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Xe–Kr laser induced collisional ionization system and experimental preparation of its initial state: Four-photon resonant excitation*

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This paper proposes a novel one-colour Xe–Kr laser induced collisional ionization system. Considering the level scheme of the system, it finds that the initial state of the reaction—the four 4f levels with even J of Xe—can be prepared through method of four-photon resonant excitation by dye laser with wavelength of ∼440 nm. Absorption of an additional photon (the transfer laser) of the same wavelength will complete the laser induced collisional ionization process. The resonance enhanced ionization spectrum of Xe by four laser photons at ∼440 nm is measured through time-of-flight mass spectrometry, this aims at the preparation of the initial state of the system proposed. The Stark broadening of the measured spectrum is observed and consistent with the previous study. Analysis of the measured resonance ionization spectrum implies the feasibility of ∼440 nm four-photon resonant excitation of the initial 4f state of the Xe–Kr system proposed in this paper, which prepares for a further experiment of laser induced collisional ionization.

Keywords: laser induced collision, ionization, four-photon resonant excitation, time-of-flight mass spectrometry

PACC: 3450R, 3280K, 3280F

1. Introduction

For many years, laser induced inelastic collisions have been the object of extensive theoretical[1,2] and experimental[3,4] studies, because on the one hand, the cross section of interparticle inelastic collisions can be increased by several orders for the presence of the laser field for a laser-induced collisional energy transfer (LICET) process, typically the collision cross section can be increased to above $10^{-16}\,\mathrm{cm}^2$ from the order of $\sim 10^{-20}\,\mathrm{cm}^2$ without laser; on the other hand, they offer a way of controlling both the relative and absolute probability of competing exit channels, and thus controlling the product of a reaction. To date, experimental observations of laser-induced energy transfer,[5] charge transfer,[6] and collisional ionization[7] have been demonstrated.

As has been described by many works,[8−10] the LICET process is an inelastic collision process between an excited atom A and another atom B of ground state. If the laser is tuned resonant to the interatomic transition, then the associative action of collision and laser field will result in an energy transfer of excited atom A to atom B, which simultaneously absorbs a photon from the laser field and consequently undergoes a transition to a higher excited state, with atom A returning to its ground state due to collisional deexcitation. The process can be described as follows:

$$\mathrm{A(i)} + \mathrm{B(i')} + \hbar\omega \rightarrow \mathrm{A(f)} + \mathrm{B(f')}, \qquad (1)$$

where i and i′ denote the initial states of atoms A and B, while f and f' are their final states, respectively.

The extension of the LICET process to the case that the final state of atom B is its ionization state provides the scheme of laser induced collisional ionization. Ionizing collisions are of particular interest because of their relevance to laser and plasma physics[11] and because of the inherently high detection sensitivity of charged particles. Experimental studies of, for example, Na-Na[12] and Li-Li[13] laser induced collisional ionization systems have been reported, most of which are metal vapour systems, only few gas systems have

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been considered. This paper proposes a new rare gas laser induced collisional ionization process in the Xe–Kr system. In comparison with those former studied metal systems that need to be heated to a high temperature for mixed vapour, the experimental setup of this system is certainly simplified.

Because of the high-lying energy levels of atomic Xe, its initial state must be excited by several photons at visible wavelength through multiphoton absorption, which is evidently different from the low excitation energy of metal atoms that can be excited by only one visible photon. For the Xe–Kr system proposed in this paper, the initial state of the reaction—the four 4f levels with even J of Xe—can be prepared through four-photon resonant excitation by dye laser with wavelength of ∼440 nm. Absorption of an additional photon at the same wavelength (i.e. the pump laser and the transfer laser are of the same frequency) will make atomic Kr ionized, therefore the laser induced collisional ionization process can be accomplished with only one laser beam, which is new to general two-colour laser induced collision systems.

This paper introduces the basic process of laser induced collisional ionization in the Xe–Kr system, and four-photon resonant excitation of the four 4f levels of Xe by dye laser at ∼440 nm wavelength is experimentally realized. The intensity spectrum of Xe^+ through four-photon resonant excitation of Xe 4f state is measured by using time-of-flight (TOF) mass spectrometry[14] and subsequently analysed.

