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PHYSICAL PARAMETER DEPENDENCE OF THE X-RAY GENERATION IN INTENSE LASER – CLUSTER INTERACTION

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Abstract : Studies on laser – cluster interaction performed on the LUCA facility (French acronym for Ultra Short Tunable Laser, CEA Saclay) allow to observe the production of hard x-rays in the 1 to 5 keV range when rare gas clusters of nanometer sizes are heated by strong optical fields ($F > 10^9$ V/cm). First complete quantitative measurements of absolute photon emission yields, as well as of charge state distributions of ionic species with inner shell vacancies have been performed as a function of several physical parameters governing the interaction. Our measurements give rise to fundamental results like an optimum heating time and an intensity threshold in the x-ray production. These data provide direct insight into the interaction dynamics and into the heating processes involved.

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Keywords: X-rays; Laser; Ionization; Clusters; Argon; Xenon

1. Introduction

The interaction of high intensity ultra short laser pulses with matter has received considerable attention in the last few years since the appearance of the CPA (Chirp Pulse Amplification) technique. The matter behavior under such extreme conditions has opened new fields of investigation with, for instance, the generation of high order harmonics, the acceleration of heavy particles, or the development of bright x-ray sources. The possibility to generate intense and short hard x-ray pulses with clusters or solids as target is a particular subject of interest for potential applications. The optimization of such sources demands however knowledge of mechanisms involved during laser – matter interaction, what can be reached through specific studies dedicated to fundamental aspects.

The use of atomic nanometer clusters gives several advantages. Clusters constitute renewable targets free of debris, contrary to solid targets. The energy coupling between radiation and clusters is very efficient due, in particular, to their local density close to the solid one ($\sim 10^{22}$ cm^{-3}). Their size much lower than the laser wavelength and slightly below the skin depth allows to assume that the electric field inside the cluster is uniform. Moreover, they offer the opportunity of observing well separated nanoplasmas: usually the mean atomic density does not exceed 10^{17} cm^{-3} leading to inter-cluster distances larger than 1 µm. Therefore, they form simple systems for the study of laser – matter interaction experimentally and theoretically.

Early studies have shown that clusters under strong laser fields ($F > 10^9$ V/cm) generate multicharged ions reaching MeV [1], hot electrons up to few keV [2] and keV x-ray photons [3, 4]. Different theories have been developed to explain the experimental features observed in the high laser intensity regime [5, 6, 7]. These first results boost the field and numerous kind of investigations have been made; a recent review is presented in [8]. The mechanisms responsible for the laser energy absorption are still not completely understood and give rise to controversy. Special attention has been paid to the nanoplasma model proposed by Ditmire et al [5], which offers a complete scenario of the interaction, taking into account ionization, heating (through inverse Bremstrahllung and laser electric field) and explosion processes simultaneously. As an example, new theoretical implementation has been recently proposed by Megi et al [9] showing the importance of electrons – surface collisions in the heating process and so far neglected. On another side, recent experimental results [10, 11, 12] on ion and/or electron angular distributions show, besides a disagreement with previous measurements [1], an anisotropy of the emission, which is not account for within the nanoplasma model [5]. All of these studies related to ion or electron emission are connected to both the dynamics of the induced plasma and the cluster explosion.

