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*Author(s):*GEORGE KYRALA, LOS ALAMOS NATIONAL LABORATORY
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MULTIPHOTON PHYSICS WITH X-RAYS

G. A. KYRALA
Los Alamos National Laboratory
Los Alamos, NM 87544

1. Abstract

With the generation of intense bursts of short wavelength radiation during the interaction of ultra-intense laser beams with solid targets, the possibility of extending the study of the non-linear interactions of radiation with matter to shorter and shorter wavelength radiation becomes possible. One would hope that an extension of the techniques to the x-ray region would lead to new tools that are similar to those used in the visible and that can be widely applied. However, a basic knowledge of the relevant efficiencies of the properties of the materials, under these unusual conditions, is necessary before one can build such sources and devices. In this work we discuss the design of the first experiments on non-linear interactions using x-rays in the kilovolt energy range, and present some of the some of the early findings.

2. Introduction

The interaction of radiation with matter has been of great interest and relevance to atomic physics since the beginning of atomic theory. Before the invention of lasers, studies concentrated on the linear interactions of the radiation with material.[1] Since the invention of lasers, intense visible-wavelength radiation has become available and has allowed the study of nonlinear interactions of radiation with matter. The interactions have allowed, among other possibilities, the generation of harmonics of the incident radiation at much shorter frequencies, as well as the generation of characteristic radiation from the plasmas that are created by the interactions with solids. In both cases the generated radiation is short, of the order of the laser irradiation time itself. Using non-linear interactions for the generation of novel sources of tunable radiation has caused an explosion in the knowledge of atomic and molecular physics spectroscopy and led to an immense number of novel devices and

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techniques that are used routinely in biological, forensic and analytic applications. One would hope that an extension of the techniques to the x-ray region would lead to similar tools that can be widely applied.

In what follows we will try to describe how we reached our experimental decision by surveying the possible x-ray sources, and then using the most intense source to design an experiment to utilize that source.

The objective of this research is to exploit the intense x-ray generation capability during the interaction of a suitable high power laser pulse with a solid target, such as of the Los Alamos Bright Source (LABS) [2]. The effort will start by demonstrating one of the non-linear processes. We will concentrate on the possibility of measuring the simplest nonlinear process, two-photon inner-shell ionization of chlorine, using keV X-rays generated from an aluminum target. An initial estimate shows that a measurable signal should be obtained using the Aluminum L_{α} line from the LABS plasma. The uncertainty in the calculation is largely due to novelty of the process. Theoretically, the problem is interesting because the closest intermediate states, in contrast to hydrogenic calculations, are not vacant. Thus, a measurement would be one of the first tests of this new approach to two-photon ionization.

3. Intense Source Regime:

How we define an intense x-ray source depends on the use of the source. In the present work we will judge a source by whether it can cause a measurable two photon signal during the lifetime of the radiation pulse. If, for scaling purposes, we use the two-photon ionization cross section for a hydrogenic ion of nuclear charge Z : [4]

$$\sigma^{[2]}(h\nu, Z) = Z^{-8} \sigma^{[2]}(h\nu/Z^2, 1) \quad (1)$$

where $\sigma^{[2]}(h\nu/Z^2, 1)$ is the two photon ionization cross section for hydrogen and $\sigma^{[2]}(h\nu, Z)$ is that for Z -ionized hydrogenic ion at the photon frequency ν . Then for an x-ray photon with an energy of a kilovolt, and ion with a nuclear charge Z of 10 the scaled hydrogenic photon energy $h\nu/Z^2$ is 10 eV [123 nm]. The two-photon ionization cross section then scales as:

$$\sigma^{[2]}(1 \text{ keV}, 10) = 3.9 \times 10^{-10} \sigma^{[2]}(10, 1) \quad (2)$$

Using the hydrogenic result for the nonresonant processes, Klarsfeld [3] gives a cross-section that linearly depends on I , the X-ray irradiance of a coherent linearly polarized field:

$$\sigma^{[2]} / I(\text{W/cm}^2) = \alpha = 1.56 \times 10^{-41} \quad (\text{cm}^4/\text{W}) \quad (3)$$

