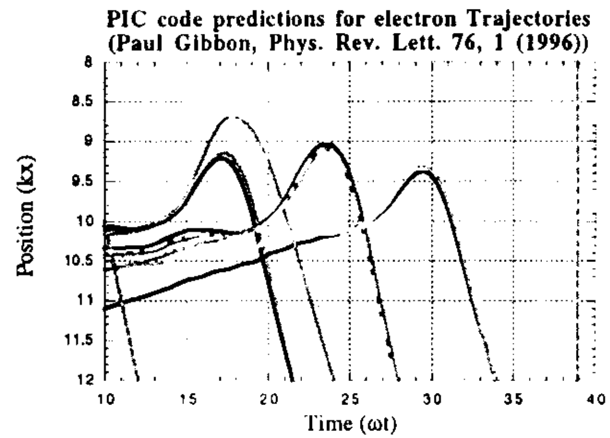
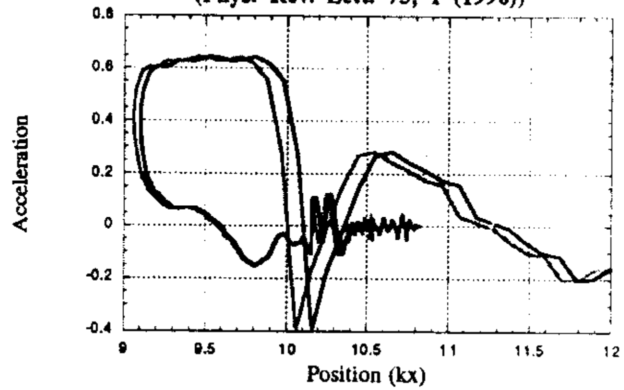


Gibbon's PIC-code predictions - Acceleration

Clarendon Laboratory, University of Oxford.

Results from Paul Gibbon's Pic code (Phys. Rev. Lett. 75, 1 (1996))



Simple Trajectory Model - "Theory"

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Let the "plasma"-vacuum surface be at z=0.

For z>0:-

$$F = m\frac{d^2z}{dt^2} = qE_0\cos(\omega t)$$

For z<0:-

$$F = m\frac{d^2z}{dt^2} = qE_0\cos(\omega t)\exp(kz)$$

Simple Trajectory Model - Trajectories

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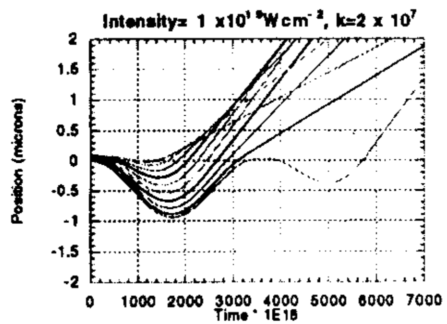

Simple Trajectory Model - Acceleration

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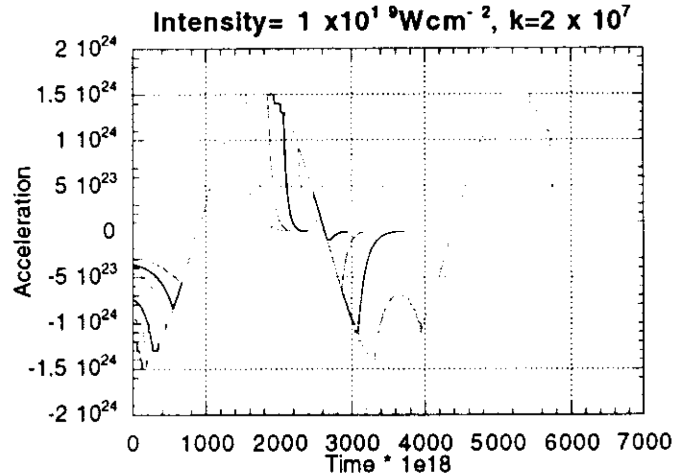

Simple Trajectory Model - Efficiency

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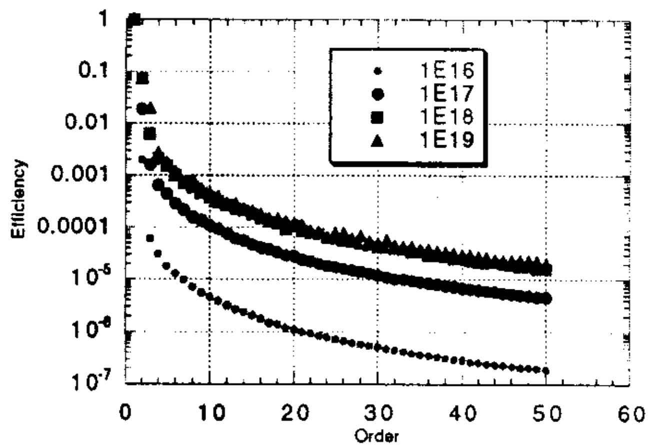

