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Nonsequential double ionization of alkaline-earth metal atoms by intense mid-infrared femtosecond pulses

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Abstract: A systematic study of nonsequential double ionization (NSDI) of alkaline-earth metal atoms with mid-infrared femtosecond pulses is reported. We find that the measured NSDI yield shows a strong target dependence and it is more suppressed for alkaline-earth metal with higher ionization potential. The observation is attributed to the differences in the recollision induced excitation and ionization cross sections of alkaline-earth metals. This work indicates that NSDI of alkaline-earth metals can be generally understood within recollision picture and sheds light on ultrafast control of electron correlation and dynamics of ionic excited states during NSDI of atoms with complex structures.

1. Introduction

As a fundamentally important strong-field process representing of electron correlation, nonsequential double ionization (NSDI) of atoms and molecules is a fascinating topic from both experimental and theoretical sides (for reviews, see [1,2]). A straightforward observable of NSDI is the enhancement of the double ionization yields at modest laser intensities with respect to the theoretical prediction assuming a sequential and independent ionization of the two electrons [3]. This so-called "knee" structure has been observed for all rare gas atoms [4–6] and some molecules [7–10]. It is now widely accepted that NSDI can be understood within the quasi-classical recollision model [11]. One learns from this model that, after tunneling through the distorted Coulomb potential, the first emitted electron may be accelerated and driven back to its parent ion by the oscillating laser electric field, where it can free a second electron through inelastic scattering. This insightful physical picture has been validated by numerous kinematically complete experiments [12–15] which were mainly performed on rare gas atoms.

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Currently accurate quantum calculations of NSDI still present a great challenge. Based on the recollision scenario, several simplified theoretical methods such as the semiclassical model [16,17], the classical ensemble model [18], and the S-matrix theory [19] have been proposed for understanding NSDI dynamics. While these approaches have shown to satisfyingly reproduce many important features of NSDI experiments [20–25], the agreement with experiments on (even close to) a quantitative level is still not easy to achieve. Recently, it has been realized that recollision impact excitation and ionization cross sections play a crucial role in NSDI of rare gas atoms [26–31]. The quantitative rescattering (QRS) model [32] incorporating electron impact excitation and ionization cross sections has been used to quantitatively reproduce the experimental ratios of Ne^{2+}/Ne^{+} and He^{2+}/He^{+} as functions of intensity [33,34].

So far atomic NSDI measurements are mainly focused on rare gas atoms. Strong-field double ionization of alkaline-earth metal atoms, which can be regarded as nearly ideal two-electron systems and contain a manifold of doubly excited states with a high degree of electron correlation, has shown interesting novel features as compare to rare gases. For instance, significant NSDI yields of Mg have been discovered for circularly polarized light [35], which is in strikingly contrast with rare gases. Previous experiments on strong-field double ionization of alkaline-earth metals were performed mainly with visible to near-infrared ns or ps laser pulses [36–38]. It was shown that various multiphoton resonant channels related to the detailed electronic structures strongly contribute to double ionization of alkaline-earth metals [39–47]. A detailed study of NSDI with femtosecond pulses, on the other side, may shed lights on ultrafast control of electron correlation in such atoms.

In this work we systematically study NSDI of alkaline-earth metals using intense mid-infrared femtosecond laser pulses. The multiphoton resonant pathways are significantly suppressed for such long wavelength and the quasistatic field approximation is more suitable for describing NSDI dynamics of alkaline-earth metals. We measure single and double ionization yields of four different alkaline-earth metals versus laser intensity. We find strong target dependence of NSDI behaviors. The higher ionization potential of alkaline-earth metal, the more suppressed ratio of doubly to singly charged ion yields measured. We demonstrate that this observation is attributed to the variation of returning electron impact excitation and ionization cross sections for different alkaline-earth metals. Our work shows that NSDI of alkaline-earth metals can be generally understood within the context of recollision scenario. This has important implications for ultrafast manipulation of electron correlation and dynamics of the excited states of alkaline-earth metal ions in NSDI since the returning electrons can be well controlled by shaping laser fields.

2. Experimental setup

In the present experiments, the mid-infrared (2000 nm) femtosecond laser pulses are generated from an optical parametric amplifier (OPA, TOPAS-C, Light Conversion, Inc.), pumped by a commercial Ti:sapphire laser system (2.5 mJ, 40 fs, 1 kHz). The variation of pulse energy is achieved by an combination of an achromatic half-wave plate and a polarizer before focused into the vacuum chamber. A home-made time-of-flight (TOF) mass spectrometer is employed for detecting the singly and doubly charged ion signals. The reader is referred to [48,49] for the details about the apparatus. By means of a turbomolecular pump and a cryopump, the base pressure in the spectrometer is below 10^{-8} mbar. The spectrometer mainly consists of a 1-cm extraction region and a ~45-cm field-free drift region. To circumvent the focus volume effect, a grounded copper slit with a width of 0.5 mm is mounted at the entrance to the drift region. This limits the collection of ions to those arising from the beam waist with a parallel beam geometry along the laser beam direction [50,51].

Collimated atomic beams are produced by an electrically heated stainless atomic oven. The atomic density in the ionization area maintains stable during the measurements by continuously monitoring the temperature of the oven. Ionization signals are detected by a microchannel plate

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located at the end of the spectrometer. The ion signal is amplified, discriminated and then recorded by a multihit time digitizer. The integrations of the ion signals have been normalized to per laser shot for each laser intensity. For high statistical accuracy, up to 10^6 laser shots are measured for lower intensities. The laser peak intensities are calibrated by comparing the measured saturation intensity of ionization of Xe at 2000 nm with the Ammosov-Delone-Krainov (ADK) calculation [51]. The uncertainty of the intensity is estimated to be about 10%.