So far, most of the studies on x-ray emission have been mainly limited to qualitative observations. Very few systematic measurements with the parameters influencing the x-ray production have been realized. Moreover, simulations are still needed to explain the production of ions *with inner shell vacancies* responsible for the strong hard x-ray generation. Thus, our goal is to understand how electrons initially produced by Optical Field Ionization with a few eV can reach enough energy (up to a few keV) to generate inner shell ionization through inelastic electron – ion collisions. KeV x-rays are studied from rare-gas clusters ($(Ar)_n$, $(Kr)_n$ and $(Xe)_n$ clusters with n between 10^3 and 10^6 atoms/cluster) irradiated with intense ($I \sim 10^{14}$–10^{17} W/cm²) infrared and blue laser pulses. Variation of the pulse duration in the 50 to 2000 fs range was available for infrared light. The observed x-rays are emitted by highly charged ions: for example, from Ar^{12+} to Ar^{16+} with K vacancies [13, 14] in the case of Ar clusters and Xe^{q+} ($q \geq 24$) with L vacancies in the case of xenon [15]. Since the lifetime of excited levels produced is rather short for highly charged ions (down to 15 fs in the case of $Ar^{16+}(^1P_1)$), the emitted x-rays allows to probe the heating mechanisms on a very short time scale (some fs). We have performed quantitative studies on the evolution of absolute photon emission yields and complete charge state distributions with different physical parameters governing the interaction; namely intensity, polarization, pulse duration and wavelength of the laser, size and density of the clusters. These studies, made under well-controlled conditions, allow to determine the sensitivity of the x-ray production upon these physical parameters and, consequently, to find conditions for optimization. They give rise, as well, to fundamental results, which will provide information on the dynamics of the interaction and on the heating processes involved. Scaling laws with the laser intensity and the cluster size have already been presented in Refs [13, 14]. With Xe clusters, we have also shown [15] that the x-ray yield does not follow a λ^{-6} law as stated in [16]. Furthermore, we have demonstrated that the charge state distribution of Xe ions strongly depends on the laser wavelength [15], while no difference has been observed for Ar clusters. In this paper, the work presented emphasizes our results on the laser intensity dependence showing evidence of a threshold in the x-ray production. In addition, we discuss the investigations over the pulse duration, which leads to an optimum heating time at constant laser energy. In the next section, a brief description of the experimental set-up is given. In section 3, we discuss, in more detail, the laser intensity dependence and, in particular, the consequence of experimental observations on the inner shell ionization probability. In section 4, the x-ray production as a function of the pulse duration is presented and finally, conclusion is given.

2. Experiment

All the experiments have been performed on the LUCA facility (French acronym for Ultra Short Tunable Laser, CEA) at Saclay. The whole experimental set-up, including the apparatus used to generate the clusters as well as the x-ray spectrometers, has been described in previous publications [4, 13, 14]. Briefly, $(Ar)_n$, $(Kr)_n$ and $(Xe)_n$ clusters with n between 10^3 and 10^6 atoms/cluster are generated within a pulsed adiabatic expansion of a gaseous jet through a conical nozzle (Hagena's type [17]). The backing pressure P_0 allows to control the cluster size according to current use scaling laws. The number of clusters in the interaction zone has been evaluated using the condensation efficiency obtained through some specific studies [18]. The intense laser beam is generated with a Ti:sapphire laser system delivering infrared pulses at a repetition rate of 20 Hz. The energy per pulse can reach 100 mJ and the pulse duration can be varied from 50 fs to 2 ps, for λ= 800 nm, leading to laser intensities up to 10^{17} W/cm² after focusing. The laser was frequency doubled to a wavelength of 400 nm using a 2 or 3 mm thick KDP crystal. The maximum second harmonic energy was 15 mJ which could be focused up to a peak intensity of 3 10^{16} W/cm^2 with a pulse duration about 150 fs. The x-rays are analyzed using two Si(Li) detectors and a high resolution high transmission Bragg-crystal spectrometer equipped with a flat mosaic graphite crystal and a position sensitive detector working in the photon counting mode. With this spectrometer, we can perform precise x-ray spectroscopy since a resolution of the order of 1.5 eV at 3 keV is easily reached. The following transitions have been recorded:

i) in the case of argon, $2p \rightarrow 1s$ from Ar^{12+} to Ar^{16+} and $3p \rightarrow 1s$ from Ar^{15+} and Ar^{16+},

ii) in the case of xenon, $3d_{3/2} \rightarrow 2p_{1/2}$ and $3d_{5/2} \rightarrow 2p_{3/2}$ from Xe^{q+} with $q \geq 24$.

The knowledge of transmission, solid angle and efficiency of the various detection systems provides measurements of absolute x-ray yields. This technique is very well suited to follow the evolution of yields and ionic distributions as a function of a given parameter when controlling the others.

3. Laser intensity dependence

The evolution of x-ray yields as a function of the laser intensity (I) has already been presented in the case of argon and krypton clusters for a 800 nm laser wavelength and a 55 fs pulse duration [14]. With xenon clusters under similar experimental conditions, the same behavior is observed (figure 1). Indeed, it is worthwhile to note that whatever the cluster species, the laser wavelength and the pulse duration, we find clearly two regimes of production. Quantitatively, we first observe a rapid increase after a threshold (~ 3 10^{15} W/cm^2 in the present case) followed by a smooth evolution like a $I^{3/2}$ law for larger intensities

