Femtosecond pumping of nuclear isomeric states by the Coulomb collision of ions with quivering electrons

Jie Feng,1 Wenzhao Wang,1 Changbo Fu,2, 4* Liming Chen,1, 3, 4† Junhao Tan,4, 5 Yaojun Li,1 Jinguang Wang,4, 5 Yifei Li,1, 5 Guoqiang Zhang,6 YUGANG Ma,2 and Jie Zhang1, 3

1School of Physics and Astronomy, Shanghai Jiao Tong University, Shanghai 200240, P. R. China
2Institute of Modern Physics, Fudan University, Shanghai 200433, P. R. China
3IFSA Collaborative Innovation Center, Shanghai Jiao Tong University, Shanghai 200240, P. R. China
4School of Physical Sciences, University of Chinese Academy of Sciences, 100049 Beijing, P. R. China
5Laboratory of Optical Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, P. R. China
6Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201210, P. R. China

(Dated: September 26, 2020)

Efficient production of metastable quantum states of nuclei (isomers) is critical for exotic applications, like nuclear clocks, nuclear batteries, clean nuclear energy, and nuclear gamma-ray lasers[1–6]. However, due to low reaction cross sections and quick decay, it is extremely difficult to acquire significant amount of isomers with short lifetimes via traditional accelerators or reactors. Here, we present femtosecond pumping of nuclear isomeric states by the Coulomb excitation of ions with the quivering electrons induced by laser fields for the first time. Nuclear isomers populated on the second excited state of 83Kr, are generated with a rate of 3.84 × 1017 per second from a table-top hundreds-TW laser system. This high efficiency of isomer production can be explained by Coulomb collision[7] of ions with the quivering electrons during the laser-cluster interactions at nearly solid densities.

INTRODUCTION

Nuclear isomers have a broad range of applications[8]. For examples, nuclear isomers like 178m2Hf and 180m7Ta etc., due to their extremely high energy-storage capabilities compared with chemical ones, are regarded to be very good battery materials[1–5]; 99mTc isomers are widely used in medical radiographic imaging of hearts, lungs and other organs[6]; nuclear clocks which use nuclear isomers like 229mTh etc. are possible candidates for the next generation of the most accurate time and frequency standards as replacements of current atomic clocks[9]; nuclear isomer gamma-ray lasers were also proposed[10]. Nuclear isomers also play important roles in nucleosynthesis[11], that is relevant with the creation of the nuclear isotopes in stars, and then eventually affect the creation of lives in the cosmos.

For nuclear isomer applications shown above, the key bottleneck today lies in the abilities to excite and de-excite isomers on demands. From the theoretical point of view, the nuclear isomer transition mechanisms are still not well understood. For example, isomer half-lives are difficult to be predicted even within an order of magnitude. From the experimental point of view, this bottleneck appears as how to excite or de-excite isomers efficiently. Traditionally, isomers are produced with accelerators or reactors. However, limited by the beam intensities of these drivers, it is very difficult to accumulate enough amounts of isomers, in many cases, especially for those short live isomers.

The rapid development of high-intensity femtosecond lasers brings great potentials to new concept of accelerators and radiation sources[12–17]. Nowadays, the laser intensity focused onto targets can be beyond 1024 W/cm2. It can create hot plasmas with extreme pressures, temperatures, and currents under which the nuclear reactions can take place[17–20]. Here we report the first proof of principle experiment of femtosecond pumping of 83Kr to its isomer states by the Coulomb excitation of quivering electron collision with ions during laser-cluster interactions. A significant amount of isomers has been detected. This opens a new path to produce nuclear isomers with an extremely high efficiency during an extremely short time. This abnormally high efficiency may lead to a deeper understanding of the laser-isomeric quantum physics.

