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Larsson, Jörgen

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J. Larsson

*Department of Physics, Lund Institute of Technology, P.O. Box 118, S-221 00 Lund, Sweden, and
Division for Synchrotron Radiation Research, Lund University, P.O. Box 118, 5-22100 Lund, Sweden*

A novel method developed to increase the temporal resolution of x-ray streak cameras is described. The method is analogous to the time-correlated single-photon-counting technique, which is commonly used in atomic physics. By use of short-pulse x-ray radiation from a laser-produced plasma, generated by an ultrafast laser, it is shown that a standard x-ray streak camera with a nominal temporal resolution of >5 ps can yield a temporal response of 1.6 ps. The readout technique also removes temporal jitter with respect to the triggering laser. Capabilities and limitations of the technology are discussed.   © 2001 Optical Society of America

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Ultrafast x-ray studies is a field that is attracting increasing attention[1–7] for applications such as studies of ultrafast melting and coherent phonons in semiconductors. There are two different approaches to performing such experiments. One is to use an ultrafast x-ray pulse in a pump–probe configuration.[1–6] The other is to use a longer pulse from a synchrotron radiation beam line,[7,8] together with an ultrafast detector, e.g., a streak camera. State-of-the-art temporal resolution of x-ray streak cameras can be 500 fs in single-shot operation.[9] If the x-ray pulses are weak but frequent, low jitter can be obtained by use of a photoconductive switch as a trigger for the sweep voltage of the streak camera. Integration times of several minutes and a temporal resolution of 900 fs have been reported[10] for high-repetition-rate data collection.

In this Letter the possibilities of completely eliminating trigger jitter and of increasing temporal resolution beyond single-shot temporal resolution by at least a factor of 3 are demonstrated. The technique is demonstrated by use of a streak camera with a nominal temporal resolution of 5 ps and a jitter of ~40 ps with respect to the laser. However, the present technological advance could easily be implemented with a streak camera with a better nominal temporal resolution. If this technique were employed with the streak camera described in Ref. 9, one could expect a temporal resolution of less than 200 fs in accumulation mode. Single-photon counting is based on the implementation of two distinct elements: First, timing fiducials are used to correct for timing jitter. Second, single x-ray photons are imaged on the readout camera, and their centers of gravity are accurately determined. Hence the arrival of an x-ray photon can be accurately determined with respect to the timing fiducial. The technique is demonstrated by determination of the temporal structure of x-ray emission from a plasma produced by a laser with a 100-fs duration. The method was suggested by Murnane *et al.*[11] but is implemented here for what is believed to be the first time.

In the current experiment an x-ray pulse was generated by irradiation of a stainless-steel target with a laser pulse with a 100-fs temporal resolution. The laser provided 100-mJ pulses at a repetition rate of 10 Hz. The laser beam was focused by an off-axis parabola to an intensity estimated (within a factor of 2) to be 5×10^{17} W/cm^2. The laser was incident at an angle of 30° to the normal with p-polarization. The x-rays were extracted at an angle of ~70° relative to the normal. This angle was chosen because of geometrical constraints dictated by the vacuum chamber and the debris shield of the laser-plasma source. A beam splitter split off a fraction of the energy in the laser pulse and was used to generate UV radiation for the two timing fiducials. The experimental setup is illustrated schematically in Fig. 1.

Because of a relatively low x-ray flux, the experiment was performed without an x-ray monochromator. The streak camera (Kentech) was positioned at a distance of 80 cm from the source. The spectrum of the emitted radiation is shown in Fig. 2. The detected signal consists of broadband bremsstrahlung and the labeled characteristic radiation. The data in Fig. 2 were registered with a CCD camera by use of pulse-height analysis. The detector was isolated from the plasma generation chamber by two beryllium windows.

For the time-resolved experiment a 120-nm-thick CsI photocathode was used, together with a 70-μm-wide 2-cm-long slit, yielding a solid angle of 10^{-4} sr. The narrow slit width was chosen so that the broadening owing to the finite width of the slit, imaged onto the phosphor, would be kept below 700 fs. The narrow slit, together with a quantum efficiency of ~0.1%, means that of the order of one x-ray photon per pulse was registered. The readout camera was limited to a 3-Hz repetition rate.

