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# Ultrafast x-ray pulse generation by focusing femtosecond infrared laser pulses onto aqueous solutions of alkali metal chloride

Koji Hatanaka, Toshifumi Miura, and Hiroshi Fukumura<sup>a)</sup>

*Department of Chemistry, Graduate School of Science, Tohoku University, Sendai 980-8578, Japan*

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X-ray pulses were generated by focusing femtosecond infrared laser pulses (775 nm, 130 fs, 0.58 mJ/pulse) onto an alkali metal (Cs or Rb) chloride aqueous solution jet. The mechanism of x-ray pulse generation was studied by measuring x-ray emission spectra and transmitted laser pulse spectra. Addition of CsCl to distilled water was found to enhance markedly x-ray intensity and x-ray cutoff energy. The frequency-up-shift observed in transmitted laser pulses was larger in CsCl solution than in distilled water. The results suggested that a high-density plasma-like medium was produced in CsCl solution due to secondary electrons. © 2002 American Institute of Physics.  
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There is now a growing interest in x-ray pulse generation from femtosecond laser-induced plasmas not only for the study of the interactions between intense laser fields and matters but also for applications such as time-resolved x-ray diffraction<sup>1</sup> and attosecond pulse generation.<sup>2</sup> To date, however, targets for the x-ray generation have been limited mostly to metals and gases. Very recently, there appeared some reports on femtosecond laser-induced x-ray pulse generation from solution targets such as carbon fluoride,<sup>3</sup> ethanol,<sup>4</sup> water,<sup>5</sup> a copper nitrate aqueous solution,<sup>6</sup> and ethylene glycol.<sup>6</sup>

We consider that a solution is an appropriate target since mechanisms of the x-ray generation can be discussed by changing solute compounds, their combinations, and their concentrations, which is different from the case of metal targets. Additionally, for practical applications, solution targets can be recycled and circulated by a pump, so that high stability is expected for long time experiments. In addition, x-ray pulse width may be controllable by changing solute concentrations since the lifetime of high-energy electrons might be short in highly concentrated aqueous solutions.<sup>7</sup>

In this letter, distilled water and alkali metal (Cs or Rb) chloride aqueous solutions are chosen as samples and x-ray pulses are generated in air by focusing femtosecond laser pulses. Both x-ray emission spectroscopy and spectroscopy of transmitted excitation laser pulses are carried out for the study of the interaction between intense laser field and conductive electrons in solution. Alkali metal chloride is a compound composed of elements with high atomic numbers and its solubility to water is high ( $\sim 6 \text{ mol/dm}^3$ ).

Figure 1 shows a top view of the experimental setup. Distilled water or a highly concentrated alkali metal (Cs or Rb) chloride ( $>98\%$ , Aldrich) aqueous solution was circulated by a pump through a flat glass nozzle of which the inner gap was about  $100 \mu\text{m}$ . Those solutions are transparent to the excitation laser light at 775 nm. Femtosecond laser pulses (Clark MXR, CPA-2001, 775 nm, 130 fs, 1 kHz) were focused by an objective lens (Mitsutoyo, M Plan Apo  $10\times$ ,

$\text{NA}=0.28$ ) onto a solution jet surface from the nozzle with an incident angle  $\sim 45^\circ$  to the jet surface (S polarization). X-ray emission spectra were measured with a Ge solid state detector (Ge SSD, EG & G Ortec, GLP-25440-S) with a  $250\text{-}\mu\text{m}$ -thick Be window. The distance between the laser focus and the Ge SSD was 150 mm. In order to decrease x-ray intensity, a Pb plate (1 mm thick) with an aperture (1 mm in diameter) was set in front of the Ge SSD. Signals from the Ge SSD were processed by a multichannel analyzer (MCA/PC98B, Laboratory Equipment Corp.). Spectroscopy for excitation laser pulses transmitted through a solution jet was also performed with a spectrometer (Hamamatsu Photonics, K. K., PMA-10). All experiments in this study were performed under atmospheric pressure at 294 K.

