Observation of Subpicosecond X-Ray Emission from Laser-Cluster Interaction

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We present the first experimental evidence of the subpicosecond duration of x-ray pulses emitted from laser-irradiated clusters, demonstrating the suitability of such a debris free target for ultrafast x-ray science applications. The K-shell emission (~ 3 keV) from large Ar clusters (6×10^5 to 4×10^6 atoms) is time resolved, when irradiated by ultrashort (40 fs to 5 ps) and intense laser pulses (10^{15-17} W/cm^2). The observations are supported by hydrodynamical and collisional-radiative calculations, that reproduce the extremely short x-ray pulse duration.

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The production of ultrashort x-ray pulses from intense laser-matter interaction has been intensively studied over the last 15 years as they offer unprecedented time resolution to image atomic structure of dynamical systems [1–4]. Several mechanisms have been proposed to get multi-keV x-ray bursts with durations down to the sub-ps range. K_α sources were first developed from laser interaction with solid targets. These monochromatic sources have a duration as short as a few hundred of fs, as demonstrated in ultrafast x-ray diffraction experiments [3,4]. The laser intensity required is a few 10^{17} W/cm^2, allowing the use of a high repetition rate laser (1 kHz) to get suitable average x-ray power for the study of complex systems [5]. More recently, a betatron x-ray source based on the interaction with a gas jet, has been proposed [6]. The duration is expected to be even shorter than 100 fs [7]. This source offers a broadband emission, extending the possible applications of ultrafast x-ray sources to the study of noncrystalline structures (liquid, warm dense matter, plasma) via x-ray absorption spectroscopy. Nevertheless, relativistic laser intensities are required (a few 10^{19} W/cm^2), and the repetition rate achievable is today limited to 10 Hz.

An alternative way to get both monochromatic and broadband x-ray emission consists of using the thermal emission of a hot and dense plasma produced from the interaction of an intense fs laser pulse with a solid density target. This thermal plasma x-ray source is currently operated as a backlighter for ultrafast absorption spectroscopy of plasmas with high energy lasers [8]. Other absorption experiments use it in conjunction with smaller laser systems compatible with high repetition rate [9]. Foil targets have been proposed to improve the heating efficiency and the subsequent x-ray emission intensity, since no thermal conduction is expected in the target depth. Furthermore, the x-ray pulse duration is expected to shorten due to faster hydrodynamical plasma expansion and cooling. Nevertheless, all the published results indicate thermal x-ray durations of a few ps with a long tail of a few 10 ps in the case of resonance lines such as He$_\alpha$ ($1s^2 - 1s2p$) line [10–12].

Cluster targets have received great attention over the last decade, since high laser energy absorption has been observed, leading to copious XUV and x-ray emission, high energy electrons and ions, and even neutrons via fusion reactions (cf. ,e.g., [13,14]). Unlike solid targets, a cluster jet is easily renewable and can be operated with high repetition rate lasers without any debris production. The main published results concerning the duration of cluster emission from XUV to x-ray [15–18] demonstrate that it shortens as the photon energy considered increases, down to a duration shorter than the 100 ps resolution of Ref. [15] for keV x rays. This is expected from the thermal emission of such laser-heated nanoplasmas. Although very short multi-keV x-ray pulses should be expected from clusters, no measurement has been yet reported to confirm this feature with sub-ps resolution.

In this Letter, we present the first measurements of multi-keV x-ray emission from laser-irradiated clusters with a subpicosecond duration, which is significantly shorter than previously obtained results with foil targets. Laser absorption as well as x-ray intensity and duration have been studied as a function of the laser pulse duration. It has been observed that an optimal laser pulse duration (400 ± 100 fs) can be set to optimize the conversion efficiency from the laser energy to the x-ray pulse, leading to a useful, clean and bright x-ray source for applications in the emerging ultrafast x-ray science. Based on absorption measurements, a simplified hydrodynamical model is proposed and coupled to collisional-radiative x-ray emission calculations. This model successfully reproduces experimental observations. The ultrashort x-ray pulse duration is understood as a combination of the very efficient laser heating with the fast expansion and cooling induced by spherical geometry of the clusters.

