Evaluation of laser-irradiated Ar clusters as a source for time-resolved x-ray studies

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Evaluation of laser-irradiated Ar clusters as a source for time-resolved x-ray studies

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We have measured the absolute average photon flux, the spectral characteristics, and the time structure of x rays emitted from Ar clusters which were irradiated by a 100 fs laser with an intensity of 10^{17} W/cm^2. The measured photon flux was 10^7 photons per shot in the K_α (at 3 keV) line in a 4π sr solid angle. The temporal structure was measured using a streak camera with a 10 ps time resolution. It was found that less than 1% of the photons were emitted within the 10 ps time-response function of the streak camera. The emission profile is roughly exponential with a time constant of 3 ns. © *1999 American Institute of Physics.* [S0034-6748(99)03605-9]

I. INTRODUCTION

Over the last five years, high-intensity laser-irradiated van der Waals clusters have been the object of intense study.[1–11] The high average x-ray flux gives reason for optimism when it comes to using laser-irradiated clusters as a source for relatively hard x rays (>1 keV). Possible application fields are lithography[12] and x-ray microscopy. The interaction of intense laser radiation with clusters is now well understood through a model first described by Ditmire *et al.*[4] In this model each cluster is treated as a small plasma ball, a few nanometers in size. Therefore the model is sometimes called the nano-plasma model. The model can be used to predict measurable features such as electron spectra, ion spectra, and the scaling of the emitted x-ray intensity with laser pulse duration and laser wavelength.

Since the clusters in many of the studies are irradiated with short-pulse lasers (100 fs), the time structure of the x-ray emission has been a topic of discussion. Short-time emission (<1 ps) would be of particular interest for ultrafast visible pump/x-ray probe experiments. Such studies are currently being performed with a wide range of x-ray sources such as synchrotrons where the temporal characteristics are modified with short laser pulses (<100 fs),[13] laser/electron beam interactions,[14] and laser-produced plasmas from solid targets.[15,16]

Compared to synchrotrons and electron beam devices, the laser-based sources are cheap and compact. Compared to solid target x-ray sources or solid cluster targets such as gold black,[17] rare gas clusters produce little debris,[18] are easily renewable and are less sensitive to alignment. All of this is important for application purposes. Many of the above mentioned advantages are also true for liquid targets.[19,20] However, few experiments using short laser pulses (<1 ps) to produce x-ray emitting plasmas from liquid targets have been carried out[21] and the temporal structure of the emission has yet to be studied.

In the present study, we investigate the temporal structure of the K_α x-ray emission from Ar clusters following irradiation by 100 fs pulses from a Ti:Al_2O_3 laser.

II. EXPERIMENTAL SETUP

A schematic of the experimental setup is shown in Fig. 1. In the experiment we made use of the newly upgraded 100 fs Ti:Al_2O_3 chirped pulse amplification (CPA) laser at the Lund High Power Laser Facility. The laser currently produces pulses with a peak power of 6 TW at 10 Hz. In this study, the 50 mm diam beam was propagated through air. For this reason, the peak power was 1.5 TW in our experiments. The beam was focused using a parabolic mirror (f-number=1). The laser intensity in the focus was deduced from a separate time-of-flight measurement of Xe ions produced in the laser focus. During these measurements, the laser was attenuated by two orders of magnitude directly after the final amplifier. By comparing the intensity dependence of the measured Xe ion production rates with calculated electron tunneling rates,[22,23] we estimate the intensity in the laser focus to be 10^{17} W/cm^2 at our peak power.