For the propagation of the x-rays we can use the generalized propagation equation: $dI/dz = -k I n - \sigma^{[2]} I n = -k I n - \alpha n I^2$, where k is the linear absorption coefficient and n is the particle density. The solution to this equation:

$$I = I_0 \left[e^{-knz} \right] \left[\frac{1}{1 + I_0 \frac{\alpha}{k} (1 - e^{-knz})} \right] \quad (4)$$

has two parts. The first part is the attenuation by the normal linear processes, the second part is due to the non-linear two-photon attenuation coefficient. When the linear attenuation is negligible, i.e. $k \rightarrow 0$, the solution reduces to:

$$I = \frac{I_0}{[1 + \alpha I_0 z n]} \quad (5)$$

The number of electron vacancies, holes, created in the target material by an irradiance I is given by $\Delta N = \Delta E / (h\nu)$, where ΔE is the energy absorbed in time ΔT by an illuminated area ΔA , $\Delta E = \Delta I \Delta T \Delta A$ with the creation of a hole with a bound energy $h\nu$. Combining all the formulas gives:

$$\Delta N = \alpha n \Delta z \Delta A \Delta T I^2 / (h\nu) \quad (6)$$

Using a solid target of thickness of one mean photon depth, [one micron thickness, or an equivalent gas target of path length $5 \times 10^{18} \text{ at/cm}^2$] an area equal to the x-ray source area [$100 \mu^2$], a pulse length of 1 psec, and a hole energy of 2 keV, then:

$$\Delta N = 2.44 \times 10^{-25} I^2 \quad (7)$$

An x-ray irradiance exceeding $2 \times 10^{12} \text{ W/cm}^2$ is required to create one hole, by two photon ionization, during one pulse. This irradiance is a very strong function of the x-ray wavelength and nuclear charge Z . Of course in an actual

experiment the number of observable events, ΔO , is much smaller than the number of generated holes, ΔO , must be large to overcome other sources of radiation, the observation efficiency that can be quite low, and the statistical uncertainty of counting. The number of observable event ΔO is equal to $\Delta N \Omega \epsilon$. Where Ω is the solid angle fraction of the detector, and ϵ is the efficiency of observation. Assuming a unit efficiency, a solid angle fraction of 10^{-4} and 100 observable events in that solid angle, then one requires an irradiance exceeding 10^{15} W/cm^2 to produce those signals in the kilovolt x-ray range.

If we want to repeat the analysis for lower Z material we will look at the Z dependence. The hydrogenic cross section varies with 6-th power of the scaled frequency. Hence the attenuation of the x-rays by two photon absorption at that scaled frequency [say $h\nu/Z^2=10$] depends on Z^8 . Since the binding energy depends on Z^2 , the required irradiance to give the same number of holes behaves as Z^5 . For a 100 eV x-ray photon one has to perform the experiment in a beryllium target with $Z=4$, and the required irradiance, $5 \times 10^{12} \text{ W/cm}^2$, is Z^5 time smaller than that at 1 keV

4. Intense Sources of Short Wavelength Radiation.

As mentioned in the introduction, various sources of intense x-ray radiation has been built. The most common sources are built around Synchrotron accelerators where the large facilities generate large average power, but with a small peak power. The maximum spectral brightness of synchrotrons for example is [5] 10^{12} photons/sec/mrad/eV at 100 eV which exceed that of a regular x-ray tube by many orders of magnitude.

Harmonic generation from a laser is another possible source. Many groups measured an unoptimized conversion efficiency of a PPM per harmonic. Thus for a 33 mJ, 100 fsec Ti:Sapphire laser, the measured x-ray yield at 100 eV [the 66-th harmonic of a Ti:Sapphire laser] is roughly 10^5 photon/mrad [6]. If one guesses that the x-ray pulse length is $\ll 100$ fsec [roughly $100 \text{ fsec}/n$, where $n \sim 100$ is the harmonic number], and that the bandwidth $\ll 1$ eV [roughly equal to the laser bandwidth $\sim 2 \text{ \AA} @ 8000 \text{ \AA} \sim 5 \times 10^{-4} \text{ eV}$], then the actual spectral brightness is much higher than 10^{19} photons/sec/mrad/eV, and much higher than the peak spectral brightness of a synchrotron or an x-ray tube. If the laser runs at 1 KHz, then the time average flux is only 10^8 photons/sec/mrad.

Black body radiation is another source of radiation.. The radiance of a black body of temperature T is $I=10^5 T^4$ where the temperature is measured in eV. A black body of temperature .32 keV emits of 10^{15} W/cm^2 , and have a peak radiance at $h\nu=3 kT= 960 \text{ eV}$ of $10^{23} \text{ X-rays/sec/cm}^2/\text{eV}$. Such conditions are not unusual, they commonly occur in laser-generated plasma. The main

disadvantage of this source is the broad-band non selective nature of the radiation.

X-ray laser sources are also available at many wavelengths[28 to 428 nm] in the soft x-ray domain [7]. A conversion efficiency of a PPM is typical of the best double-passed x-ray lasers[8]. However since they use kiloJoule style lasers to produce lasing, an output radiance of 10^{14} W/cm²/sr is available at 23 nm with divergence of 10 mr x 10 mr. Thus their irradiances can exceed 10^{31}

Soft-Xray/Sr/cm² at an energy of 54 eV.