EXPERIMENTAL SETUP

The experiment is carried out using a Ti: Sapphire laser system at the Laboratory of Laser Plasmas of Shanghai Jiao Tong University. The experimental setup is shown in Fig. 1. 30 femtosecond laser pulses with an energy of 3.6 J are focused at an intensity of 1 × 1019 W/cm2 on natural Kr gas, with 11.5% 83Kr isotope, from a jet in the vacuum chamber with a backing pressure up to 7 MPa. During the adiabatic expansion process, the Kr gas jets are cooled down and being frozen to nanoparticles or clusters which serve as the targets. During the laser-plasma interaction, Kr clusters are ionized at the laser focus, and then electrons are rapidly heated up by various collective and nonlinear
FIG. 1: The schematic experimental setup. The femtosecond laser pulses are focused on the Kr nanoparticles ejected from a high-pressure Kr gas nozzle. A Faraday cup (FC) is used to measure the ion spectrum. After the interaction between the laser and clusters, the gas is pumped and frozen on surfaces of the liquid-nitrogen cold trap. The possible radioactive decays are recorded by a NaI detector under the trap. A probe laser pulse is aligned through the gas/plasma during interaction to provide interferograms.

processes to a non-equilibrium state with mean energies of many keV before the clusters disassemble in the laser field[13]. During this process, the low-lying excited states of $^{83}$Kr, including the first excited isomeric state $^{83}_{1}m_{Kr}$ with $E_1 = 9.4$ keV, the second $^{83}_{2}m_{Kr}$ with $E_2 = 41.6$ keV and the third $^{83}_{3}m_{Kr}$ with $E_3 = 562.5$ keV (Fig. 2a), could be populated due to physical processes to be discussed later. After the laser-cluster interaction, the Kr gas is collected by a turbo pump, and then goes into a cold trap to be frozen on it (Fig. 1). A NaI detector is used to record the decay event through a Be window.

ANALYSIS AND RESULTS

Ion Temperature

A typical time-of-flight spectrum obtained by the Faraday cup is shown in the extended data set in Fig. 4. The energy spectrum of Kr ions from the coulomb explosions follows the quasi-Boltzmann distribution[21, 22]. The temperature of Kr ions is fitted to be $T = (15 \pm 4)$ keV. By integrating the area under the quasi-Boltzmann distribution curve, and taking the average Kr charge state to be 14$^{+}$ according to the ADK model[23], we estimate the total number of the Kr atoms in the laser spot to be $4.5 \times 10^{15}$ in one shot, and then the total number of the $^{83}$Kr atoms in the laser field is,

$$N_{FC} = (5.0 \pm 0.3) \times 10^{14}/\text{shot},$$

where the error comes from the fitting of the Faraday cup spectrum.
FIG. 2: $^{83}$Kr Isomeric states and decay characteristics. (a) The decay scheme of $^{83}$Kr. The $J^\pi$, energies, and half-life times of the ground state, first, second, and third excited states are listed. (b) A typical spectrum measured by the NaI detector. The blue square markers represent the data measured while the red crosses the background. The red solid line is the fit with sum of the decay (9.4 keV, 5.5%, black dash line), $k_\alpha$ (13.6 keV, 13.8%, black dot line), and $k_\beta$ (14.1 keV, 2.1%, black dot-dash line). (c) The decay time spectrum. Blue squares represent the experimental data, and error bars denotes $\pm 1\sigma$. The red solid line represents a fit with an exponential function. The fitted half-life is $T_{1/2}^{exp} = (1.80 \pm 0.05)$ h.

$^{83m}$Kr$_2$ generating efficiency

A typical energy spectrum measured by the NaI detector is shown in Fig 2b. The decay spectrum is shown in Fig. 2c. The half-life time is measured to be $1.80 \pm 0.05$ h. This agrees well with $^{83m}$Kr$_2$ lifetime. Both the energy and time spectra prove that the decays are from $^{83m}$Kr$_2$. The total number of radiation photons detected from $^{83m}$Kr$_2$ isomers in 100 shots is fitted to be 2283 $\pm$ 30 particles. For each experimental run, we have 100 shots with the same shooting speed in about 67 mins. Considering each shot with an energy of 3.6 J, as well as the cold-trap detecting efficiency of 0.20% (see Methods), we deduce that the $^{83m}$Kr$_2$ producing efficiency for single shot is

$$N_{exp}^{zp} = (3203 \pm 42) p/J = (1.15 \pm 0.02) \times 10^4 p/\text{shot}. \quad (2)$$

Noticing that the abundance of $^{83}$Kr is 11.5%, The reaction ratio from $^{83}$Kr g.s. to $^{83m}$Kr$_2$ is estimated to be $2.2 \times 10^{-11} \, \text{p/p}$.