2. The fundamental process of laser induced collisional ionization in Xe–Kr system

Figure 1 illustrates those energy levels relevant to the laser induced collision process of Xe–Kr system. Before the reaction, atom Xe is excited initially to the 4f excitation state, while atom Kr is in its ground state ($4p^6\ ^1S_0$). As it is shown in Fig.1, there are four levels of Xe 4f state with even J, 4f$[7/2]_4$, 4f$[5/2]_2$, 4f$[9/2]_4$ and 4f$[3/2]_2$ level, which can be resonantly excited by four pump laser photons of frequency ω (in the wavelength region of ∼440 nm). With the conjunct effect of radiation and interatomic collision, the excitation energy of atom Xe, which then undergoes a transition to its ground state, is transferred to atom

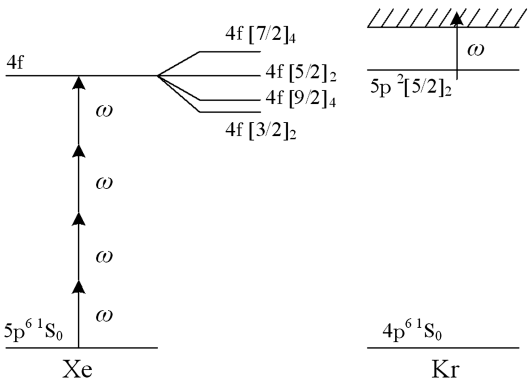


Fig.1. Energy levels of Xe–Kr laser induced collisional ionization.

Kr through collisional interaction because of the presence of Kr $5p^2[5/2]_2$ level which is near resonant with Xe 4f state, while atom Kr is ionized by simultaneous absorption of an additional photon (the transfer laser) with the same frequency ω from the laser field. Because of the strict selectivity of resonant ionization for the excitation wavelength, resonant excitation can take place only when the excitation energy is strictly or near resonant to the target atomic level. In the experiment of laser induced collisional ionization in this Xe–Kr system, the detuning (about 1400 cm^{-1}) between Kr $5p^2[5/2]_2$ level and four-photon resonance of ∼440 nm is large enough that atom Kr can not be resonantly ionized directly. This process can be expressed by the following formula:

$$\begin{aligned} &\mathrm{Xe(4f)} + \mathrm{Kr}(4p^6\ ^1S_0) + \hbar\omega \\ &\rightarrow \mathrm{Xe}(5p^6\ ^1S_0) + \mathrm{Kr}^+ + \mathrm{e}. \end{aligned} \tag{2}$$

In comparison with the diagram of a LICET process,[10] laser induced collisional ionization is virtually a special case in which the discrete final state of the LICET process is replaced by an ionization continuum. Because the pump laser and the transfer laser are of the same wavelength for this Xe–Kr system, only one laser beam is needed to accomplish the laser induced collision process which ordinarily requires two laser frequencies.

3. Experimental preparation of the initial state of Xe–Kr system

Multiphoton resonant excitation is required to prepare the Xe 4f initial state of the Xe–Kr laser in-

duced collisional ionization system because these four levels of Xe lie much higher than that of metal atoms in traditionally studied the LICET process. Considering the feature of the Xe 4f state, we use the four-photon resonant excitation by dye laser with wavelength in the region of ∼440 nm to prepare this initial state. Once excited to this state, atom Xe can be easily ionized by absorbing an additional laser photon, therefore the method of resonance enhanced multiphoton ionization, which is generally used to study high-lying atomic Rydberg states,[15] is used to measure the spectrum of four-photon resonance enhanced ionization via the Xe 4f state through TOF mass spectrometry, with the purpose of detecting the initial state of the reaction.