Ponderomotive force

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$$\frac{d^2z}{dt^2} = \frac{qE_0 Cos(\omega t)}{m}$$

$$\frac{dz}{dt} = v = \frac{qE_0 Sin(\omega t)}{m\omega}$$

$$\therefore U = \frac{m\bar{v}^2}{2} = \frac{q^2 E_0^2}{4m\omega^2}$$

$$F_{pond} = -\nabla U = -\frac{q^2}{4m\omega^2}\nabla\left(E_0^2\right)$$

Electrons are expelled from regions of high intensity towards regions of low laser intensity by the ponderomotive force.

Moving mirror model (1)

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University of Oxford.

Consider a mirror oscillating in the z-direction at a frequency ω_m:

$$s(t) = s_0 \sin(\omega_m t)$$

If, for the sake of simplicity, we neglect retardation effects, a beam of frequency ω_0, with angle of incidence θ reflected from the mirror will undergo a phase shift compared to a stationary mirror of:

$$\phi(t) = \left(\frac{2\omega_0 s_0}{c}\right)\cos(\theta)\sin(\omega_m t)$$

R. Lichters et al, Phys. Plasmas, in the press

Moving mirror model (2)

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University of Oxford.

Therefore the reflected beam will have an electric field that varies as:-

$$E(t) = E_0\left[\cos(\omega_0 t + \phi(t))\right]$$

$$\therefore E(t) = E_0 \cos\left[\omega_0 t + \left(\frac{2\omega_0 s_0}{c}\right)\cos(\theta)\sin(\omega_m t)\right]$$

$$E(t) = E_0 \cos\left[\omega_0 t + b\sin(\omega_m t)\right]$$

Orbit of electron in polarized plane wave (1)

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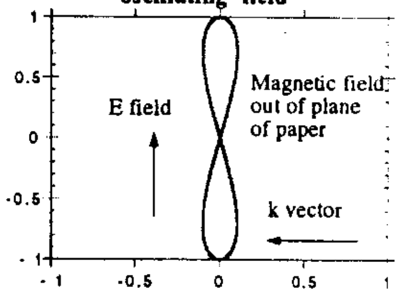
- It is well known that during one fundamental cycle of an oscillating field the electron undergoes a "figure eight" motion in the plane containing the electric field vector and the wavevector.



Orbit of electron in polarized plane wave (2)

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- Note that parallel to the wavevector (longitudinal component) there is a component oscillating at twice the fundamental, whereas parallel to the electric field (transverse compnent), the electron oscillates at just the fundamental.
- Recall that for weak fields the longitudinal component is small - the transverse component dominates.
- The relative strength of the components depends on the relativistic parameter, which is a function of the amplitude of the vector potential:-

$$a_0 = \frac{eA_0}{mc^2}$$

Reflected Light in Moving Mirror Model (1)

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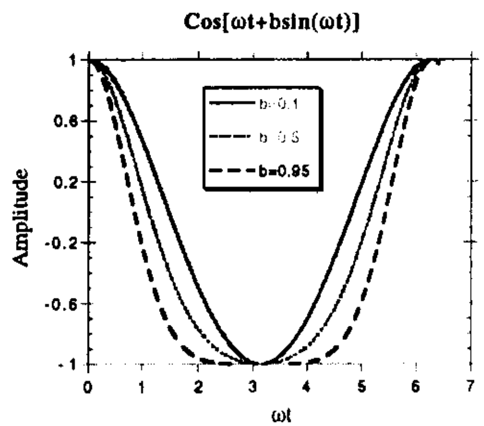

Reflected Light in Moving Mirror Model (2)

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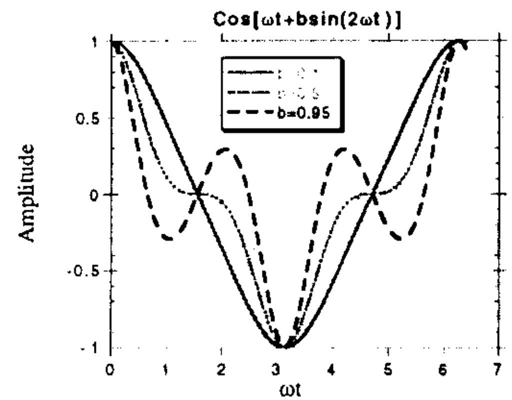