Possible mechanisms of $^{83m}$Kr$_2$ generating

Many processes could, of course, be responsible for the production of $^{83}$Kr isomers in our setup,

$$A + ^{83}\text{Kr} \rightarrow ^{83m}\text{Kr}_i + A, \quad (3)$$

where $i = 1, 2, \text{or} 3..., \text{representing different nuclear excited states of Kr}$. Here $A$ can be electrons, or isotopes of Kr, i.e. $^{78,80,82,83,84,86}$Kr, considering the fact that the Kr gas are natural in this experiment. Due to the fact that electron densities, as well their energies, are much higher than those of ions according to the following simulation, the contributions from ion-ion collisions could be neglected.

There are several possible transitions for $^{83}$Kr excitations, as are shown in Fig. 2a. The transitions which are responsible for the $^{83m}$Kr$_2$ could be g.s. $\rightarrow$ 2nd ($T02$), g.s. $\rightarrow$ 1st. $\rightarrow$ 2nd ($T012$), and g.s. $\rightarrow$ 3rd. $\rightarrow$ 2nd ($T032$). Excited levels above 3rd one are also considered but having negligible contributions.
FIG. 3: Particle-in-cell simulation of nuclear Coulomb excitation in interaction of laser pulses with clusters. (a-d) Laser driven electrons from a cluster, corresponding to four different moments ($t = 6.75$ fs, 8 fs and $t = 30$ fs, 31.25 fs correspond to the process of linear resonance\cite{24, 25} and nonlinear resonance\cite{26, 27} respectively), where the arrow represents electrons motion direction and the color shows electrons energy. (e) The average density evolutions of electron and krypton ions in cluster region, and the insets are the density distributions at $t = 60$ fs, the $n_c$ is critical density equaling to $1.736 \times 10^{27} m^{-3}$, the red cycle is the cluster region. (f) The energy evolution of electrons in cluster region. (g) The excitation number evolution for three different energy states, and the inset is total excitation number in the laser irradiation region. The light red shaded area represents the temporal window of laser interaction with cluster. Most isomers are generated in this window. The $^{83m}$Kr$_2$ producing rate here is $(3.84 \pm 0.05) \times 10^{17}$ p/s.

If the transitions of T02, T012, and T032 are due to the coulomb excitation mechanism, their strength could be estimated following the Ref. \cite{7}. For electric excitation, the cross section can be written as,

$$\sigma_{E\lambda} = c_{E\lambda} E^{-2} (E - \Delta E')^{-1} B(E\lambda) f_{E\lambda},$$

where $E$ is the projectile's energy, $\Delta E' = (1 + A_1/A_2)\Delta E$, $A_1$ ($A_2$) is the mass of the projectile (target), $\lambda$ is the gamma multiplicity number, $f_{E\lambda}$ is the f-function described in Ref. \cite{7}, $c_{E\lambda}$ is

$$c_{E\lambda} = \frac{Z_1^2 A_1}{40.03} [0.07199(1 + A_1/A_2)Z_1 Z_2]^{-2\lambda+2},$$

where $Z_1$ ($Z_2$) is the charger of the projectile (target).

