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Time-Resolved Investigation of Ionization of Clusters Irradiated by a Strong Laser *

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Ionization dynamics of clusters irradiated by chirped femtosecond lasers is investigated by using a linearly chirped pulse spectral interferometry with a time resolution of less than 100 fs. The production of an average charged $\sim Xe^{18+}$ and $\sim Kr^{9+}$ ions indicates a strong energy coupling between laser and cluster. Ultrafast depletion of the probing laser is observed to be strictly coincident with the ionization front as seen in other experiments. Moreover, a two-step ionization process for Xe and Kr clusters irradiated by high-intensity lasers has been observed, which implies the role of resonance enhancement during the cluster explosion.

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The knowledge of ultrafast dynamics of gas atoms, solids and clusters irradiated by a strong laser field is important in theory and experiment to understand the mechanism of laser-matter interaction such as the evolution of electron density, the dissembling of clusters, and nonlinear effects. Time-frequency domain techniques such as FROG, MI-FROG and spectral interferometry (SI) have been employed in many experiments to measure ultrafast ionization dynamics of laser-irradiated gas,[1] and laser-driven wakefields.[2] Kim *et al.* have recently developed the SI by using a supercontinuum light source as a probe to investigate the ionization process of helium and the explosion of Ar clusters.[3−5]

Many researchers have studied the intense laser interaction with clusters by characteristics of generation of x-ray,[6] high-energy ions and electrons,[7−9] and nuclear fusion.[10] Several models have been developed to explain the manner in which a cluster ionizes, dissembles and explodes when it is irradiated by a high-intensity laser,[11−14] but there is still much uncertainty over how these phenomena occur. The resonance absorption and enhancement of laser field at critical density regions proposed by these models has attracted more interests but not yet observed directly. In this Letter, for the investigation of ultrafast dynamics in laser-cluster interaction, a linearly chirped pulse spectral interferometry has been employed to investigate the ionization of clusters, and the time-resolved depletion of the probe pulse.

In our experimental scheme, the probing and pump pulses were stretched to several hundreds of femtoseconds from an unchirped pulse laser with time duration of 40 fs. The unchirped laser pulse can be assumed to be a Gaussian pulse with a central frequency of ω_0 and pulse duration τ_0, $E(t) = \exp(-a_0 t^2 + j\omega_0 t)$, and $\tau_0 = \sqrt{2\ln 2/a_0}$. It is assumed that the stretching fold is N, the linearly chirped pulse laser may be written as $E_1(t) = \exp(-at^2 + j(\omega_0 t + bt^2))$, and its pulse duration $\tau = \sqrt{2\ln 2/a} = N\tau_0$, $2b$ is the chirp rate, and $b = a\sqrt{\tau_1^2/\tau_0^2 - 1} \approx Na$. When the chirped probe laser traverses through laser-driven plasma, the transmitted probe pulse is given by

$$E_2(t) = E_1(t) \times T(t) = E_1(t) \times G(t)\exp(-\mathrm{i}\Delta\phi(t)), \tag{1}$$

where $G(t)$ corresponds to the absorption, and $\Delta\phi(t) = 2\pi(n(t) - 1)z/\lambda_0$ corresponds to the phase shift impressed upon the probe laser by electrons, $n(t) = \sqrt{1 - n_e(t)/n_{cr}} \approx 1 - n_e(t)/(2n_{cr})$ is the refractive index of plasma with critical density n_{cr}, z is the length of plasma the pulse has traversed, and λ_0 is the central wavelength of the probe laser. The interferogram in frequency domain between the reference pulse $E_1(t)$ and the τ-delayed probe pulse $E_2(t-\tau)$ is given according to

$$\begin{aligned} I(\omega) = &|\tilde{E}_1(\omega)|^2 + |\tilde{E}_2(\omega)|^2 \\ &+ 2|\tilde{E}_1(\omega)\tilde{E}_2(\omega)|\cos(\omega\tau + \Delta\varphi(\omega)), \end{aligned} \tag{2}$$