(> 7 10^{15} W/cm²). Dealing with studies on laser - atom interaction, this behavior is associated to a saturation regime [19]: the physical signal grows with the number of partners contained in a volume, called the *effective focal volume* ($V_{\text{eff. foc.}}$), where a given intensity threshold (I_{th}) is reached. In the case of laser – cluster interaction, we found that the number of emitted x-rays is also proportional to this effective focal volume given by the following equation [19]:

$$V_{\text{eff.foc.}} = \pi \cdot z_R \cdot w_0^2 \left\{ \frac{4}{3} \cdot \left(\frac{I}{I_{th}} - 1 \right)^{1/2} + \frac{2}{9} \cdot \left(\frac{I}{I_{th}} - 1 \right)^{3/2} - \frac{4}{3} \cdot \operatorname{arctg} \left(\frac{I}{I_{th}} - 1 \right)^{1/2} \right\} \quad (1)$$

where z_R is the Rayleigh length and w_0 the beam waist. This evolution is well reproduced by the inner shell ionization model developed in our group and described in Refs [13, 14]. Briefly, in this simple dynamical model, the electrons, first produced by Optical Field Ionization, acquire energy only through oscillations in the laser field. This leads to an intensity threshold but at least one order of magnitude larger than the observed one (predictions of this model is plotted in dashed line in figure 1). Efficient inner shell ionization mechanisms at low laser intensities are obviously missed. Charge polarization effects [20], already invoked to explain the anisotropy in the electron emission for instance [11], are investigated. New simulations taking into account the field inside the cluster induced by this effect are under progress and could explain the observed low intensity threshold. Very recently, new measurements on the evolution of absolute yields with the laser intensity for larger pulse durations (> 50 fs) exhibit even lower intensity thresholds: I_{th} ~ 3 10^{14} W/cm^2 has been found at 575 fs for argon clusters. These last experimental results will be published in a forthcoming paper and discussed within new theoretical approach [20].

Nevertheless, our experimental results highlight a fundamental point. Indeed, the evolution of absolute yields with the effective focal volume has a consequence on the inner shell ionization probability, which is simply given by the ratio between the number of emitted x-rays and the number of emitting atoms. Since the absolute yields follow the evolution of the effective focal volume, we can conclude *that inner shell ionization rates per atom are constant whatever the peak intensity, provided the threshold's value is exceeded*. For the experimental conditions given in figure 1, we find a probability of a few 10^{-5} per atom. So far, none of the available models are able to explain such a large probability for laser intensities in the range of a few 10^{14} W/cm^2.

4. Pulse duration dependence

To better understand the time competition between heating processes (inverse Bremsstrahlung, laser driven electrons, polarization charge effect…) or even cooling (cluster expansion), we have investigated the influence of the pulse duration (τ) for a constant laser energy but different cluster sizes (figures 2 and 3). We first observe a strong increase of the x-ray production up to an optimum followed by a decrease. In the case of Ar clusters, a maximum is found to be 130 fs independently of the cluster size within the range of backing pressure used (figure 2). For Xe clusters, the maximum is in the range of 200 to 300 fs as illustrated by the results plotted figure 3. At a constant laser energy value, the x-ray yields exhibit an optimum heating time. The difference between Ar and Xe clusters may be due to different time scales for the ionization of L (for Ar) and M shells (for Xe). At present, only the decrease in x-ray yields for large pulse durations can be understood by the inner shell ionization model. Actually, in this approach, the number of x-rays (N_X) depends on the effective focal volume ($V_{\mathrm{eff.\,foc.}}$) and on the number of optical cycles (n_{cycles}) by simply writing [13, 14]:

$$N_X \propto n_{\mathrm{cycle}} \times V_{\mathrm{eff.foc.}} \qquad (2)$$

Since $n_{\mathrm{cycles}} \propto \tau$ and $V_{\mathrm{eff.foc.}} \propto I^{3/2}$ with $I \propto \tau^{-1}$, N_X should decrease as $\tau^{-1/2}$, which may reproduce the experimental data observed after the optimum (solid curves in figures 2 and 3).

However, the observed behavior, at least before this optimum time, seems most certainly related to the fact that the intensity threshold appears to be also pulse duration dependant (see section 3). For short pulses, this threshold is larger than for long pulses, resulting in smaller effective focal volumes (see equation 1) and then x-ray yields. Nevertheless, the variation of the intensity threshold value with the pulse duration is far to be understood and, once again, quantitative predictions are needed.