Excursion amplitude of the surface

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- In the non-relativistic regime the excursion amplitude is simple to derive.

$$\frac{d^2z}{dt^2} = \frac{qE_0 Cos(\omega t)}{m}$$

$$\frac{dz}{dt} = v = \frac{qE_0 Sin(\omega t)}{m\omega}$$

$$z = \frac{-qE_0 Cos(\omega t)}{m\omega^2}$$

Relativistic parameter

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- The maximum amplitude of the electron is obviously one laser wavelength (when driven at the fundamental frequency). This occurs when it is moving at close to the speed of light.
- The laser intensity for such relativistic motion can be calculated by requiring the ponderomotive, or quiver energy, to be equal to the rest mass energy of the electron.

$$\frac{e^2 E_0^2}{4m\omega^2} = mc^2$$

This occurs at an irradiance of approximately $I\lambda^2 > 10^{18} Wcm^{-2}\,\mu m^2$

Selection rules - p polarized light

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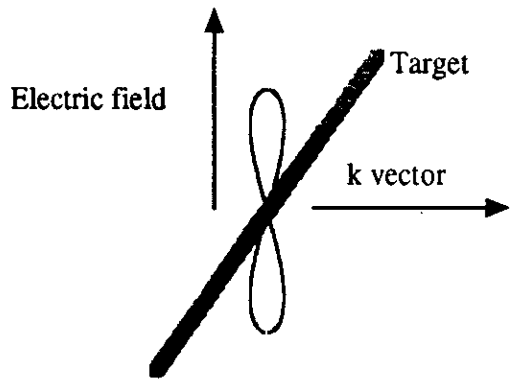

- The electron boundary is driven at both ω and 2ω - both odd and even harmonics are produced.

Selection rules - s polarized light

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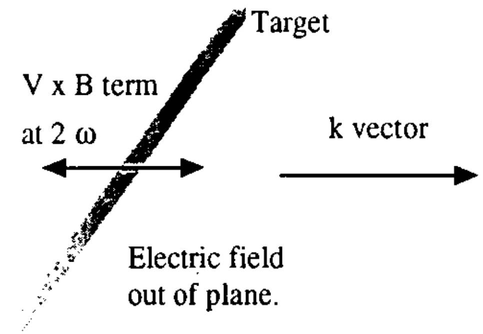

- The electron boundary is driven at 2w - only odd harmonics are produced.

Dipole sheet radiation

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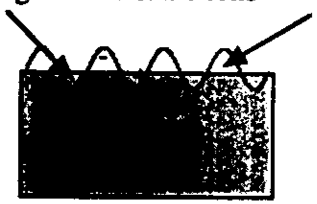

- s polarized light drives the dipole sheet at 2ω. It is obvious that the induced dipoles radiate p-polarised even harmonics.

Final selection rules

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	s-pol harmonics	p-pol harmonics
s-pol fundamental	odd	even
p-pol fundamental	forbidden	odd&even

Lichter's PIC code

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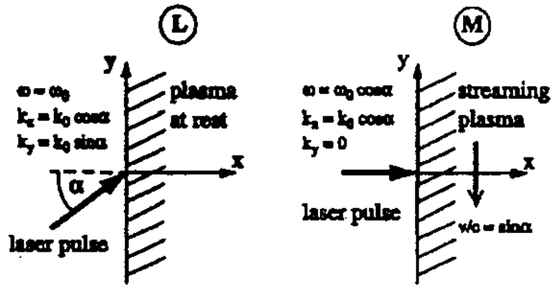

Lichter's results for the shape of the critical surface

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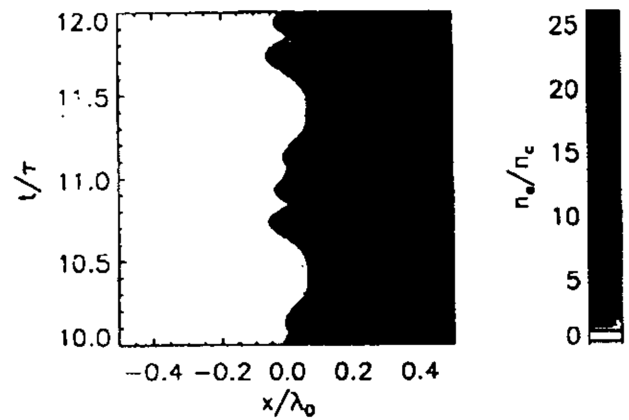