$\Delta\varphi(\omega)$ is the measured spectral phase difference between the reference and probe pulses. A full Fourier extraction[3] can be used to extract the temporal phase shift and amplitude of the probe pulse

$$\Delta\phi(t) = \mathrm{Im}\ln\left\{\left[\int \tilde{E}_2(\omega)\exp[j\varphi_1(\omega)\right.\right.$$

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$$+ j\Delta\varphi(\omega) - j\omega t]d\omega\Big] \cdot \Big[\int \tilde{E}_1(\omega)\exp[j\varphi_1(\omega) - j\omega t]\mathrm{d}\omega\Big]^{-1}\Big\}, \quad (3)$$

where $\varphi_1(\omega)$ is the spectral phase of the reference. $\eta = I_{\mathrm{pro}}/I_{\mathrm{ref}}$ is defined as the intensity depletion coefficient, which includes the absorption, reflection and scattering of probe laser. The time resolution is $\Delta t \approx 2\pi/\Delta\omega$, and $\Delta\omega$ is the full spectral width of the probe laser. For our laser with a full spectral width of 48 nm, the time resolution is about 50 fs.

On the other hand, a more direct approach which includes blueshift can be used to extract the temporal phase shift. The spectrum of the probe pulse is given by

$$\tilde{E}_2(\omega) = \int \exp\Big[-\frac{1}{4}\frac{a}{a^2+b^2}(\omega_a-\omega')^2 - j\frac{1}{4}\frac{b}{a^2+b^2}(\omega_a-\omega')^2\Big]\tilde{T}(\omega')\mathrm{d}\omega', \quad (4)$$

where $\omega_a = \omega - \omega_0$, $\tilde{T}(\omega)$ is the spectrum of $T(t)$ in frequency domain. By considering that $T(t)$ is slowly varied compared with the unchirped laser pulse, the amplitude term in the integral can be placed outside, and the above equation is given as

$$\tilde{E}_2(\omega) \approx \tilde{E}_1(\omega)\int T\Big[\frac{1}{2}\frac{b}{a^2+b^2}(\omega_a-\omega')\Big] \cdot \exp\Big(-j\frac{1}{4}\frac{b}{a^2+b^2}\omega'^2\Big)\mathrm{d}\omega', \quad (5)$$

where $\frac{1}{2}\frac{b}{a^2+b^2}\omega_a = A\omega_a = t(\omega)$ corresponds to the frequency-time mapping relation. The phase shift $\Delta\phi(t-t')$ can be expanded in Taylor series at t

$$\Delta\phi(t-t') = \Delta\phi(t) - \frac{\partial\Delta\phi}{\partial t}t' + \frac{\partial^2\Delta\phi}{\partial t^2}t'^2 + \cdots \quad (6)$$

If the first three terms are included in Eq. (6), the spectrum of the probe laser may be written as $\tilde{E}_2(\omega) = \tilde{E}_1(\omega) \times \tilde{G}(\omega)\exp(-j\Delta\varphi(\omega))$ and

$$\Delta\varphi(\omega) \approx \Delta\phi(t(\omega)) + \frac{1}{2}(A\partial\Delta\phi/\partial t)^2/(1-2A\partial^2\Delta\phi/\partial t^2). \quad (7)$$

If the blueshift satisfies $A/2 \times (\partial\Delta\phi/\partial t)^2 \ll \Delta\phi(t(\omega))$, a simple mapping approach to extract phase shift as $\Delta\phi(t(\omega)) = \Delta\varphi(A(\omega-\omega_0))$ gives a good result with less distortion.