The isomer products through the coulomb excitations by electrons can be estimated by

$$N^{CE} = \int \int n_e n_0 \langle \sigma_{E\lambda} v_e \rangle dV dt.$$
fields about 20 times, which is defined as nonlinear resonance mechanism in fs laser-cluster interactions [26, 27]. Because of the flips, many energetic electrons which are resonant with laser field can go back and forth, and collide the relative static heavy ions (see Figs. 3a-d), and then get $^{83}$Kr nuclei excited. The high densities of ions (about $5n_c$) and electrons (about $40n_c$) (Fig. 3e), as well as the high electron temperature, result in the high productivity of the isomers. Figure. 3g shows the Coulomb excitation rates for three different paths at different time. For all paths, the generation rates of $^{83}$Kr isomeric states dominated by laser-on period, which is about 20 fs. The ratio of T01:T012:T03 $\simeq 5\times 10^{14}:1:2\times 10^8$. Because T32 is almost equal to 100%, we have T032=T03×T32=T03. Our simulation results demonstrate that the most possible path of $^{83m}$Kr observed in experiment is T032, which is clearly shown in the inset of Fig.3g. The disagreement in Fig. 3g may come from the PIC simulations errors, as well as other processes not considered in the previous calculations. For examples, processes including nuclear excitation by electron transfer (NEET), electron bridge (EB), nuclear excitation by electron capture (NEEC), may also contribute significantly[28–30].

DISCUSSION

In many isomer applications, lifetimes of the isomers needed are relatively short. For example, in nuclear gamma-ray laser applications, nuclei with two excited states are needed, in which the lower state has a shorter lifetime, enabling a population inversion between them. Furthermore, in order to reduce the line-width difficulty[10, 31], isomers with lifetime down to sub-nanoseconds are needed. it was thought to be impossible to produce such isomers with so short lifetimes with traditional accelerators or nuclear reactors[31]. The similar difficulty also arises in nuclear clock applications. $^{229}$mTh and $^{235}$mU are promising nuclear isomers to be used in nuclear clocks[9]. Their half lifetimes are 2 and 26 mins respectively. How to pump them in very short temporal duration to isomer levels is still a big challenge. Our experimental result shows an efficient way to quickly pump short lifetime isomers in femtosecond temporal duration via femtosecond laser-cluster interaction. Furthermore, our method provides a novel tool for the exploration in inter-disciplines of nuclear physics, plasma physics, and atomic physics.

SUMMARY

In summary, for the first time, we have presented efficient femtosecond pumping of nuclear isomeric states by Coulomb collision of ions with quivering electrons at nearly solid densities of interaction of fs laser pulses with clusters. By interaction of Kr cluster targets with a 30 fs laser pulses at 120 TW, the nuclear isomers of $^{83m}$Kr($E = 41.6$ keV and $T_{1/2} = 1.83$ h) have been generated with a producing rate of $(3.84 \pm 0.05) \times 10^{17}$ particles/s, which is much higher than traditional methods. Our simulation shows that the high efficiency comes from the electron-quivering effect. The effective $^{83m}$Kr generation are dominated during the 30 fs laser pulses duration, due to the collisions of high densities ions with high energy electrons accelerated by the electron-quivering effect. We also find that the $^{83m}$Kr($E = 41.6$ keV) isomers are mainly produced through the middle state, i.e. g.s. $\rightarrow$ 3rd $\rightarrow$ 2nd, and the direct transition g.s. $\rightarrow$ 2nd may be negligible. This high efficiency, femtosecond pulse duration, and easy accessibility of production of short lifetime of nuclear isomers could be greatly beneficial for potential applications such as clinic nuclear imaging, nuclear batteries, nuclear clocks and nuclear gamma ray lasers.

METHODS

The fs laser pulses

The laser pulses are from commercial femtosecond laser system, which delivers 3.6 J/pulse in 30 fs (full width half maximum, FWHM) at center wavelength of 800 nm and operated frequency of 0.5 Hz. The laser pulses are focused with an f number of 20 off-axis parabolic mirror into the front edge of a supersonic gas jet, and the height of focus relative to the jet is about 1.5 mm. The typical diameter of the focus spot is about 30 $\mu$m (FWHM), producing a laser intensity of about $1 \times 10^{19}$ W/cm$^2$. 
The Vacuum Pumps

To improve the gas collecting efficiency, vacuum pumps in the system are used as follows. There are two pump sets (Fig. 1) on the optical chamber and target chamber respectively. Each set has a roughing pump and a turbo pump. Before the experiment, they are turned on to take the vacuum down. During experiment, the pump set at the optical chamber, as well as the roughing pump at the target chamber, are turned off. Only the turbo at the target chamber is on. In this way, we estimate that over 95% Kr gas can be collected into the cold trap.