Experiments have been performed with the CELIA Ti:sapphire kHz laser facility [19], delivering a minimum pulse duration of 40 fs full width at half maximum

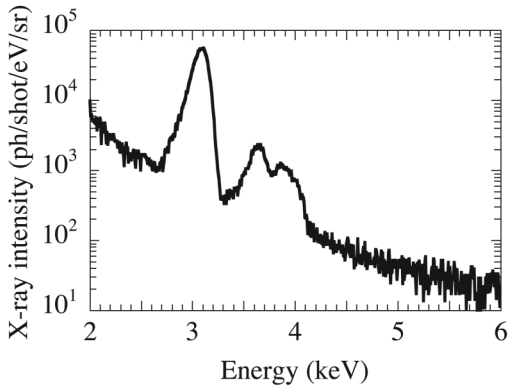

FIG. 1. X-ray emission spectrum recorded by the broadband spectrometer. 275 Å average radius Ar clusters are irradiated by 2.0 mJ, 320 fs FWHM, linearly polarized laser pulse. The laser intensity is $7.5 \pm 2.5 \times 10^{15}$ W/cm^2. 25 μm Be and six other filters, each composed by 6 μm Mylar and 0.3 μm Al, are set up in front of the detector.

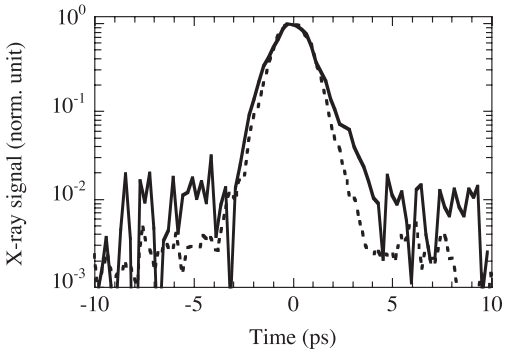

FIG. 2. Temporal profile of the x-ray pulse (with arbitrary origin). 275 Å average radius Ar clusters are irradiated by 2.0 mJ, 320 fs FWHM, linearly polarized laser pulse. Full line: x-ray measurement. Dashed line: 0.8 ps rms instrumental response as measured on the same setup with third harmonic generated from 40 fs laser pulses.

(FWHM) and up to 4 mJ on target. The experimental setup is basically the same as previously reported [20]. We use an Ar cluster jet delivering relatively large clusters (180 to 350 Å average radius, respectively, corresponding to $6 \times 10^5 - 4 \times 10^6$ atoms [21]). A broadband x-ray spectrometer, based on a charge coupled device (CCD) camera operating in photon counting mode, has been set at 45° from the laser axis in the forward direction. A cumulative x-ray streak camera, offering 0.8 ps rms temporal response has been set at 10° (forward) from the laser axis [22]. In this geometry, the temporal broadening induced by the integration over the 600 μm long plasma column is negligible (~ 30 fs).

The x-ray emission spectrum is presented in Fig. 1, obtained from 275 Å average radius Ar clusters irradiated by 2.0 mJ, 320 fs FWHM laser pulses. One can see a continuous bremsstrahlung emission, and three different spectral structures identified as the K-shell emission, corresponding to transitions from the L, M, N shells (left to right) down to the K shell. The brightest one is centered near 3 keV. It contains the whole set of $2p - 1s$ transitions (including the He_α line) which are not spectrally resolved by this spectrometer (resolution $\sim$150 eV). After filter transmission correction, we estimate that 3×10^8 photons per shot are emitted in 4π sr, integrated over the 2.7–3.3 keV range. In order to remove any XUV contribution from the time resolved signal, up to 4 aluminized Mylar filters (described in Fig. 1) are set in front of the x-ray streak camera. Also, the use of a KI photocathode optimized for multi-keV detection warrants that the signal measured with the x-ray streak camera is dominated by the K-shell emission.

The time resolved x-ray measurement is shown in Fig. 2. The x-ray temporal profile is essentially limited by the instrumental response, and unambiguously demonstrates a subpicosecond x-ray pulse duration. The cumulative mode offers a signal-to-noise ratio (SNR) reaching 2 orders of magnitude, which allows a correction of the instrument temporal response. This response has been demonstrated to be largely dominated by the residual jitter between the x-ray pulse and the sweeping ramps of the streak camera [22]. The streaked signal of an ultrashort UV pulse (obtained by the third harmonic generated from a 40 fs laser pulse) has been used to determine this jitter (i.e., the instrumental response $\Delta t_{\mathrm{instr}}$), with the same setup, and between each x-ray measurement. The corrected x-ray pulse duration Δt_{cor} is then deduced from the measured duration Δt_{mes} via a quadratic subtraction ($\Delta t_{\mathrm{cor}}^2 = \Delta t_{\mathrm{mes}}^2 - \Delta t_{\mathrm{instr}}^2$), considering that Δt_{cor} and $\Delta t_{\mathrm{instr}}$ are independent data. The measurement presented in Fig. 2 leads to $\Delta t_{\mathrm{cor}} = 290$ fs FWHM. A large error bar comes from the small fluctuations observed in the measurement of $\Delta t_{\mathrm{instr}}$, leading to an upper limit of 700 fs FWHM for the x-ray pulse duration.