5. Conclusion

We have measured the laser intensity dependence of x-ray production for Ar and Xe rare-gas clusters. A clear threshold behavior is observed, showing that inner-shell ionization probabilities are constant for given experimental conditions (namely cluster species, size and density, laser wavelength and pulse duration). The observed increase of x-ray emission with laser intensity is solely due to the increase of the effective focal volume provided an intensity threshold is achieved. Hence, for a given laser energy, an optimum of the x-ray yields is reached by defocusing the laser to some extent since the effective focal volume presents a maximum as a function of the beam waist. We have also measured the laser pulse duration dependence of x-ray production for the same species keeping the laser energy constant. An optimum for the pulse duration is also observed, somewhat larger for Xe than for Ar clusters. Since we also observe a decreasing intensity threshold for increasing pulse duration, this optimum pulse duration value may well be due to a combined effect of threshold value and laser intensity variations on the effective focal volume, together with the number of optical cycles. These new results bring challenging questions that must be answered by new models to come.

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Figure captions:

Figure 1 : Evolution of the absolute x-ray yield with the laser intensity in the case of 9 bar xenon clusters (λ = 800 nm; τ = 55 fs). The curves show the evolution of the effective focal volume with the experimental intensity threshold (3 10^{15} W/cm²) in solid line and with the predicted – see text – intensity threshold (4.5 10^{16} W/cm²) in dashed line.

Figure 2 : Evolution of absolute yields with the pulse duration (τ) for a 800 nm laser at constant energy (20 mJ) in the case of argon clusters for different backing pressure (i.e. different sizes): a) P_0 = 20 bar, i.e. 2 10^5 at/cluster ; b) P_0 = 30 bar, i.e. 5 10^5 at/cluster and c) P_0 = 40 bar, i.e. 10^6 at/cluster. The solid curve corresponds to the evolution as $\tau^{1/2}$ – see text.

Figure 3 : Evolution of absolute yields with the pulse duration (τ) for a 800 nm laser at constant energy (35 mJ) in the case of xenon clusters for a 3.5 bar backing pressure – i.e. 4 10^4 at/cluster. The solid curve corresponds to the evolution as $\tau^{1/2}$ – see text.