Since in the actual experiment the beam was propagated through 3 m of air and 7 mm of glass (BK 7), intensity dependent effects, not present in the low-power intensity determination, might have distorted the temporal and spatial characteristics of the laser. The implication is that the extrapolation to high powers introduce a large error of about 50%, compared to the 10% error in the time-of-flight measurements. The contrast with respect to prepulses was studied with different techniques for different times relative to the main pulse. A diode was used to monitor prepulses arriving 0.5 ns or more before the main pulse. The contrast was found to be better than 10^{-5}. Measurements with a streak camera showed a contrast better than 10^{-3} for prepulses outside 10 ps from the main pulse. However a 1% postpulse was detected 300 ps after the main pulse. No prepulses or postpulses were observed using a second-order autocorrelator with a dynamic range of 10^{-2} in a 20 ps window around the main pulse. Clusters were produced by expansion of argon gas into vacuum. The technique and diagnostics are very similar to that described in detail by Ditmire *et al.*[4] A gas

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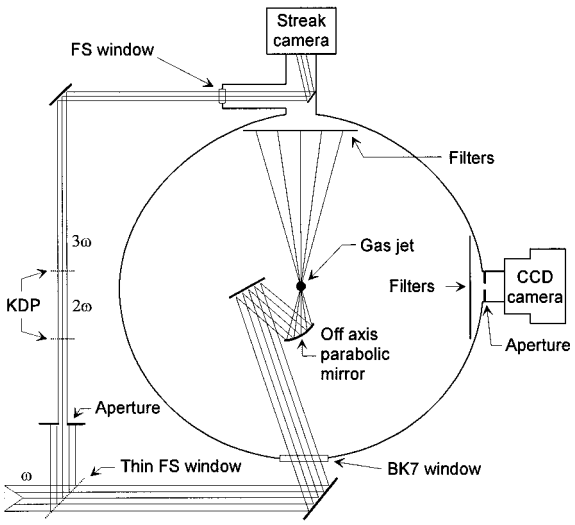

FIG. 1. Experimental setup.

nozzle allowing for backing pressures up to 80 atm was used. The diameter of the nozzle opening was 0.8 mm. The extent of clustering in an expanding gas jet can be estimated through the so-called Hagena parameter. It is given by the semiemperical formula

$$\Gamma^* = k \times [d/\tan(a)]^{0.85} \times p_0 / T_0^{2.29}.$$

The units for different parameters are the ones given below. The empirical constant $k=1700$ for Ar. The nozzle diameter was 500 μm, the pressure was 80 000 mbar. The temperature was 300 K and the cluster half angle was 15° yielding $\Gamma^* = 2\times 10^5$. For this hagena parameter a cluster size of 10^5-10^6 is expected. The onset of cluster formation was experimentally studied through the intensity of Rayleigh scattered laser light versus backing pressure. The same laser was used for this purpose. However, the laser intensity was attenuated and a 1 mm aperture was inserted. The power was measured to 10 μJ. The laser was focused in the gas jet to an intensity of about 10^{11} W/cm^2 and the scattered radiation was imaged onto a charge coupled device (CCD) camera. From the angular acceptance of the imaging lens and the number of electrons generated on the CCD chip the total number of scattered photons were estimated. The Rayleigh scattering cross section is

$$dS/d\Omega = 2\times \pi \times r^6/\lambda^4 \times (n^2-1)/(n^2+2)\,\text{cm}^2/\text{sr}.$$

We observed 30 000 photons in a 10^{-5} sr solid angle using a detector with 50% quantum efficiency. Inserting the refractive index of solid argon ($n=1.38$) and $\lambda=800$ nm yields a cluster size of 15 nm given a gas density of 10^{19} atoms/cm^3. This cluster size corresponds to about 10^5 atoms/cluster atoms per cluster taking the inter-atomic spacing to be the same as in solid argon. The gas density was estimated from a measurement of the rise of static pressure in the volume of the vacuum chamber for a fixed number of gas puffs. The duration of the gas puff was estimated through the Raleigh scattering experiment.

The streak camera cathode had an estimated 3% quantum efficiency and an acceptance angle of about 3 $\times 10^{-6}$ sr. Hence, the recorded x-ray emission was weak in the time-resolved measurements. The strongest x-ray emission was obtained using the highest backing pressure (80 bar), giving the largest clusters (10^5 atoms/cluster) and the highest density (10^{19} atoms/cm^3). These numbers are accurate to the order of magnitude.