Line emission from laser generated plasmas is another source of intense radiation. For long laser pulse lengths, exceeding a nanosecond, the conversion efficiency has been measured for the hydrogenic and helium-like lines of many elements [9,10] the data for the He-like resonance line can be summarized as:

$$\zeta = 6554 (h\nu)^{-4} \text{ Photon/sphere/Joule} \quad (8)$$

where the photon energy is measured in keV.. For an aluminum plasma the 1.6 keV He-like resonance line output can exceed 10^{14} photon per joule of incident laser light, which corresponds to a conversion efficiency of the laser light to x-ray lines of around 2%. Since the few nanosecond x-ray pulse length of these plasmas is relatively long the output radiance from a plasma irradiated by a joule of laser light is around 10^{12} W/cm². The way around this is the use of a short pulse laser, say shorter than a picosecond, to generate the x-rays.[11,12] The short pulse work of [11] that generated an intense source of x-ray radiation at the Aluminum He-like line of 1.6 keV motivated us to study whether we can use such intense radiation.

Choice Of Material

H- and He-like Aluminum

Figure 1.

The choice of material depends on the K-shell ionization energy and the X-ray line used.

5. Two-Photon Photoionization Physics Using Non-Coherent Sources:

Three atomic targets Cl, Ar, and K are possible candidates that allow the observation of two-photon K-shell ionization by the various lines from an aluminum plasma [Figure 1]. Because the He(2p-1s) resonance line is the most intense and has the smallest photon energy, the combination with a chlorine atom leads to the largest signal. The chlorine atom was the atom with the largest nuclear charge where two photon ionization was possible. Chlorine has a further advantage over smaller nuclear charge material where the cross sections were larger because of its lower sensitivity to photoionization by the rest of the plasma radiation.

Figure 2.

Schematic of the levels and processes in a chlorine atom.

A simplified energy levels of chlorine is shown in [Figure 2] where the possible photoionization mechanisms are shown as well. A unique feature of photo ionization from the inner shell of a multi electron atom is the presence of intermediate energy levels states that are fully occupied and that are quite far off resonance. This is in contrast to two-photon ionization with visible light where electrons can be promoted to the intermediate energy-level states that are unoccupied, and where those intermediate states are not far off-resonance. The actual calculations of [13] showed that there are two paths to photoionization, the first is through exciting the intermediate levels then photoionizing that level, and the second through photoionizing an electron from the outer shell followed by exciting that level from the ground state of chlorine. Although each transition is far off resonance, the final state conserves energy, generates a

x-ray source are: a source diameter of less than 5 micrometers, thermal electron temperature of 300 eV, hot electron temperature of 12 keV, line width of few eV, and little hot electron generation. The x-ray radiance exceeded 5×10^{13} W/cm² in the Aluminum H-like resonance line at 1.6 keV. The flux, 3×10^{27} Xray/cm²/Sr/eV, is large enough to be usable as a source for our experiment [Figure 3]. What remained was to design a target geometry that uses this source and removes the extraneous x-ray radiation that accompanies the required line.

Figure 3.

Measured
Spectrum from
Aluminum at a
XeCl laser
irradiance
 5×10^{18} W/cm²

7. Target Hydrodynamic:

Positioning a chlorine target presented a challenge. The x-ray flux decreased rapidly with distance from the x-ray source, and the signal decreased as the fourth power of distance. Thus a gaseous target, though attractive, would not have produced a measurable signal because not enough density could have been placed close to the x-ray source without perturbing the laser interaction itself. A solid target NaCl provided a convenient solid target that could be deposited close to the source and had the added convenience of providing a null measurement through the presence of the sodium which requires 3 photons to give an x-ray signal at 3.384 keV. That signal would be used to measure a competing rate for the production of inner shell holes by hot electron collisions. The design of the target requires that we filter the extraneous x-rays, mostly that lie at low photon energies, thus a silicon substrate was needed to do two tasks. First, to filter the low energy x-rays completely, and attenuate

hole in the inner K-shell core, and produces a free electron with an energy of 300 eV. This hole can be filled by emission of an Auger electron or a fluorescent x-ray at 2.62 keV. The results of the calculation for the total two-photon ionization cross section, for a neutral chlorine atom with linearly polarized coherent radiation at $h\nu=1.6$ keV was

$$\sigma^{[2]} / I(\text{W/cm}^2) = \alpha = 1.032 \times 10^{-41} \quad (\text{cm}^4/\text{W}) \quad (9)$$

in contrast the scaled hydrogenic result, at this particular energy, gives a coefficient that is about a factor of two lower,

$$\sigma^{[2]} / I(\text{W/cm}^2) = \alpha = 5 \times 10^{-42} \quad (\text{cm}^4/\text{W}) \quad (10)$$

Since the light that we use is chaotic, non-coherent and occurs from all directions, one has to modify these estimates to account for the difference in behaviour from a laser induced two-photon ionization. The net result is to increase these estimates by a factor of two. This is still within the estimates used to judge the feasibility of the experiment. The next task is to create the necessary flux of radiation.