The cold trap

The cold trap is filled with liquid nitrogen. There has a 380 µm thick Be window close to the trap. If Kr isomers are produced via coulomb collision, γ-rays from the isomer decays would be detected by a detector located behind the window.

The cold-trap detecting efficient

The cold-trap detecting efficiency is the product of the following factors: the chamber-to-trap gas transferring efficient (95%), the area ratio the cold trap’s bottom to the whole surface (26.5%), the factor due to Be window absorption (95.6%), the factor due to the delayed measuring time (81.8%), the geometrical factor due to the distance between the detector and the Kr ice (4.7%), and the decay branching ratio (21.39%) [32]. Therefore, we estimate the cold-trap detecting efficient to be 0.20% in X-ray energy range of 9 to 15 keV.

The NaI detector and detection

The NaI detector is compose of a NaI crystal of 50 mm diameter and 3 mm thickness, a Be window of 200 µm thickness, and a photomultiplier tube. The NaI detector’s resolution is measured to be FWHM = 4 keV at E = 10 keV. For $^{83m}$Kr$_2$, there have several decay lines around 10 keV, i.e. 9.4, 12.6(K$_\alpha$), and 14.1(K$_\beta$) keV, with intensities of 5.5%, 13.8%, and 2.1% respectively, from NNDC database [32]. The energy spectrum is fitted with assuming the energies and line strengths shown above.

The Faraday Cup Measurement

A copper Faraday cup with a diameter of 0.75 cm is located 133 cm away from the target to detect the ion spectra. The signals from Faraday cup is recorded by an oscilloscope with input impedance of 50 Ω.

The gas density monitor camera

A camera was installed as the monitor to measure the gas/plasma density from the nozzle. A probe laser pulse passes through the plasma induced by the main laser pulse to form interferograms, providing the gas/plasma density information.

The nuclear Coulomb excitation calculation

The rate of nuclear Coulomb excitation is calculated based on the PIC simulation data. In the PIC simulation, the cluster area is divided into many cubic cells, whose shape is similar to that of a disc. In Eq.6, $\langle \sigma E \lambda v_e \rangle$ is expressed as the weighted average of product of cross-sections and electron velocities in single cell, and $dV$ the volume of single cell. In order to prevent electrons from running out of cluster area in the time of $dt$, here we interpolate the $n_e$, $n_i$ and $\langle \sigma E \lambda v_e \rangle$ in every cell, and the corresponding interpolation $dt = 1$ attosecond. Then, substituting these interpolated variables into Eq.6, and integrating in the whole cluster area. Actually, the real shape of cluster is close to sphere,
and here the cell number of sphere is \(4N_r/3 \approx 50\) times higher than discs. Therefore, the yield of excitation states of single cluster should be \(N_{SCE} = 50N_{CE}\).

According to the hydrodynamics simulation of Kr cluster generation from the nozzle used in experiment[26, 35], the spacing within clusters is about 1.35 \(\mu\)m and the average radius of cluster is about 30 nm. If the radius of cluster plasma expands to 1.35 \(\mu\)m, the plasma density would be reduced by four orders of magnitude. Therefore, the influence of the Coulomb excitation from surrounding clusters plasma and ion-ion collisions can be ignored. In addition, according to the laser plasma interaction area detected by a probe laser (Extended Data Fig. 6), the estimated cluster number is about \(3 \times 10^6\). Therefore, the total yield of excitation states of one single shot can be expressed as \(N_{TCE} = 3 \times 10^6N_{SCE}\).