The experiments were performed with a 10-Hz chirped-pulse amplification Ti:sapphire laser system. The pulse duration can be adjusted in a range of 40–400 fs by introducing a slight offset in the distance between the two gratings of the compressor, and it was measured with an autocorrelator and also monitored by our spectral interferometry: scanning the delay time between the pump and probe pulses, the onset of the interferometric fringe shift will change in the spectral interferogram, and the pulse duration can be calculated with a resolution of 66 fs. The ratio of a femtosecond prepulse to a main pulse was measured less than 10^{-5}. The experimental arrangement is shown in Fig. 1. A linearly chirped pulse laser was divided into two parts by a beamsplitter with reflectivity of 20% and transmission of 80%. The higher-energy laser was used as a pump laser and focused by a parabolic mirror with a focal length of 20 cm into clusters. The lower-energy laser was divided into two separate laser pulses with a time separation of 2 ps by a Michelson interferometer, and the two laser pulses which transmitted through the laser-driven plasma were imaged on to the slit of the spectrometer by a $4f$ system with a magnification of 15.

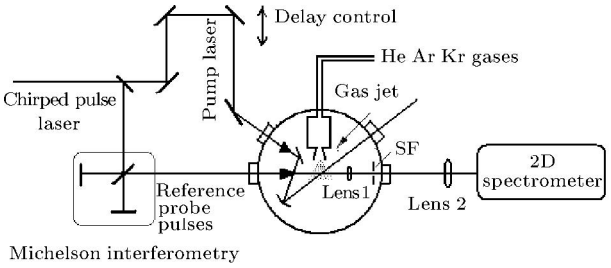


Fig. 1. Experimental set-up.

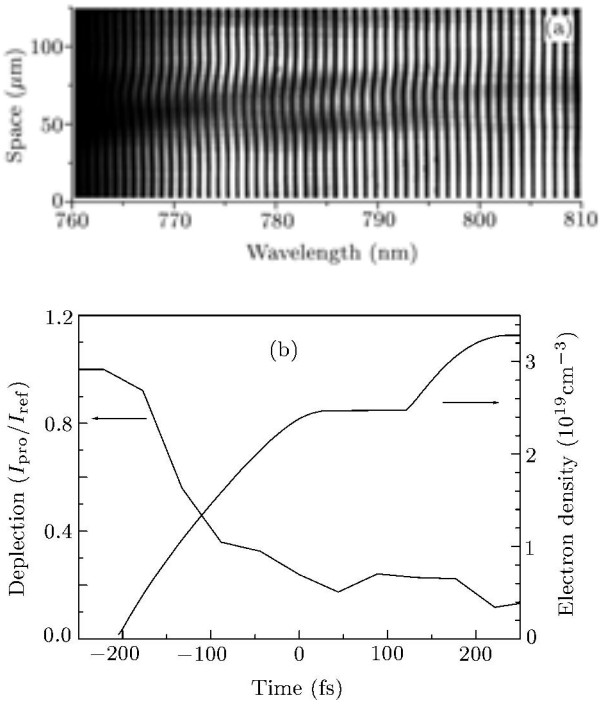


Fig. 2. (a) Spectral interferogram recorded in Kr cluster plasma driven by a 10 mJ, 220-fs chirped laser at 790 nm. The backing pressure was 8 bars. (b) Temporal evolution of depletion of the probe laser and electron density extracted from (a).

In the experiments, a pulsed sonic gas jet with

a nozzle diameter of 0.7 mm and an expansion angle of 5° was employed. The mean cluster size was estimated to be $\sim 6 \times 10^4$ for Kr at a backing pressure of 8 bars. The gas density was estimated to be about $3.8 \times 10^{18}\,\mathrm{cm}^{-3}$ by the measurement of ionization of helium. For the investigation of the ionization dynamics of Kr clusters, a 10-mJ of chirped pulse laser with pulse duration of 220 fs was employed, the peak intensity was $\sim 4 \times 10^{15}\,\mathrm{W/cm}^2$. The recorded spectral interferogram under this condition was shown in Fig. 2(a), the spot size was about 30 μm. The evolution of electron density and depletion of the probe laser by laser-driven plasma extracted from the spectral interferogram was shown in Fig. 2(b). A final average charge state of $\sim Kr^{9+}$ was produced which was approximately in agreement with the calculation result by the hydrodynamic model.[12] If only optical field ionization (OFI) was included, only an average charge state Kr^{3+} at this laser intensity could be obtained. It can be seen from Figs. 2(a) and 2(b) that a span of time exists when the electron density changes very slowly, and follows by another rapid increase until it reaches a stable level. The depletion of the probe laser which is defined as I_{pro}/I_{ref} was also given in Fig. 2(b), ultrafast depletion of the probe laser is found to be strictly coincident with the ionization front as seen in other experiments.[2]