Relativistic parameter = 1, upper shelf density = 4 x critical

Lichter's PIC code harmonic predictions + modified moving mirror model

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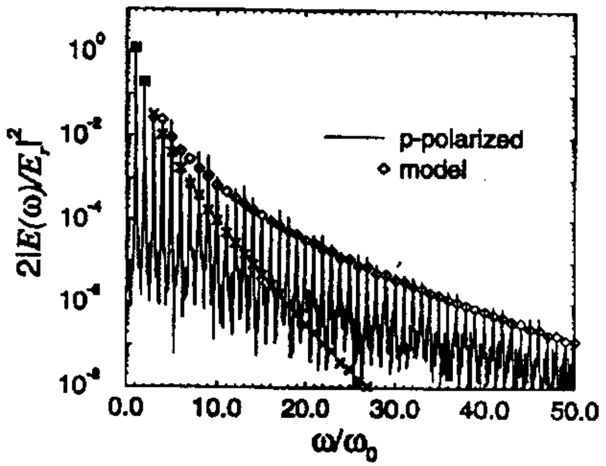

Comparison between s and p polarised light

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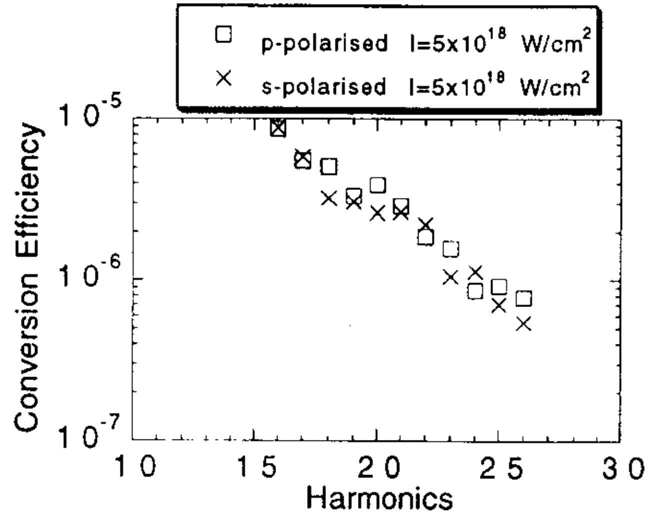

High Resolution of Harmonic Spectrum

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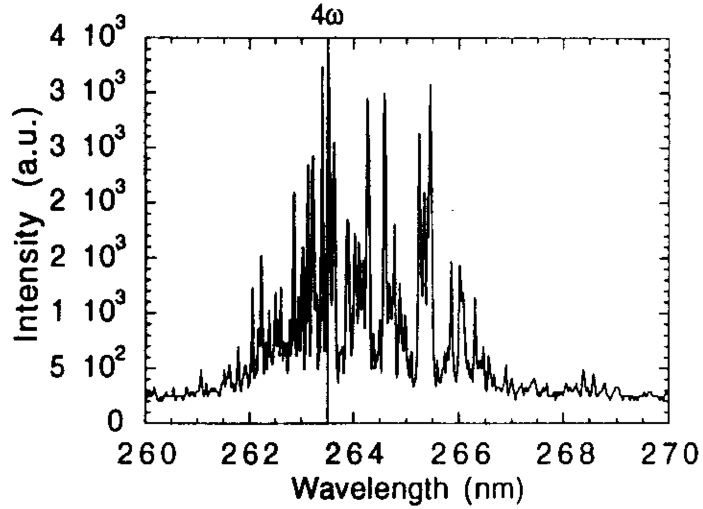

Comparison with and without Prepulse

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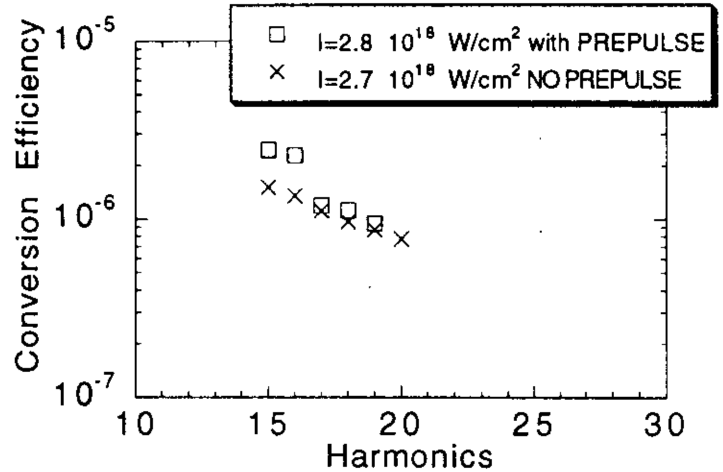

So what's going on?