**Particle-in-cell Simulations**

We carry out 2D PIC simulations with the EPOCH code[33], the ADK model code[34] is adopted for the field ionization. The simulation box size is \(6\mu m \times 6\mu m\) in \(x \times y\) directions. We take the cell sizes in the two directions as 0.8 nm, the time step as 1.78 attosecond, and 10 quasi-particles per cell. The p-polarized laser pulses propagate along the \(x\)-direction, and the laser-focusing plane is located at \(x = 1.5\mu m\). The laser pulses have a Gaussian transverse profile with \(w_0 = 30\mu m\) and a Gaussian longitudinal envelope with a pulse duration of 30 fs (FWHM). The normalized vector potential \(a_0 = 1.8\). One circular spot is set at \((x, y) = (1.5 \mu m, 0 \mu m)\) with a uniform krypton gas density distribution \((N_{kr} = 1 \times 10^{28} m^{-3})\) to represent a cluster. The cluster radius is 30 nm according to the Measurement of Rayleigh scattering (Extended Data Fig. 5). Moreover, we utilize the convolutional perfectly matched layer boundary to absorb particles and electromagnetic fields.

**Data availability**

Experimental raw data were obtained using a commercial 200 TW Ti: Sapphire laser system at the Laboratory of Laser Plasmas of Shanghai Jiao Tong University. All of the relevant data that support the findings of this study are available from the corresponding authors upon reasonable request.

**Code availability**

Computer code used for particle-in-cell (PIC) calculation in this study is available at https://www.archer.ac.uk/community/eCSE/eCSE03-01/eCSE03-01.php

**ACKNOWLEDGEMENTS**

This work is supported by the Science Challenge Project (TZ2018005), the National Nature Science Foundation of China (11875191, 11991073, 11421505, 11721404), the Strategic Priority Research Program of the CAS (XDB1602), the National Key R&D Program of China (2017YFA0403301), and the Key Program of CAS (XDA01020304, XDB17030500). We would like to acknowledge the 200TW laser operating staff for running the laser facility.

**AUTHOR CONTRIBUTIONS**

C.B.F. and L.M.C. proposed the study. J.F., W.Z.W., J.H.T., J.G.W., Y.J.L., G.Q.Z. and C.B.F. set up the experiment, and participated in the data collection. L.M.C., C.B.F., and J.F. led the experimental running, the data
analysis, and the interpretation of the results. J.F. and Y.F.L. carried out PIC simulation. C.B.F., L.M.C., J.Z., J.F., and Y.G.M. wrote the text and led the discussion. Y.G.M. and J.Z. are the principle investigators of the laser nuclear research project. All listed authors contributed to discussion and helped to improve the manuscript.

COMPETING FINANCIAL INTERESTS

The authors declare no competing financial interests.