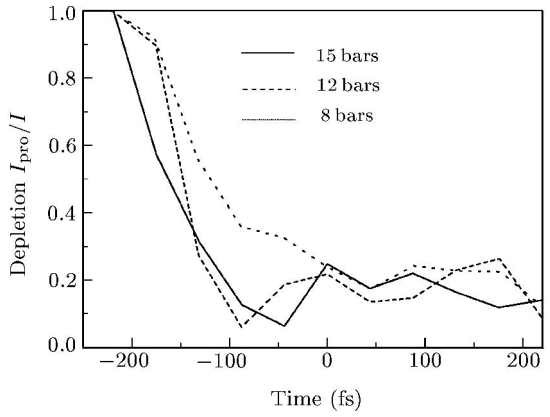


Fig. 3. Depletion of the probe laser in Kr cluster plasma at different backing pressures.

As the backing pressure increases up to higher level (> 15 bars), the final phase shift has an increase smaller than increasing backing pressure. However, in the case of helium, the final phase shift increases almost linearly as the backing pressure increases from 5 to 30 bars in our experiments. The possible cause may be explained as follows: as the backing pressure increases, the gas density increases up to a high level $> 10^{19}/\mathrm{cm}^3$, the dense electrons in cluster plasma would make defocusing of the pump laser. On the other hand, intense scattering together with the absorption of the pump laser would distort the temporal shape of the pump laser as it travels through plasma. Figure 3 shows the time-resolved depletion of the probe laser at the backing pressures of 8, 12, 15 bars, respectively. As the backing pressure increases, the depletion also increases. Therefore, an analysis of beam propagation for the pump laser will be needed if the gas density is larger than $10^{19}/\mathrm{cm}^3$.

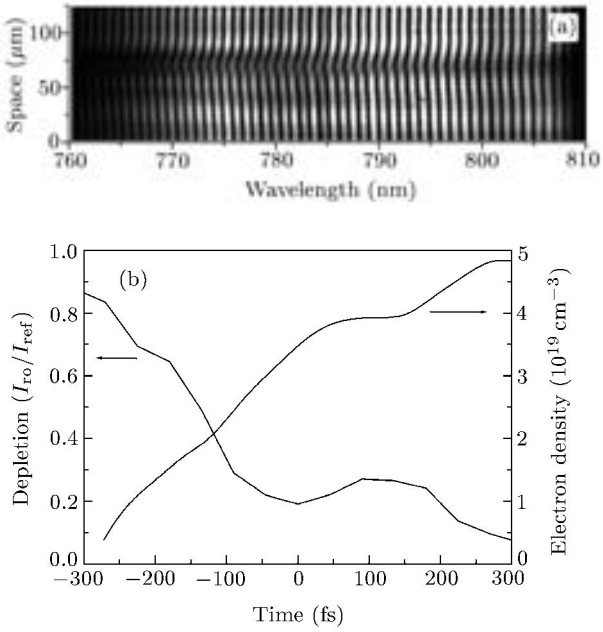


Fig. 4. (a) Spectral interferogram recorded in Xe cluster plasma driven by a 12-mJ 300-fs chirped laser at 790 nm. The backing pressure was 6 bars. (b) Temporal evolution of depletion of the probe laser and electron density extracted from (a).