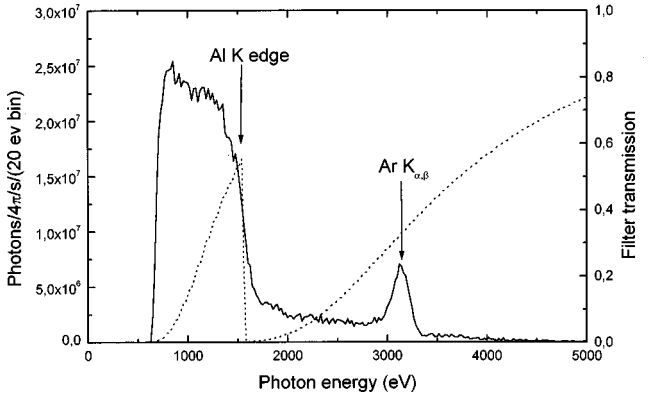

FIG. 2. Emission spectrum (solid line) as observed behind a filter. The photon count has not been modified to account for the filter transmission profile (dotted line).

The x-ray detectors were put behind filters in order to suppress stray light and soft x-ray emission. Two types of filters were used. Either a 6 μm thick Al filter or a combination of 6 μm Al and 140 μm plastic film. We measured the x-ray energy spectrum using a Peltier-cooled CCD camera using the fact that the number of electrons produced and stored in each CCD pixel corresponds to certain photon energy (3.6 eV/electron). The CCD chip was back thinned giving >90% quantum efficiency for 3 keV radiation. The x-ray temporal structure was measured with a commercial streak camera with a 120 nm thick solid density CsI photo cathode.

III. RESULTS AND DISCUSSION

The spectrum is shown in Fig. 2. As can be seen, the *K* edge from the Al filter is clearly visible in the thermal background radiation. The smearing of this edge compared to the calculated filter response (also shown in Fig. 2), yields an estimate of the spectral resolution to about 200 eV. From the average number of photons per pulse we determine the absolute flux of Ar *K*-shell radiation to be 10^7 photons per pulse at 10 Hz in a 4π sr solid angle.

As seen in Fig. 2, a substantial amount of the radiation originated from the continuous background with temporal emission characteristics that might differ from the line emission. In order to filter out the thermal radiation, a plastic filter was inserted. The emission spectrum as detected behind this filter combination is shown in Fig. 3. The spectrum is clearly dominated by the *K*-shell emission. From the recorded spectra we estimate that 50% of the radiation arriving at the detector came from emission near the K_α line. The rest was emission near the K_β line radiation and incoherent brehmsstrahlung. Multiple pulses had to be accumulated to get sufficient photon statistics.

In Fig. 4 we show the emission characteristics over several nanoseconds. The data consist of 100 accumulated pulses. This recording was made using the slowest sweep of

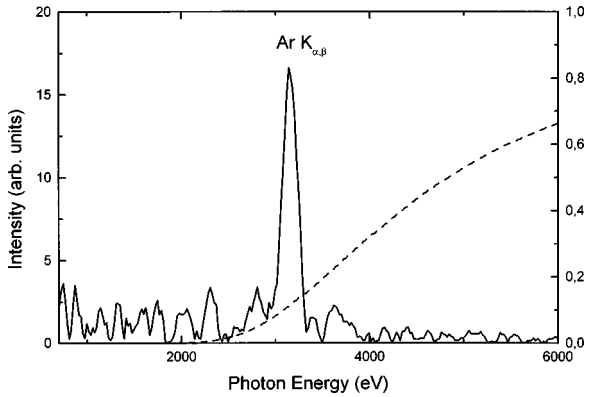

FIG. 3. Emission spectrum (solid line) as observed behind the filter combination used in the time-resolved measurements. The calculated filter transmission profile is also shown (dotted line).

the streak camera (700 ps/mm). The streak camera response to a 100 fs pulse obtained by frequency tripling of the 800 nm laser is also shown. The temporal response is limited by the slit size as well and the trigger jitter.