Figure 2 shows normalized x-ray emission spectra from jets of distilled water, a CsCl aqueous solution ( $6.5 \text{ mol/dm}^3$ ), and a RbCl aqueous solution ( $6.0 \text{ mol/dm}^3$ ). The excitation laser intensity was 0.58 mJ/pulse. In the case of distilled water, a broad spectrum was observed with a tail up to  $\sim 15 \text{ keV}$  ( $\sim 0.09 \text{ nm}$ ). The intensity degradation in the lower energy region was due to the absorption effect. In the case of CsCl and RbCl aqueous solutions, in addition to a broad spectrum with a gentler slope, sharp x-ray lines were also observed clearly which were assigned to the following

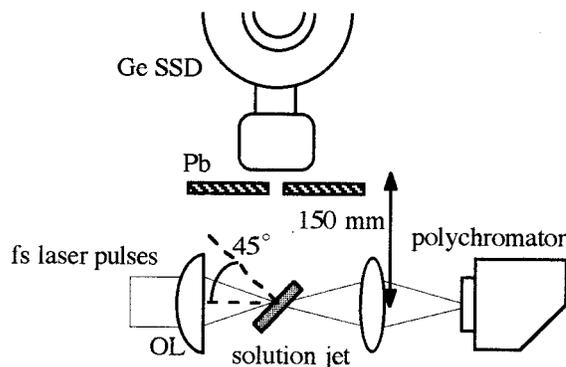


FIG. 1. Top view of experimental setup for x-ray pulse generation with solution jets, x-ray spectroscopy, and transmitted excitation laser pulse spectroscopy. OL: an objective lens ( $\text{NA}=0.28$ ). Pb: a lead plate (1 mm thick) with an aperture (1 mm diameter).

<sup>a)</sup>Author to whom correspondence should be addressed; electronic mail: fukumura@orgphys.chem.tohoku.ac.jp.

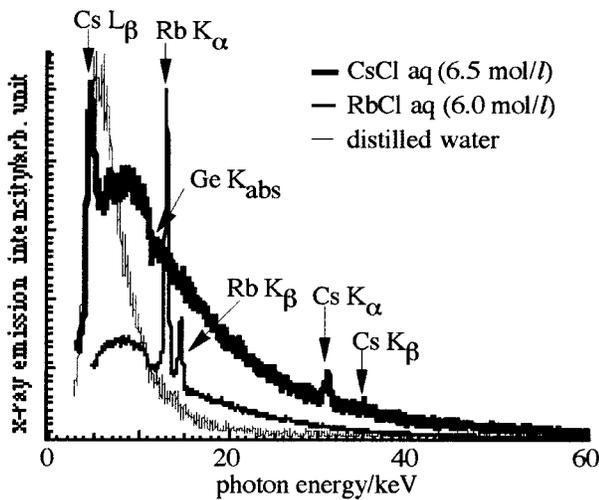


FIG. 2. X-ray emission spectra of distilled water and alkali metal (Cs or Rb) chloride aqueous solution irradiated by focused femtosecond laser pulses (775 nm, 130 fs, 0.58 mJ/pulse, 1 kHz). X-ray emission intensities are normalized by their maxima.

characteristic x-ray lines: Cs  $K\alpha$  (30.9 keV), Cs  $K\beta$  (34.9 keV), Cs  $L\beta$  (4.61 keV, 4.93 keV), and Rb  $K$  (13.3 and 14.9 keV). The observation of these high-energy  $K$  lines certifies that the measurement condition is free from the pileup effect, which is characteristic to solid state detectors.<sup>8</sup> Characteristic x-ray lines of Cl ( $K\alpha=2.62$  keV,  $K\beta=2.81$  keV) are out of the detection range. The depression observed commonly in all the spectra at  $\sim 11$  keV are due to the Ge  $K$  absorption edge (11.1 keV). Energy conversion efficiency of laser pulse to x-ray pulse in the range 3–60 keV, in the case of a CsCl solution (6.5 mol/dm<sup>3</sup>), was calculated to be  $\sim 10^{-8}$  under the assumption that x-ray radiation was spherically homogeneous.