A recorded image is shown in Fig. 3. Two timing fiducials with a known spacing (in this case ~300 ps) can be seen, together with two x-ray photons and intensifier noise. Each such image is analyzed by use of software, following the protocol described below. First a lineout is made along the horizontal direction to identify and determine the positions of the fiducials. Then all spots in a region between the two fiducials
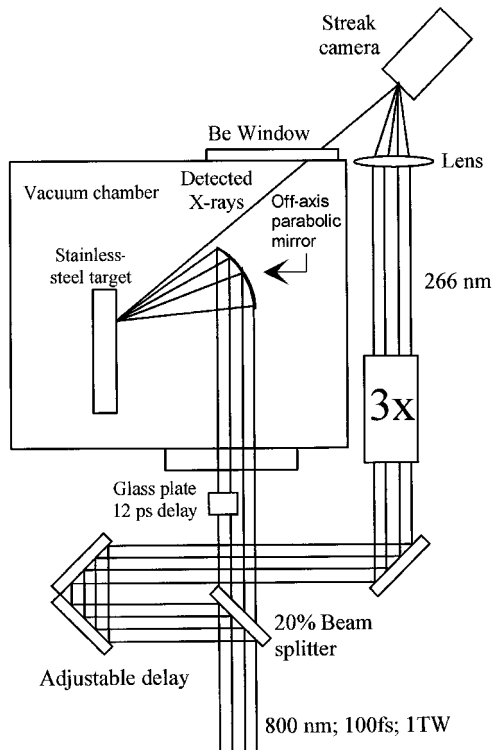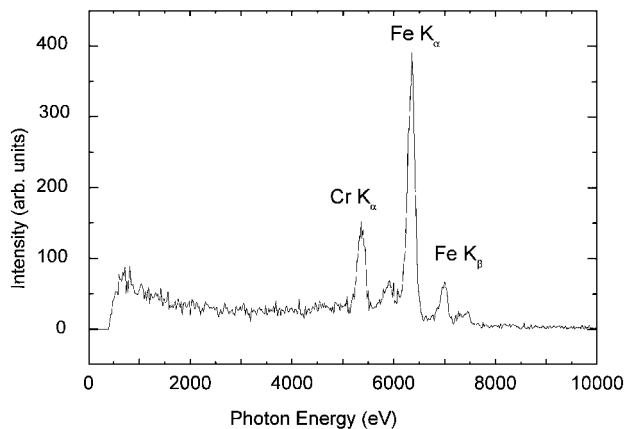
Fig. 1.   Experimental setup.

Fig. 2.   Spectrum of the laser-driven plasma source.   The intensity corresponds to the number of photons per pulse in a 4π sr solid angle.

intensity over the larger area are counted as real photons, whereas the others are discarded as noise.

The effect of employing the readout technique can be seen by comparison of Figs. 4 and 5. The single-shot resolution is determined from a trace of the intensity across the fiducials, which yields a FWHM of 5.5 ps and a 10–90% rise time of 6.5 ps. It should also be noted that jitter of ~40 ps would be present if multiple images were added. Counting the photons in approximately 1000 images yields a single jitter-free image. The temporal resolution is improved by a factor of 3 compared with single-shot resolution. A lineout of the central portion of the image reveals a rise time of ~1.6 ps (1.6 ps for the first pulse, and 1.0 ps for the second). The difference in rise time is believed to be due to noise. The 1.0-ps risetime of the second peak is obtained after subtraction of the base level of ~0.2 units, which is due to the first pulse. (Note that using the central part of the image is essential, as there is a substantial trace curvature that is due to the different path lengths of the photoelectrons generated on different parts of the photocathode.) The exponential fall of the recorded plasma radiation has a decay constant of 3.5 ps. This constant is attributed to the temporal
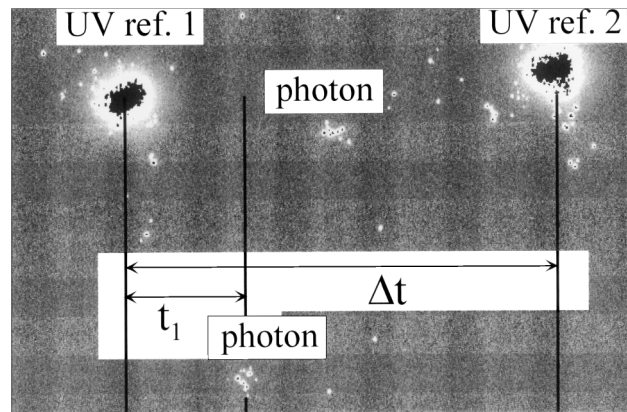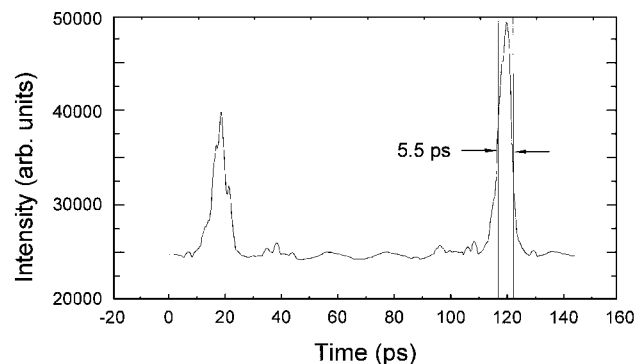