[1] J. Carroll, J. Burnett, T. Drummond, J. Lepak, R. Propri, D. Smith, S. Karamian, J. Adam, F. Stedile, and F. Agee, Hyperfine Interact. **143**, 37 (2002).
[2] D. Belic, C. Arlandini, J. Besserer, J. De Boer, J. Carroll, J. Enders, T. Hartmann, F. Käppeler, H. Kaiser, U. Kneissl, et al., Phys. Rev. Lett. **83**, 5242 (1999).
[3] J. Carroll, S. Karamian, L. A. Rivlin, and A. Zadernovsky, Hyperfine Interact. **135**, 3 (2001).
[4] J. Gunst, Y. A. Litvinov, C. H. Keitel, and A. Pálffy, Physical Review Letters **112**, 082501 (2014).
[5] Y. Wu, C. H. Keitel, and A. Pálffy, Phys. Rev. Lett. **122**, 212501 (2019).
[6] S. Banerjee, M. R. A. Pillai, and N. Ramamoorthy, Semin. Nucl. Med. **31**, 260 (2001).
[7] K. Alder, A. Bohr, T. Huus, B. Mottelson, and A. Winther, Rev. Mod. Phys. **28**, 432 (1956).
[8] A. Aprahamian and Y. Sun, Nat. Phys. **1**, 81 (2005).
[9] L. von der Wense, B. Seiferle, M. Laatiaoui, J. B. Neumayr, H.-J. Maier, H.-F. Wirth, C. Mokry, J. Runke, K. Eberhardt, C. E. Düllmann, et al., Nature **533**, 47 (2016).
[10] E. Tkalya, Phys. Rev. Lett. **106**, 162501 (2011).
[11] P. M. Walker and J. J. Carroll, Phys. Today **58** (2005).
[12] T. Tajima and J. M. Dawson, Phys. Rev. Lett. **43**, 267 (1979).
[13] T. Ditmire, J. Tisch, E. Springate, M. Mason, N. Hay, R. Smith, J. Marangos, and M. Hutchinson, Nature **386**, 54 (1997).
[14] J. Zhang, A. MacPhee, J. Lin, E. Wolfrum, R. Smith, C. Danson, M. Key, C. Lewis, D. Neely, J. Nilsen, et al., Science **276**, 1097 (1997).
[15] J. Zhang, A. MacPhee, J. Nilsen, J. Lin, T. Barbee Jr, C. Danson, M. Key, C. Lewis, D. Neely, R. O’Rourke, et al., Phys. Rev. Lett. **78**, 3856 (1997).
[16] B. Remington, Science **284**, 1488 (1999).
[17] T. Ditmire, J. Zweiback, V. Yanovsky, T. Cowan, G. Hays, and K. Wharton, Nature **398**, 489 (1999).
[18] X. Zhang, J. Zhao, D. Yuan, C. Fu, J. Bao, L. Chen, J. He, L. Hou, L. Li, Y. Li, et al., Phys. Rev. C **96**, 055801 (2017).
[19] Y. Kim, J. M. Mack, H. W. Herrmann, et al., Phys. Plasmas **19**, 056313 (2012).
[20] C. Fu, J. Bao, L. Chen, et al., Sci. Bull. **60**, 1211 (2015).
[21] M. Barbui, W. Bang, A. Bonasera, et al., Phys. Rev. Lett. **111**, 082502 (2013).

FIG. 4: A typical ion spectrum measured by the Faraday cup. The black solid line represents a Boltzmann distribution fit with Kr ion temperature $T = 15$ keV. The black dash line is an exponential fit corresponding to the X-ray induced by the main laser. The red solid line is the sum of the two fits.
FIG. 5: Formation characteristics of krypton cluster. (a) Rayleigh scattering signal from a CW laser focused on krypton cluster detected by an ultra-fast photodiode. (b) The blue square markers represent the peak intensity ($I$) of scattering signal at different pressure ($P$), error bars denote $\pm 1\sigma$ uncertainties, and the red line is the fit $I \propto P^{2.64}$. The power factor 2.64 is lower than Hagenas value 3-3.5, and the Hagen parameter $\Gamma^*$ is located in the range of $10^4$ to $10^6$[36]. According to the formula correction of F. Dorchies[37], the number of atoms per cluster can be expressed as $N = 100 \left( \frac{r^*}{1000} \right)^{1.8}$. The Van der Waals radius of krypton atom $r = 202$ pm, therefore the average radius of cluster can be estimated as $R = rN^{1/3}$ (black circle markers).
FIG. 6: Optical interference diagnosis. The diagnosis is based on Nomarski interferometer. (a) The interference fringes of 7 MPa Kr gas (left) and the calculated atom density distribution (right) with the Abel inversion method[38]; (b) The interference fringes of laser gas target interaction.

[36] O. F. Hagena and W. Obert, Journal of Chemical Physics 56, 1793 (1972).
[37] F. Dorchies, F. Blasco, T. Caillaud, J. Stevefelt, C. Stenz, A. S. Boldarev, and V. A. Gasilov, Physical Review A 68 (2003).
[38] Y. Yasutomo, K. Miyata, S.-I. Himeno, T. Enoto, and Y. Ozawa, IEEE Trans. Plasma Sci. 9, 18 (1981).