3.1. Experimental setup

Figure 2 shows the experimental setup for measuring the spectrum of four-photon resonance enhanced ionization via the Xe 4f state through TOF mass spectrometry. The TOF mass spectrometer, which is the principal part of the whole experimental setup, consists of the vacuum chamber and its gas extraction system, the pulse valve, the accelerating electric field and the free flight region. Driven by the Iota One Pulse Driver System, a 009-1 pulse valve (with orifice diameter of 0.2 mm) from Parker Company, America, jets high-purity xenon at a frequency of 10 Hz, with the gas pressure within the valve maintaining at 0.4 MPa. The xenon ejected from the valve enters the ionization region in the form of a pulsed beam. The 355 nm laser generated from frequency tripling of 1064 nm output of the Nd: YAG laser is used to pump the dye laser, providing pulsed laser output in the wavelength region of ∼440 nm, with 10 Hz frequency, 7 ns pulse width and 0.002 nm linewidth. The laser beam is focused by a lens of 6.5 cm focal length into the interaction region, where xenon is resonantly excited and subsequently ionized. A 4-channel Digital Delay / Pulse Generator DG535 (Stanford Research Systems) is used to control the time sequence of the dye laser and the valve driver, which keeps synchronization between the laser pulse and the xenon beam. The xenon ions created from ionization gain sufficient flight speed through the two accelerating field of 800 V and 1200 V, and then fly through a free flight region of 1.2 m length to the microchannel plate (MCP) detector with a gain of 10^6, which converts the ions into electric signals. After amplified by an amplifier, the ionization signals are either observed directly by a digital oscilloscope, or integrated and averaged by a Boxcar gated integrator/averager, with the data stored on a computer disk for further analysis. The four-photon resonance enhanced ionization spectrum via Xe 4f state can be measured by scanning the laser wavelength near the region of ∼440 nm. The vacuum

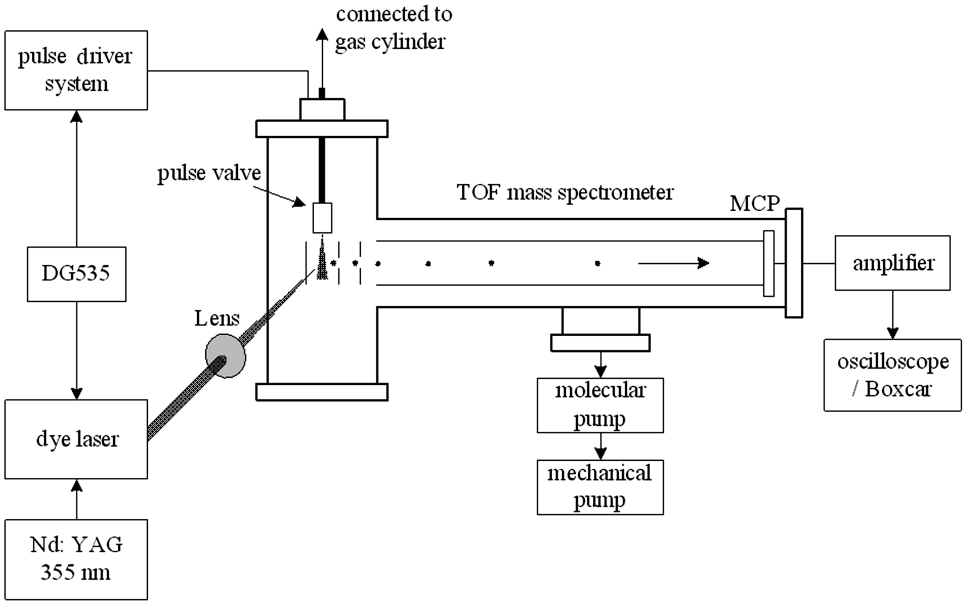


Fig.2. Experimental setup for detecting four-photon resonance enhanced ionization via the Xe 4f state through TOF mass spectrometry.

pump keeps working during the whole experimental process to maintain the environmental vacuum around the interaction region and the vacuum degree in the free flight region, insuring that the MCP detector can work normally and that the generated ions can fly to it successfully. During the experiment the background vacuum is 1.0×10^{-5} Pa, while it is 7.5×10^{-3} Pa during gas injection.

3.2. Results and analysis

The xenon beam was targeted by a femtosecond laser of 10 Hz before we measured the spectrum of four-photon resonance enhanced ionization via the Xe 4f state by ∼440 nm dye laser, so that we could determine the flight time of Xe^+ under our condition through the abundant ion signals created by the intense femtosecond laser pulse ionizing xenon.