Fig. 3.   A single shot as captured by the readout camera. In this image the two timing fiducuals (t_1 and Δ_t) can be seen, together with two x-ray photons. The contrast of the image has been modified to improve the quality of the printed version.

(horizontally) and below them are found. Spots with a high enough intensity and a large enough spatial extent are registered as photons. The criterion on spatial extent is what separates the photons from the intensifier noise and is crucial for long integration times. As can be seen from Fig. 3, there are several isolated smaller spots that are due to intensifier noise but only two clusters of spots that are real photons. The computer algorithm that identifies photons is implemented in two steps. A simplified description is given below. First, all spots with high enough intensity are identified as potential photons. Then the intensity over a larger area (in this case 10×10 pixels) around each spot position is calculated. The potential photons that have a high enough integrated

Fig. 4.   Nominal single-shot temporal resolution of the streak camera was determined by measurement of the FWHM of UV pulses generated by frequency tripling of the radiation from the femtosecond laser.
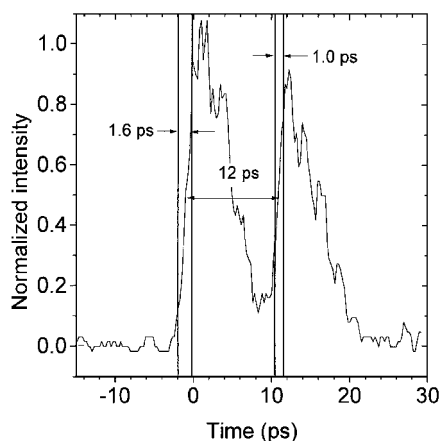
Fig. 5. After averaging of 1000 laser shots, the temporal structure of the laser-plasma source can be seen. The double-pulse structure depends on the fact that two laser pulses with a separation of 12 ps interact with the target. The temporal resolution of the system is deduced from the 1.6-ps rise time of the registered temporal shape of the emitted radiation from the plasma.

characteristics of the laser-produced plasma source. Work is currently in progress to shorten the emission time of the source. Owing to the fact that we now have a source that provides a pulse with a sharp rise and a slow fall, we determine the temporal resolution as the 10–90% rise time of the detected signal. With this criterion, the temporal resolution of the detector was found to be 1.6 ps.

When a streak camera is designed, one must take into account a number of considerations to ensure the optimum temporal response. Only some of the mechanisms for temporal smearing are compensated for by the single-photon-counting readout presented here. Collective effects owing to spreading of the ensemble of secondary electrons, such as imperfect focusing and smearing owing to the velocity spread of secondary electrons, are canceled. Statistical processes involving single x-ray photons or primary electrons cannot be canceled. Such an effect is the broadening that is due to the finite width of the slit. The position where a single photon hits the photocathode will influence the position at which center of gravity of the ensemble is imaged. A mechanism that may ultimately be the limit of the temporal resolution of streak cameras has not been an issue so far. It will take a finite time for the primary electrons with a kinetic energy corresponding to the x-ray photons to be converted to secondary electrons, which then will escape from the cathode in a diffusive process. Since this stage involves statistics of the primary as well the secondary electrons, this effect will be only partially improved on by the single-photon-counting

technique described here. The readout camera was limited to a 3-Hz repetition rate. However, there exist slow-scan, intensified CCD cameras that allow for a full-frame readout at 40 Hz. To go beyond that to the kilohertz regime, one must use another type of readout camera. Over the past 5–10 years camera chips, often based on complementary metal-oxide semiconductor technology, have integrated on-chip image processing capabilities. One can foresee that cameras based on such a technology could be used to bring single-photon readout into the kilohertz regime.

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