For Xe clusters at a backing pressure of 6 bars, the average gas density is about $2.8 \times 10^{18}\mathrm{cm}^{-3}$, the cluster size is estimated to be about 20 nm in diameter. The laser intensity is $5 \times 10^{15}\,\mathrm{W/cm}^2$ with a pulse duration of 300 fs and energy of 12 mJ. The recorded spectral interferogram is shown in Fig. 4(a). It could be seen that under this condition, a very small highly ionized spot size of less than 20 μm was produced. The evolution of electron density and depletion of the probe laser extracted from Fig. 4(a) is shown in Fig. 4(b), a final average charged Xe^{18+} is obtained, and an appearance of a period can also be found when the electron density and depletion vary slowly. In our experiment, the probing pulses copropagate with the pump laser at an angle of 45°, as shown in Fig. 1. Therefore, the measured electron density corresponds to an average value over a range of 20 μm and 30 fs in a scale of space and time. Employing the hydrodynamic model, we have calculated the ionization process of an Xe cluster with a diameter of 20 nm. It was shown in Fig. 5 together with the 300-fs chirped pump laser. As the hydrodynamic model described, the electrons were produced at early stage by optical field ionization, then the cluster absorbed

laser energy by inverse bremsstrahlung heating and electron-impact ionization acted as a main ionization mechanism. The cluster expanded at a slow speed for a large-size cluster at early stage under the forces of hydrodynamic pressure and Coulomb explosion. The ionization will be saturated if the atomic ionization energy is high enough, and the electron yield increases very slowly in a long span of time. When the electron density decreases owing to the expansion, the resonant laser-cluster coupling occurs. The resonant absorption and enhancement of electric field would, in a short time, yield high electron temperature and large ponderomotive force on electrons which could ionize highly charged ions. Therefore, the electron yield will rapidly increase after a slowly varied period as shown in Fig. 5, which may be reflected in the measured spectral interferogram even if it occurs in a very short time.

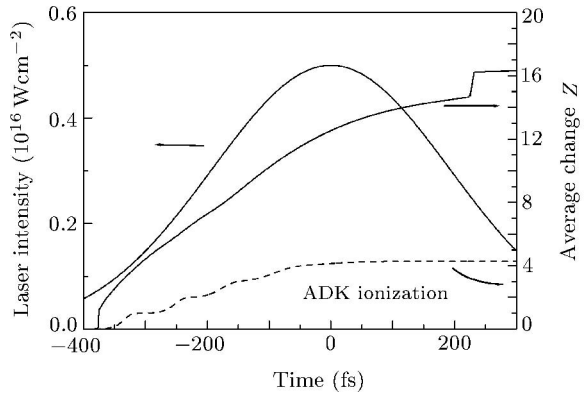


Fig. 5. Ionization process of an Xe cluster with a diameter of 30 nm calculated by the hydrodynamic model for a 12-mJ 300-fs chirped laser at 790 nm (solid). Ionization process of the Xe atom calculated by ADK formula (dashed).

The ionization process (dashed line) of the Xe atom by OFI was also shown in Fig. 5. It could be seen that only an average charge state Xe^{4+} at this laser intensity could be obtained and ionization of Xe atom saturates at the peak intensity of the laser. Therefore, the ionization dynamics of clusters is very different from that of atoms, and ultrafast diagnostics such as SI could be a useful tool to obtain an insight into the mechanism of laser-cluster interaction.

In conclusion, a linearly chirped pulse spectral interferometry has been employed to investigate ionization dynamics of clusters irradiated by chirped femtosecond lasers. The production of highly charged ions indicates that there is a strong laser-cluster coupling in laser-cluster interaction. Ultrafast depletion of the probing laser has been found to be coincident with the ionization front. The observed two-step ionization dynamics for Xe and Kr clusters irradiated by high-intensity lasers may imply the role of resonance enhancement as the cluster explodes.

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