X-ray emission spectra of a CsCl aqueous solution (6.5 mol/dm<sup>3</sup>) with different laser intensities are shown in Fig. 3. The spectra are corrected by considering the absorption effect of air, the Be window, and the Ge absorbing layer. As the laser intensity increased, x-ray emission intensity increased, the slopes of broad spectral components became less steep. The slopes can be analyzed quantitatively by using an

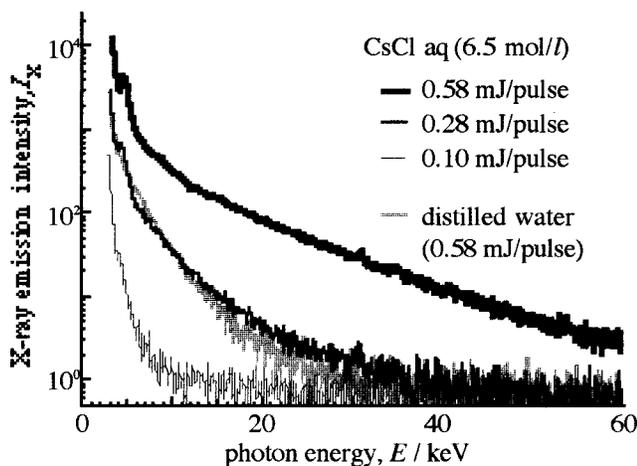


FIG. 3. X-ray emission spectra of CsCl aqueous solution (6.5 M) with different laser intensities and an x-ray emission spectrum of distilled water with 0.58 mJ/pulse laser intensity. The spectra are corrected by absorption effect of air, Be input window of Ge SSD, and Ge absorbing layer.

equation,  $I_x(E) = \exp(-E/T) \times \text{const}$ , where  $I_x$ ,  $E$ , and  $T$  represents x-ray intensity, x-ray energy, and the slope parameter, respectively. The parameter  $T$  for distilled water was calculated to be 2 keV, while  $T$  for a CsCl solution was 8 keV even with the same laser intensity. Among the spectra of CsCl solution with different laser intensity, the parameter  $T$  increased from 1 and 3 to 8 keV nonlinearly as the laser intensity increased from 0.10 and 0.28 to 0.58 mJ/pulse.

Laser power at the focus is calculated to be  $\sim 2 \text{ PW/cm}^2$  from the focus spot size ( $\sim 17 \mu\text{m}$ ), estimated by plasma emission photography in air (not shown). As for the ionization mechanism in this intense laser field, it may involve not only multiphoton absorption but also tunneling ionization by atomic potential distortion.<sup>9</sup> The Keldysh parameter<sup>10</sup> is calculated from the values of ionization potential of H<sub>2</sub>O and Cl<sup>-</sup>, indicating that the tunneling ionization is the major process for initial ionization. In general, electron acceleration is considered to occur through ponderomotive forces when metal targets were used.<sup>11,12</sup> In this study, however, the ponderomotive potential of laser field is calculated to be only 110 eV, which is much lower than the electron temperature obtained from the slope in Fig. 3. Thus, we must invoke other mechanisms such as inverse bremsstrahlung and/or stimulated Raman scattering to explain the observed high electron temperature. The confirmation of the mechanism details is now under consideration.