Measurements of the x-ray duration have been performed as a function of the incident laser pulse duration. The results are plotted in Fig. 3 after correction of the instrumental response. From all these data, the x-ray duration is observed to generally follow the laser pulse duration. In this regime combining large clusters and subrelativistic laser intensities, no significant dependence is observed on laser polarization, as previously reported in time-integrated x-ray spectra measurements [20]. The x-ray intensity reaches a maximum value for an optimal laser pulse duration depending on the cluster size. With 275 Å average radius clusters, x-ray intensity reaches its maximal value with 400 ± 100 fs FWHM laser pulses. This is in very good agreement with the results reported in Ref. [23].

In order to present a scenario of the laser-cluster interaction as completely as possible, absorption measurements have been performed with the same setup. We used an integrating sphere to measure the laser energy absorbed in

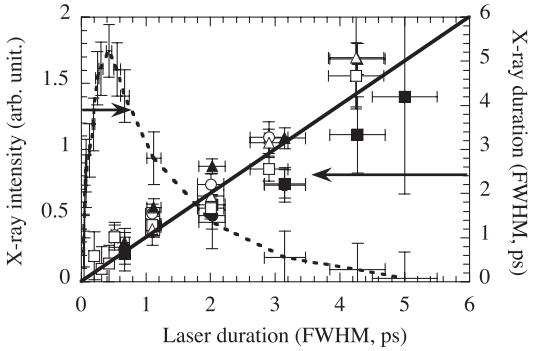

FIG. 3. X-ray emission duration measured as a function of the laser pulse duration. Ar clusters with 180 Å (circles), 275 Å (squares), and 350 Å (triangles) average radius are irradiated by 2.0 mJ, linearly (black) and circularly (white) polarized laser pulses. Full line: guiding line equating the x-ray pulse duration to the laser pulse duration. Dashed line: time-integrated K-shell x-ray emission from 275 Å Ar clusters.

the entire focal volume. The results are plotted in Fig. 4. The absorption is found to be the most efficient for laser pulses longer than a duration of a few 100 fs, increasing with the cluster average radius. This observation is consistent with previous observation [20] and with the nanoplasma models proposed first by Ditmire [16], then improved by Milchberg [24]. In these models, the laser absorption is enhanced during the cluster expansion when the electronic density becomes close to the critical density.

The plasma has been spatially resolved using shadowgraphy and x-ray pinhole diagnostics [25]. From the cluster density previously determined [21], we get the number of clusters contributing to the observed laser energy absorption. A maximum value of 15 ± 5 nJ is then deduced for the average absorbed energy per cluster (275 Å average radius) in a 6 μm plasma radius. Assuming that all of this energy is first absorbed by the electrons, and considering an average ionization state $Z^* = 15$ [20], a total energy of $T_{e0} = 3100 \pm 500$ eV per electron is deduced. This is

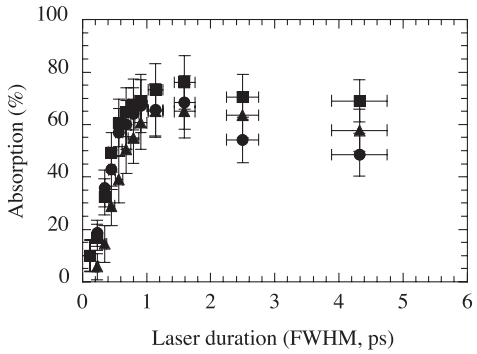

FIG. 4. Laser energy absorption measured in the cluster jet as a function of the laser pulse duration. Ar clusters with 180 Å (circles), 275 Å (squares) and 350 Å (triangles) average radius are irradiated by 2.0 mJ, linearly polarized laser pulses.

consistent with the observations previously reported by Ditmire [26] for Ar and deuterium clusters and by Zweiback [27] for deuterium clusters. This value, which is quite high in view of the modest laser intensity involved and compared to that observed with solid and foil targets, confirms the high efficiency laser-matter coupling with clusters.