Figure 3 shows the ion signals created by targeting of the femtosecond laser. For TOF mass spectrometry, the relationship between mass-to-charge ratio (m/q) and the flight time (t) of an ion is as follows:

$$\sqrt{m/q} = a + b \cdot t. \tag{3}$$

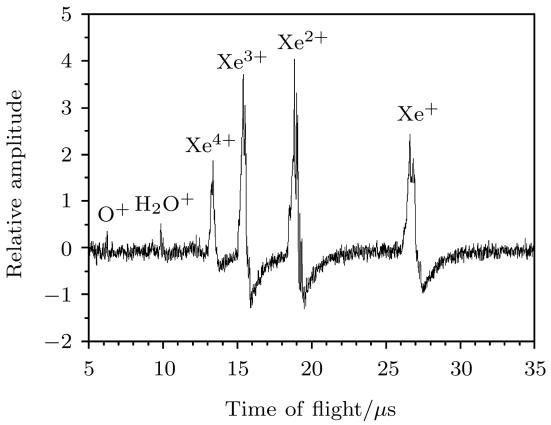


Fig.3. The ion signals created by intense femtosecond laser.

Based on our experience, the two peaks on the left in Fig.3 were determined as signals of O^+ and H_2O^+ remained in the vacuum chamber, whose flight times were 6.252 μs and 9.848 μs, with the corresponding values of $\sqrt{m/q}$ equal to 2.8284 and 4.2426, respectively. We estimated the four stronger peaks on the right, from right to left, were signals of Xe^+, Xe^{2+}, Xe^{3+} and Xe^{4+}, respectively. According to Eq.(3), the $\sqrt{m/q} - t$ data points corresponding to these signals should be on a straight line if this estimation was right. The linear fit of these data is shown in Fig.4, from which one can see that there is very good linear correlation of them, confirming that the flight time of Xe^+ is 26.6 μs under the present condition. The split of Xe ion signals of all orders in Fig.3 arises from different Xe isotopes.

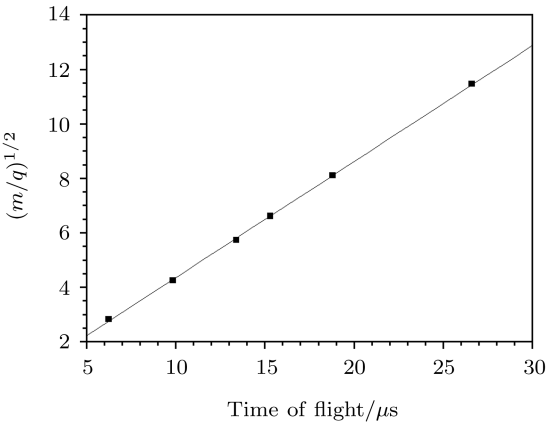


Fig.4. The $\sqrt{m/q}$ value vs. flight time of the ion signals in Fig.3.

Figure 5 shows the ion signals created by targeting atomic xenon beam using dye laser at 440 nm under the same condition. The laser beam used with pulse energy of ∼3.2 mJ is focused by a 6.5 cm focal-length lens. With a beam divergence of ∼1mrad, a power density of $\sim 1.3 \times 10^{10}$ W/cm^2 is generated in the focal volume. In comparison with Fig.3, there is only Xe^+ produced by multiphoton resonant ionization presented in Fig.5, because the power density of dye laser is much lower than that of femtosecond laser. Moreover, the Xe^+ yield is very small because of the small cross section of four-photon resonant absorption, so that the signal-to-noise ratio in Fig.5 is relatively worse than that in Fig.3.

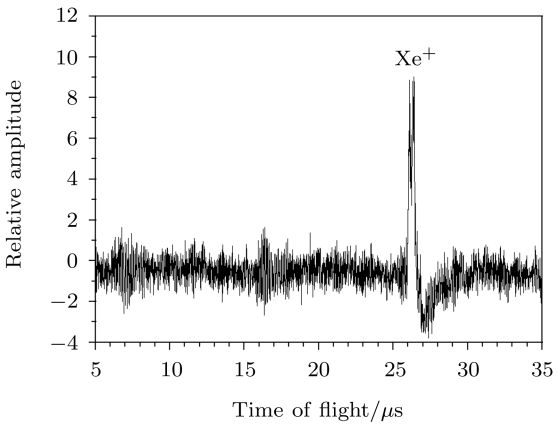


Fig.5. The ion signals created through four-photon resonant ionization via Xe 4f state.