Figure 1

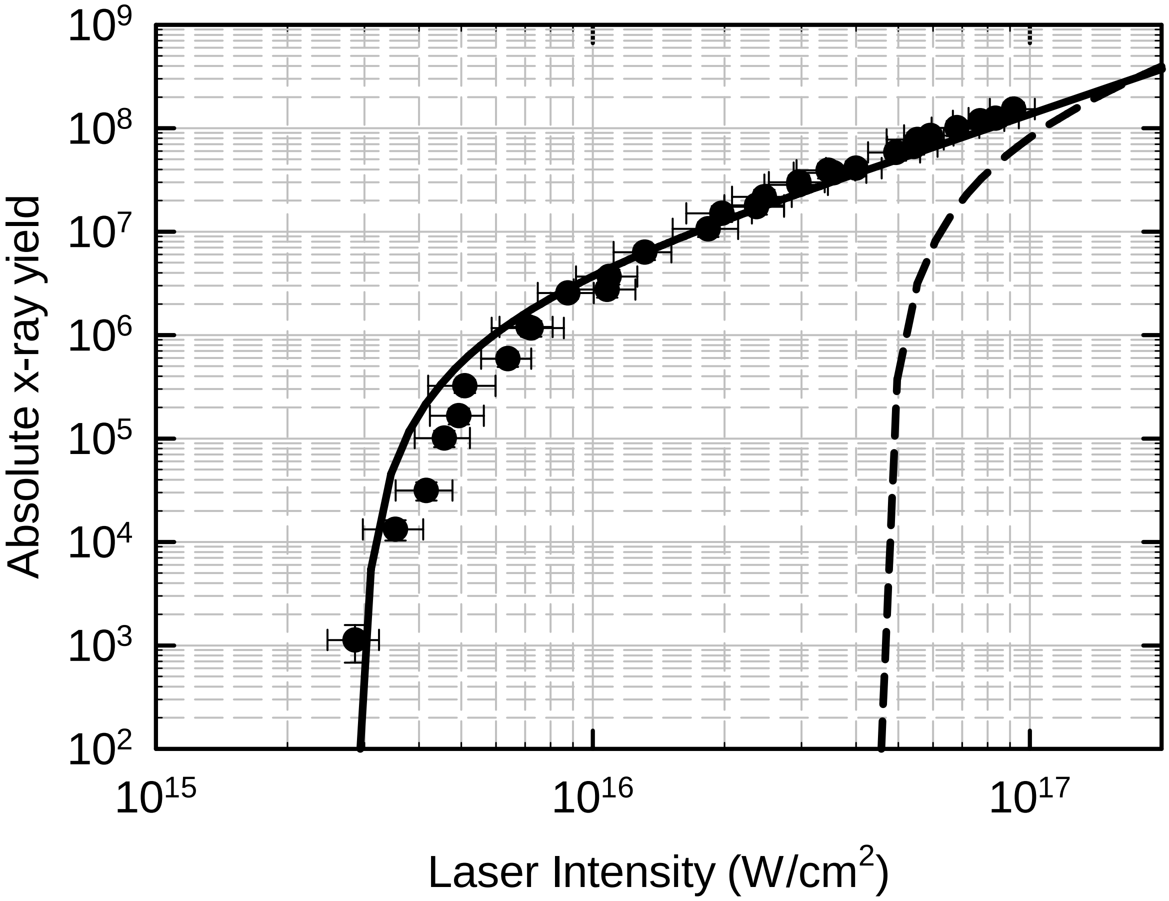

10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
Absolute x-ray yield
10^{15}
10^{16}
10^{17}
Laser Intensity (W/cm^2)