A simple hydrodynamical model is proposed in order to reproduce the extremely short x-ray duration observed from laser-cluster interaction compared with laser-solid foil interaction. It is based on calculations previously proposed, that reproduce fairly well the time resolved x-ray spectra measured in laser-foil experiments [12]. The laser energy is supposed to be converted into electron thermal kinetic energy. This is described as an energy source term following the temporal profile of the laser pulse. Its temporal integration is set to the values of energy deposited deduced from the absorption experiment. The specific geometry of the target is taken into account, assuming a self-similar expansion of (i) a foil with initial thickness $2R_0$ (thin foil), and (ii) a sphere with initial radius R_0 (cluster). The plasma dynamics is driven by an adiabatic hydrodynamic expansion, described by three equations: (i) energy conservation, (ii) entropy conservation, (iii) kinetic pressure assumed to be dominated by its electronic component. Long after the interaction, the target dimension is found to evolve as $R_0 \sim c_{s0}t$, where c_{s0} is the sound speed deduced from the energy deposited per electron ($c_{s0} = \sqrt{Z^* k_B T_{e0}/m_i}$). We deduce the asymptotic temporal behavior of the electron density and temperature to be, respectively, $N_e \sim 1/t$ and $T_e \sim 1/t^{2/3}$ for foils. In the case of clusters, just due to spherical geometry, we get $N_e \sim 1/t^3$ and $T_e \sim 1/t^2$. This simple geometrical effect leads to a significantly faster expansion and cooling for clusters, that should shortens the x-ray emission duration.

To assess this picture, hydrodynamical calculations have been performed with 275 Å average radius Ar clusters as a function of the laser duration. A non steady-state collisional-radiative atomic physics code (TRANSPEC [28]) has been used as a postprocessor, in order to calculate the temporal behavior of the x-ray emission. The code includes all the ionization states with a detailed description of the H, He, and Li-like ionization stages. All of other stages uses a superconfiguration description of both the collisional and radiative rates and the supertransition arrays properties [29]. Time-dependent results have been spectrally integrated over the K-shell range and are displayed in Fig. 5. A very good agreement is found between these calculations and the measurements plotted in Fig. 3. A laser duration of 450 ± 100 fs FWHM is found to optimize the x-ray intensity. With shorter laser pulse, our absorption measurements show that the cluster heating is not efficient leading to poor x-ray emission. If the laser pulse duration is too long, then most of the absorbed energy is transferred in cluster expansion, limiting the temperature achieved and the subse-

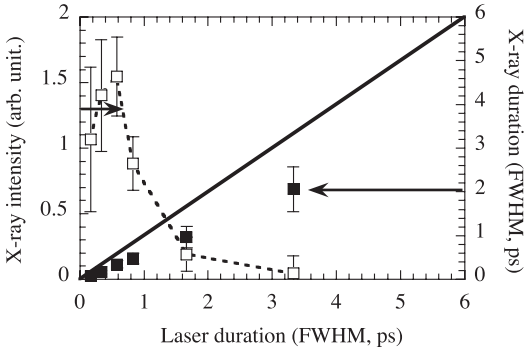

FIG. 5. Black squares: x-ray emission duration calculated as a function of the laser pulse duration from a 275 Å average radius Ar cluster. Full line: guiding line equating the x-ray pulse duration to the laser pulse duration. White squares and dashed line: time-integrated K-shell x-ray emission deduced from the calculation.

quent x-ray emission. The calculated x-ray pulse duration is found to increase linearly with the laser duration, but with a slightly smaller slope compared to experiment. Close to the optimal x-ray emission (i.e., with 320 fs FWHM laser pulse) an x-ray duration of 170 ± 40 fs FWHM is estimated (error bars come from the energy deposition measurement uncertainty).

In conclusion, we report the first experimental evidence of subpicosecond x-ray bursts produced by laser-cluster interaction in the multi-keV range. Large Ar clusters (average radius from 180 to 350 Å) have been irradiated by ultrashort (down to 40 fs) and intense subrelativistic laser pulses (up to 10^{17} W/cm^2). The K-shell x-ray emission ($\sim$3 keV) is selected from the intense XUV emission and measured with a 0.8 ps rms temporal response cumulative streak camera. The K-shell x-ray emission intensity is observed to be optimal with 400 ± 100 fs FWHM laser duration, reaching up to 3×10^8 photons per shot emitted in 4π sr. The correction of the instrumental response leads to a corresponding x-ray duration with an upper value as short as 700 fs FWHM. An hydrodynamic expansion model is proposed, based on laser energy absorption measurements and coupled with collisional-radiative calculations. Calculations reproduce the behavior of the x-ray emission with laser pulse duration, as well as the subpicosecond durations observed. They even suggest an x-ray duration as short as 170 ± 40 fs FWHM when the x-ray emission is optimal. This extremely short x-ray duration is understood as the combined effect of both the very efficient laser energy coupling with clusters and the ultrafast expansion (and cooling enhancement) of such spherical nanotargets. Since no high laser energy is required (a few mJ in a few 100 fs), this ultrashort multi-keV clean x-ray source can be operated with up to 10 kHz repetition rate commercially available lasers. It presents a great potential for applications in ultrafast x-ray science.

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