With the laser wavelength scanned between 439.4 nm∼441 nm, we measured the four-photon resonant ionization spectrum via Xe 4f state, which is shown in Fig.6. The power density of the focused laser is the same as that in Fig.5. From Fig.6 one can obviously see the peaks of four-photon resonance at 439.83 nm and 439.995 nm, which correspond to 4f $[7/2]_4$ and 4f $[5/2]_2$ level, respectively; while the peaks of 4f $[9/2]_4$ and 4f $[3/2]_2$ level, which correspond to four-photon resonance at 440.23 nm and 440.29 nm, merge into one because of the spectrum broadening. Moreover, the fact that no ion signal was observed beyond the interested wavelength range proves that the ion signal shown in Fig.6 is indeed produced through four-photon resonant ionization by laser at ∼440 nm.

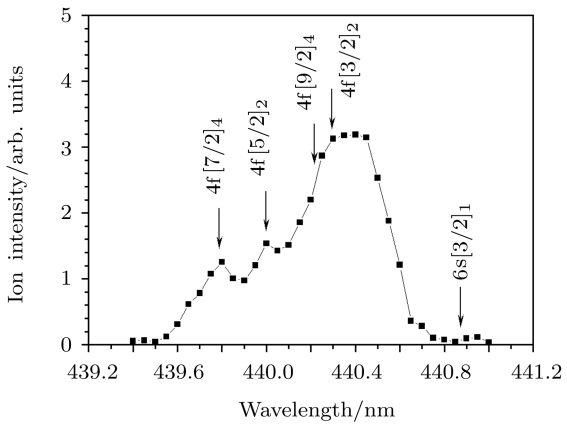


Fig.6. Spectrum of four-photon resonant ionization via Xe 4f state by ∼440 nm laser.

Atomic energy levels can be easily affected by an external electric field within which the reaction takes place, resulting in the Stark broadening of the Xe 4f state, which accounts for the broadening of the ionization spectrum shown in Fig.6. As it has been reported by Jackson *et al*,[16] the Stark broadening of the Xe 6*s* state induced by an electric-field intensity of 260 V/cm is approximately 4 cm^{-1}@440 nm, while for the 4f state of Xe which can be more likely affected by an external electric field, this broadening will be 1.5–2 times larger than that of 6*s* state. The electric-field intensity in the interaction region is 400 V/cm in our experiment, therefore the Stark broadening of the Xe 4f state is estimated between 0.18–0.24 nm, which is consistent with the experimental result.

Another fact which needs to be explained is that, the wavelength range measured in our experiment is in the negatively dispersive region of three-photon (440.8 nm) resonance of Xe $6s^2[3/2]_1$ level (68045.156 cm^{-1}), then is there any probability that the ion signals measured in this experiment are created through three-photon resonant process via 6*s* state? According to numbers of studies and discussions on this issue,[17−19] once excited to the 6*s* state, the atomic Xe will undergo a radiative transition to its ground state, generating third-harmonic photons with frequency of 3ω. Studies by Peet *et al*[17] indicate that for a Gaussian beam, the excitation of 6*s* state can be strongly suppressed by the cancellation effect between pathways of one-photon excitation at 3ω and three-photon excitation at ω. This is also proved by the fact that no ion signal was observed at wavelength of 440.8 nm in our experiment.

4. Conclusion

This paper introduces a new rare gas laser induced collisional ionization process in the Xe–Kr system. Preparation of its initial state through the method of four-photon resonant excitation by dye laser with wavelength of ∼440 nm is proposed by considering the level scheme of the system. Absorption of an additional photon (the transfer laser) of the same wavelength will accomplish excitation of Kr^+, therefore just one laser beam is needed to complete the traditional two-colour laser induced collisional ionization process. Aiming at the preparation of the initial state of this system, we measured the resonance enhanced ionization spectrum of Xe by four ∼440 nm laser photons through TOF mass spectrometry, which is accomplished through calibration of the ion signals created by femtosecond laser ionizing atomic xenon first. The Stark broadening of Xe 4f state, which is indicated by the measured spectrum, is in fair to good agreement with the previous study. Analysis of the experimental results implies the feasibility of ∼440 nm four-photon resonant excitation of the initial 4f state of the Xe–Kr system proposed in this paper. A further experimental study is anticipated to be crossed-beam laser induced collisional ionization in the Xe–Kr system.

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