The remedy for the influence of the slit size was to increase the sweep speed, this, however, made the signal appear substantially weaker. To avoid trigger jitter in the accumulation, a timing fiducial, obtained from the third harmonic of the 800 nm laser and a photon-counting algorithm, was used. The principle for this technique is described in Ref. 24. However, the temporal resolution in our study was limited by the fact that already at a sweep speed of 30 ps/mm, we found that few photons arrived within the window of detection. A recording of 500 averaged shots at this sweep speed is shown in Fig. 5.

The long duration of the x-ray emission indicates that cooling predominantly occurs through expansion of the cluster, as three-body recombination is expected to show a faster time dependence.[4]

Due to flux limitations, it was not possible to use a high resolution crystal spectrometer in combination with a temporally resolving detector. Although the filter could make the *K*-shell emission dominate the spectrum, we were not able to resolve emission from the different charge states, which, due to different screening potential, would be separated in wavelength.

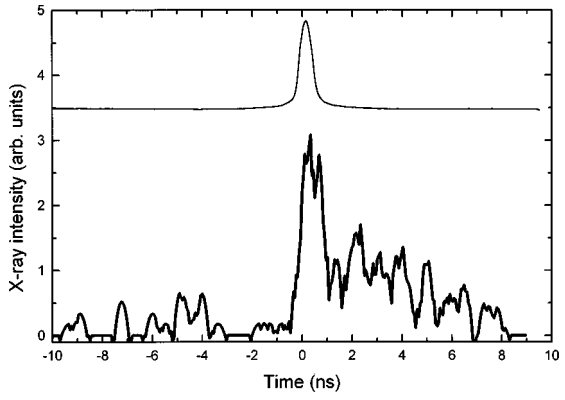

FIG. 4. Time-resolved emission from clusters as detected by a streak camera. The time response from a series of 100 fs UV pulses is also shown. The data for both the x-ray and UV radiation consist of 100 averaged pulses.

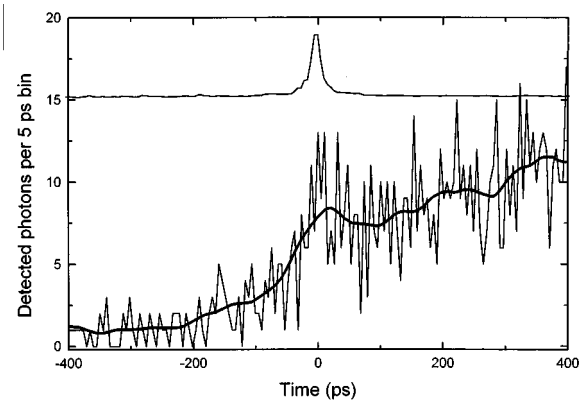

FIG. 5. The early time history of the emission from the cluster target studied by a streak camera in photon-counting mode. The number of averaged pulses is 500. The single-shot time response to a 100 fs UV pulse is also shown.

From our measurements, we cannot rule out that x rays from highly ionized Ar ions are emitted during a shorter time than the average. Recombination could quench the emission from high charge states. An interesting experiment that would enable the measurement of the emission duration from different charge states would be to use time-resolved x-ray diffraction. The monochromator crystal would then be melted by a short pulse laser with a variable delay relative to the x rays. The emission from different ionization stages would be spatially separated on a position sensitive detector. The temporal structure of the emission could be deduced from the registered intensity as function of optical delay in a cross-correlation measurement similar to that described in Refs. 13 and 15. However, to successfully carry out such an experiment, a higher x-ray flux must be obtained.

We find that, although x-ray radiation from nano-plasma clusters shows promise for time-integrated applications, the long pulse duration prevents time-resolved probing of ultrafast processes.

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