6. X-ray Source Design

Using a small KrF laser system Cobble et al[11], measured a large conversion efficiency to the Aluminum x-ray radiation, nevertheless their x-ray-flux was short too low to perform this experiment. The cause of the large conversion efficiency, when compared to other laser based sources, was not well understood, but was suspected to be caused by the low contrast pulse that interacted with the target. Since, we have built a well diagnosed XeCl laser system where we could monitor, and to a certain extent control, the prepulse in the final laser output[14]. At the highest laser irradiance, we measured a conversion efficiency was 0.8 % of the laser energy into one aluminum line[15]. The reason for the large efficiency, compared to other measurements[16,17,18] was due to the presence of the prepulse that was controlled and that provided the appropriate conditions in front of the target for x-ray conversion.. The pulse length was also measured to be less than 5 psec [2], much longer than the interacting laser pulse, and of the order of the atomic ionization equilibrium times. While shorter x-ray pulses have been generated with high-contrast no prepulse lasers, their conversion efficiency were abysmal[16,17,18]. For these experiments, what matters is the x-ray flux, not the x-ray pulse length per se.[2, 17, 18]. Some other characteristics of the

the high energy x-rays, but transmit the aluminum x-rays. Second, and because of its crystal structure, to form a supersmooth optical surface where the aluminum was deposited [The depth of focus of the optical system was few tens of microns].

Figure 4.

Schematic of
the compound
target

Hydrodynamic calculations were carried out to find the appropriate thickness to be used in order to maximize the x-rays and to mitigate the effect of damage from the prepulse before the experiment could take place. The calculations used a radiation transport code that handled the laser radiation interactions including resonance absorption, above threshold ionization, multiphoton absorption, and that treated the hydrodynamics in 1-D [19]. Some of the results show that the shock from the prepulse, which occurs 3 ns before the main laser pulse, destroys the target well after the end of the x-rays from the main pulse are over. The final design is shown in Figure 4.

8. Measurement.

The resultant x-rays were measured with a filtered CCD camera that was used in the pulse height analysis mode. Each absorbed x-ray deposited its energy in a pixel or two, and 3.6 eV were required to create one free electron that was collected. The CCD camera was filtered by an 8 μm thin Nickel filter that absorbed the visible light and attenuated the intense Aluminum radiation by a factor of 8×10^{-11} .

Figure 5. The measured x-ray spectrum from 100 laser shots, using 120 eV wide energy bins.

The result of 100 shots on the target were accumulated with the CCD. The pulse height distribution, excluding readout noise, and the background counts, is shown in Figure [5]. The error bars are the 1σ statistical error bars. The histogram has energy bins that are 120 eV wide about the resolution of the CCD detector, as measured with an electron beam source of x-rays. The histogram shows the Aluminum lines around the 1.6 keV bin as well as a weak signal where the Chlorine K_α line at 2.672 keV. With a bin that large, we do not observe the K_α from the Potassium target, indicating that the hot electron production rate of holes was less than that from the two-photon absorption rate. Other features of the spectrum is some energy at 2.2 keV which may come from the series limit in Al-XIII emission, and some emission at 2400 eV which may come from the late time emission of the Si XIV species. The Aluminum line at 1.48 keV can not be resolved.

The amplitude of the unresolved aluminum He-Ly $_\alpha$ line at 1.48 keV gives us a source strength of 10^{12} photon/shot roughly three times the estimate from flat crystal spectrometry. The amplitude of the Chlorine feature gives us a hole production rate of 1.1×10^6 vacancies per shot. The estimated number of vacancies is 3 times that predicted using crystal spectrometry and the theoretical cross sections. These measurements should be treated with care, since the x-ray pulse length was not measured accurately, the laser was

generating multiple x-ray pulses, and the immense attenuation of the Nickel filter amplify any published attenuation cross section measurements errors.

9. Conclusion

We have performed an experiment that should be viewed as proving the feasibility of the measurement of the two x-ray-photon absorption rate. Many difficulties must be overcome before a cross section measurement can be performed with any accuracy.

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