Broad x-ray emission spectra can be a result of recombination between electron and ionic species and/or bremsstrahlung. Comparing the x-ray emission spectrum of CsCl aqueous solution with that of distilled water, we found that x-ray intensity was higher in CsCl solution than in distilled water. This x-ray intensity enhancement by adding electrolyte with high atomic number elements is reasonable because recombination or scattering cross section is much larger in Cs<sup>+</sup> or Cl<sup>-</sup> than in H<sub>2</sub>O. However, the higher  $T$  in the CsCl solution compared to the distilled water cannot be explained only by the atomic number effect. Although it is difficult to clarify the mechanism of x-ray emission at present, it is plausible that nonlinear interaction between electrons and ions under intense laser field are responsible for the enhancements of x-ray intensity and x-ray cutoff energy observed in CsCl solution.

Figure 4 shows spectra of transmitted laser pulses through the CsCl solution jet with different laser intensities and a spectrum of distilled water. Compared to the original spectrum of laser pulse, spectra of laser pulses transmitted through the two solutions become broader and contain frequency-up-shifted components. The amount of frequency-up-shift is larger in CsCl solution than in distilled water. Among the spectra of CsCl solution, the frequency-up-shift is larger as the laser intensity becomes higher. Similar frequency-up-shift of femtosecond laser pulses have been observed in gases by Downer *et al.*,<sup>13</sup> while to our knowledge few studies with solutions have been reported. They reported that free electrons are generated through tunneling ionization at the leading edge of a laser pulse. Consequently, plasma is produced during a laser pulse. The refractive index of plasma is lower and expressed as follows;

$$n_p(t, x) = \{1 - [N(t, x)e^2 / m\omega_L^2]\}^{0.5}, \quad (1)$$

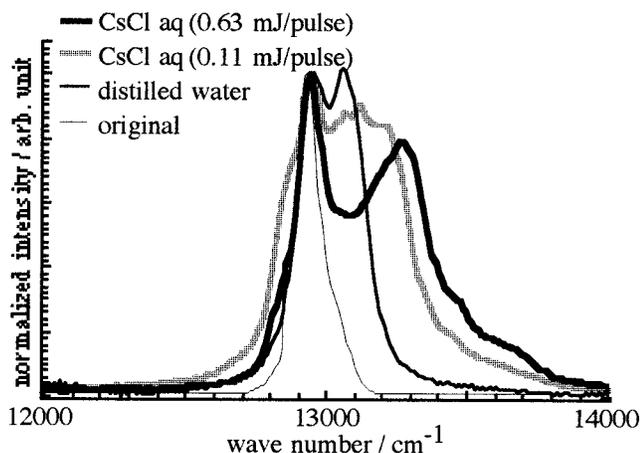


FIG. 4. Spectra of transmitted excitation laser pulses through CsCl solution (6.5 mol/l) or distilled water irradiated by 0.63 mJ/pulse laser pulses.

where  $n_p$ ,  $N$ ,  $t$ ,  $e$ ,  $m$ , and  $\omega_L$  represents a refractive index of a plasma, plasma density, time, elementary electric charge, electron mass, and angular frequency of a laser pulse, respectively. A laser pulse experiences a frequency-up-shift due to a change of the refractive index of a plasma generated by the laser pulse itself, because a phase velocity of laser pulse increases in a medium with a low refractive index. The amount of frequency-up-shift can be calculated by integrating a time-dependent refractive index. From the discussion above, the frequency-up-shift observed reflects long interaction length and a fast decrease of refractive index in the plasma wave front, in other words a fast increase of electron density. Therefore, the larger frequency-up-shift observed in the CsCl solution with the higher laser intensity indicates that faster rise of electron density occurs in the CsCl solution. In the case of CsCl solution, conductive electrons can be produced not only through tunneling ionization but also through accelerated-electron impacts on ions and molecules because the cross section of electron collision is larger than distilled water. Conductive electrons produced in this way

will absorb laser energy effectively through inverse bremsstrahlung. This may be the reason why the x-ray intensity increased and its cutoff energy extended to the higher energy region in the CsCl solution. In order to study mechanisms of ionization and electron acceleration leading to x-ray emission, we are performing experiments with solutions with a variety of conditions such as excitation laser energy, solute concentration, and compounds of different atomic numbers.

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