Figure 2

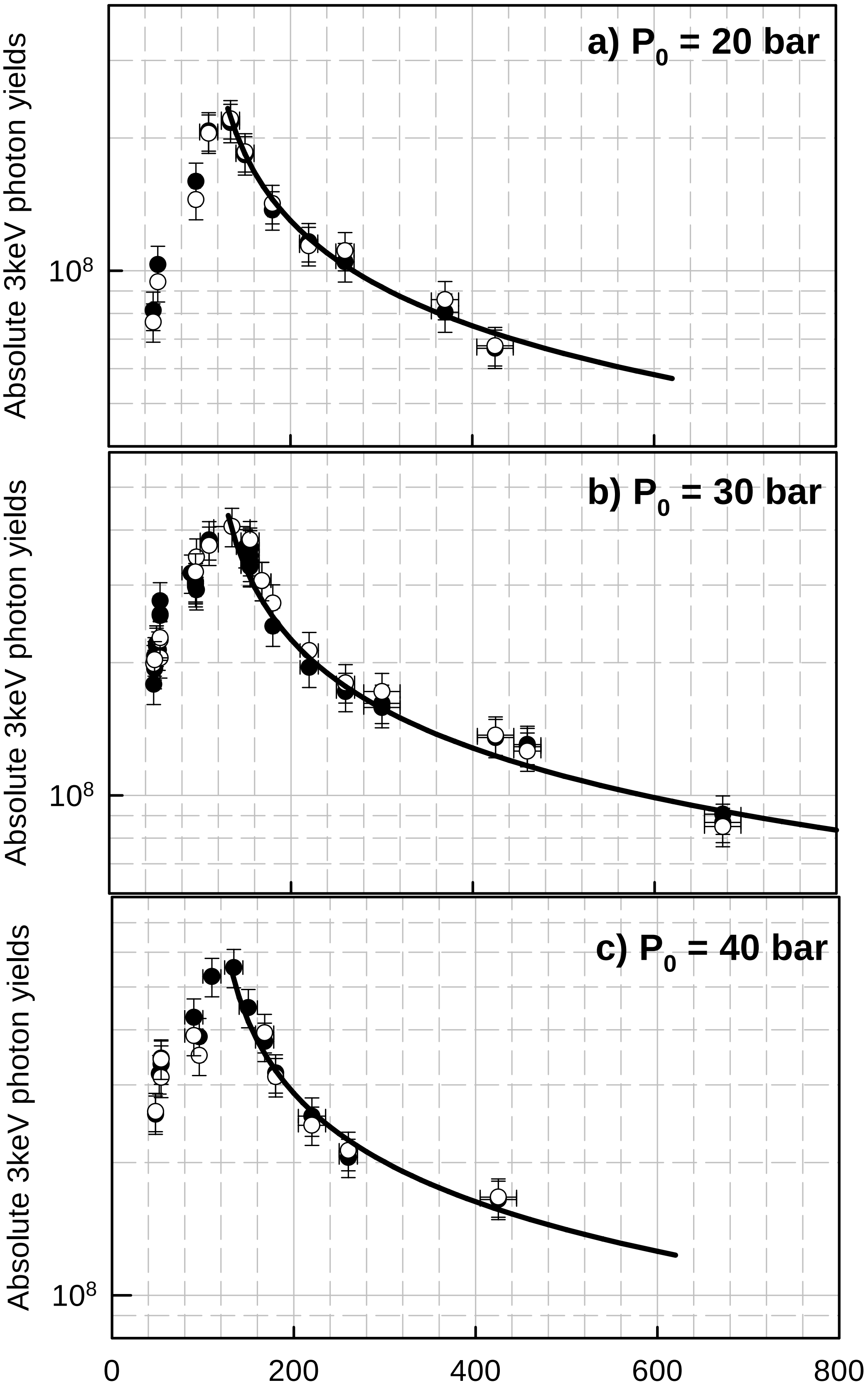
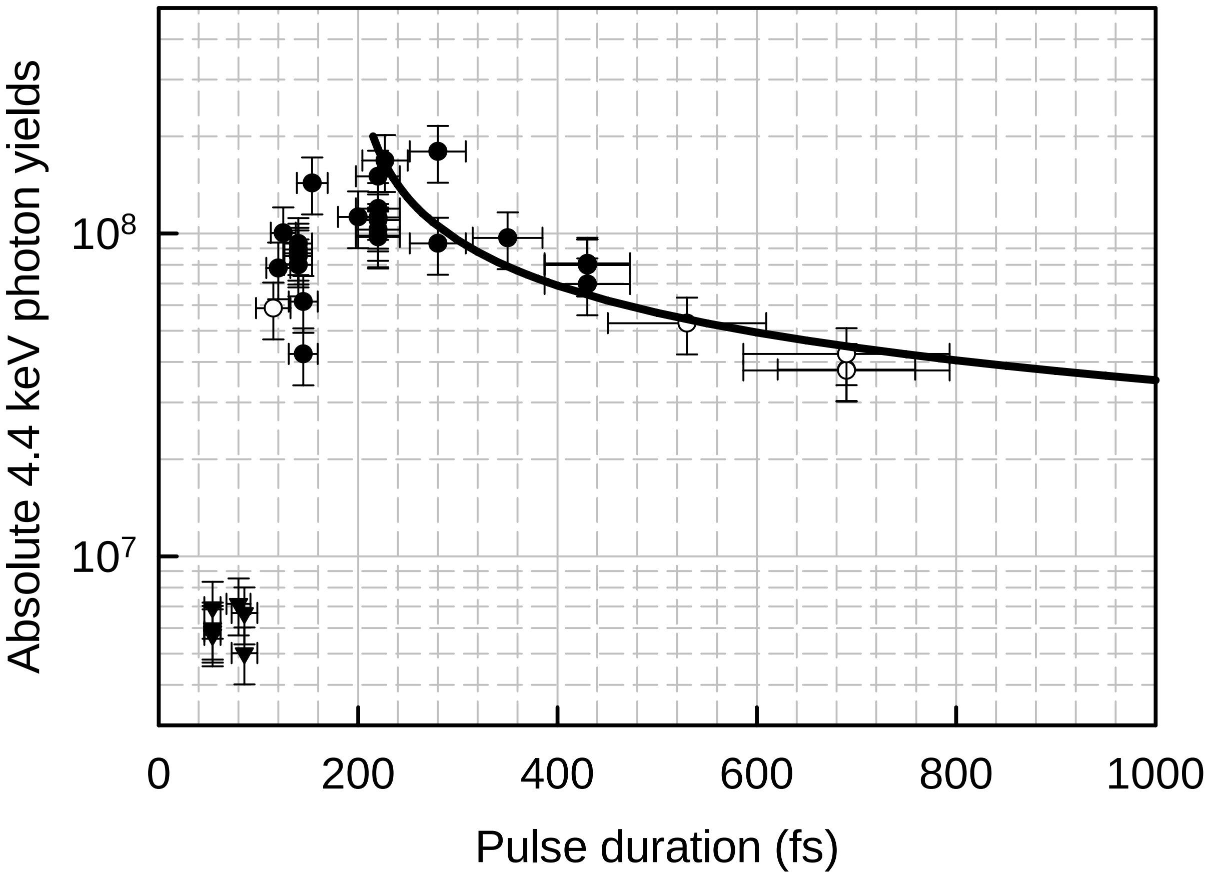

a) P_0 = 20 bar
b) P_0 = 30 bar
c) P_0 = 40 bar
Absolute 3keV photon yields
10^8
0
200
400
600
800
Pulse duration (fs)

Figure 3



Absolute 4.4 keV photon yields
10^8
10^7
0
200
400
600
800
1000
Pulse duration (fs)