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- The PIC code and moving mirror model assume a "clean" density step.
- In reality, in our experiments a plasma is formed before the main pulse, but a density step forms as a result of the ponderomotive force.
- The ponderomotive force is pushing a dense plasma - therefore the interface is unstable to a "Rayleigh-Taylor like instability.
- The emitting surface is therefore fragmented, and s and p polarisations lose their meaning.
- The source loses coherence - do the harmonics are emitted in all directions in the presence of this instability.

Imaging of the harmonic source

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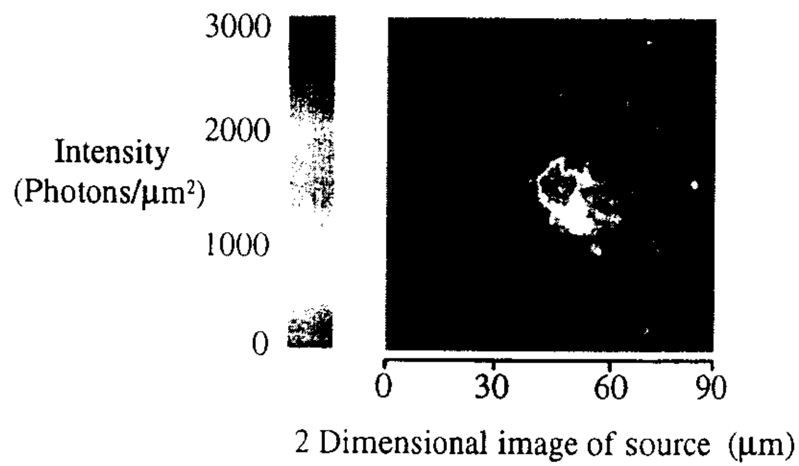

Holeboring diagnosed with harmonics

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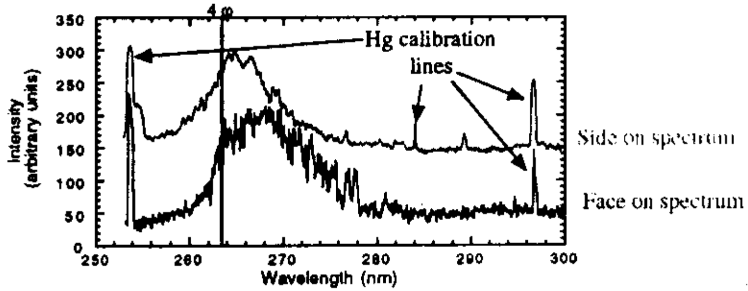
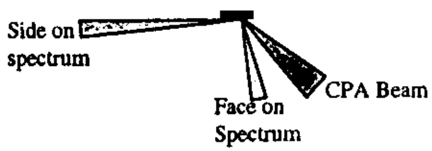


Experimental Geometry



Holeboring Velocities

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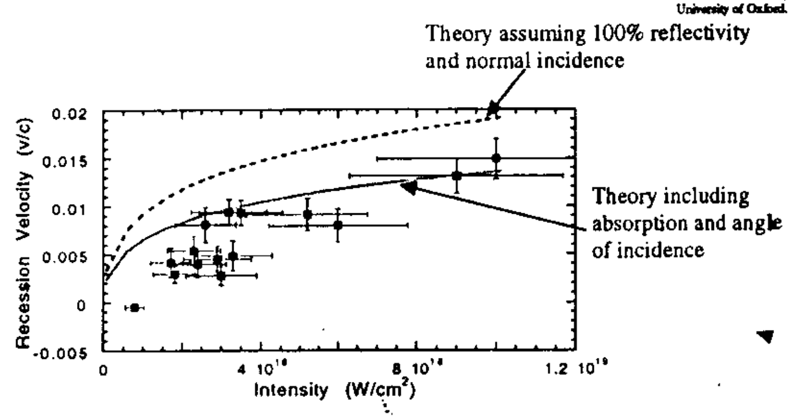

Which model is correct ?

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- It is interesting to question which is the better model - the moving mirror model, or the electron trajectory model.
- Perhaps they are two different ways of looking at the same phenomenon.
- Work is ongoing in this area

Summary

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- Harmonics from solids produce a very bright source of XUV radiation.
- At present, only one experiment has seen such high harmonics, but more will inevitably follow.

Future Work

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- Water window harmonics from solids using KrF lasers.
- What is the ultimate efficiency of harmonics from solids?
- Can we increase the brightness by getting specular reflection whilst still using high intensity lasers?
- Can we phase match harmonics in gases?
- Applications.