3. Results and discussions

Figure 1 shows the measured single and double ionization yields of different alkaline-earth metal atoms by 2000 nm laser pulses. For Sr or Ba with relatively low ionization potential, single ionization is saturated already below 8×10^{13} W/cm^2 or 6×10^{13} W/cm^2. The double ionization yield thus exceeds the single ionization yield due to its depletion above this intensity. Notably, the "knee" structure of double ionization yield representing of NSDI can be identified for both Mg and Ba results. Apparently, the "knee" is more suppressed for Mg. Interestingly, for Ca and Sr such structure seems absent. This, however, dose not indicate that NSDI contributions are missing for these two targets. In Fig. 2 we show the experimental ratios of doubly to singly charged ions with respect to the ADK calculations using the single active electron approximation. One can see that significant NSDI channels exist for all the targets. Note that for each target, the range of the NSDI ratio is limited by the range of the measurement of the doubly charged ion yield. The experimental NSDI ratio displays a strong target dependence, i.e., it decreases when the first and second ionization potentials of the atoms become higher, which is similar to that observed for rare gas atoms (see, e.g., [52]).

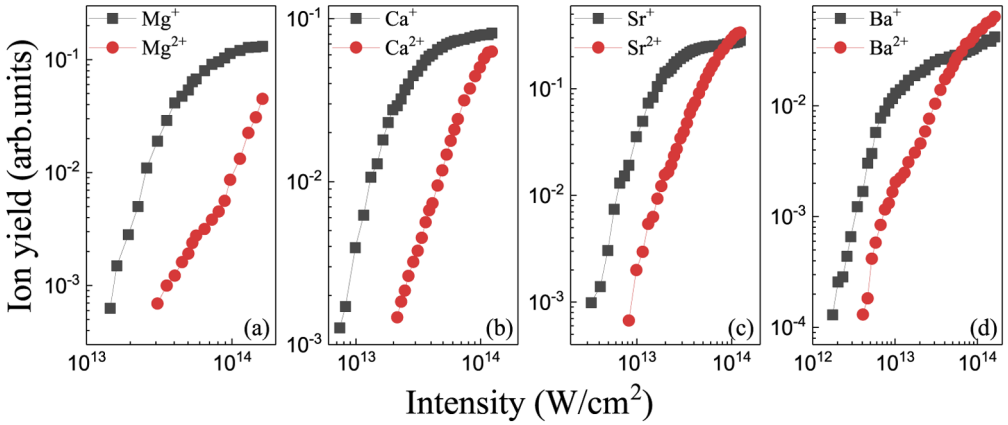

Fig. 1. Experimental singly and doubly charged ion yields of four different alkaline-earth metal atoms with 2000 nm, 45 fs pulses.

To interpret the experimental data, we resort to a 3D Monte Carlo model fully considering recollision impact ionization and excitation cross sections. In this model, NSDI occurs via recollision excitation with subsequent ionization (RESI) and recollision-induced direct ionization (RDI) pathways. The NSDI ratio can thus be expressed as

$$R = \frac{\int dE\sigma_e(E)W_r(E)\frac{e^{-b^2/a_e^2}}{\pi a_e^2} \cdot P_{OBI} + \int dE\sigma_i(E)W_r(E)\frac{e^{-b^2/a_i^2}}{\pi a_i^2}}{\int dEW_d(E)}, \quad (1)$$

where $\sigma_e(E)$ and $\sigma_i(E)$ are the field-free impact excitation and ionization cross sections, respectively. These values can be inferred from previous measurements with electron guns. For

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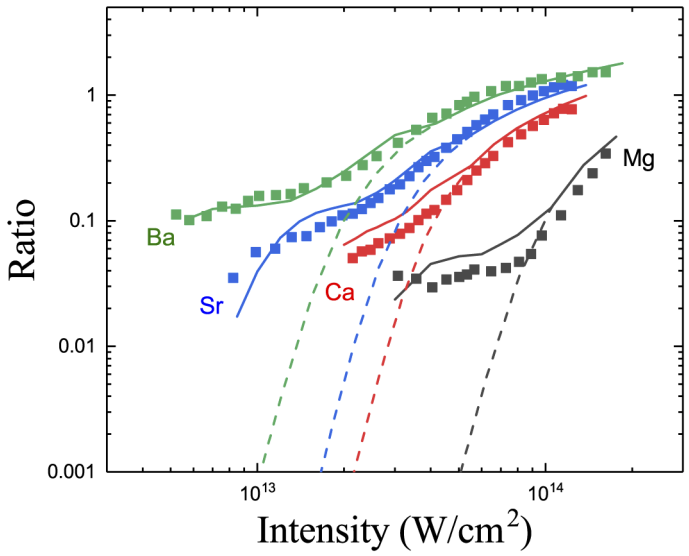

Fig. 2. Experimental (dots) and calculated (solid and dashed lines) intensity dependent ratios of doubly to singly charged ion yields for different alkaline-earth metal atoms. The dashed lines represent the ADK calculations with the same experimental parameters. The solid lines are the sum of the calculations from Eq. (1), which are performed only for the "knee" regions, and the ADK calculations.

the RESI pathway, we consider Mg^{+*} 3p, Ca^{+*} 4p, Sr^{+*} 5p, and Ba^{+*} 6p excited states in the calculations of Mg, Ca, Sr, and Ba, respectively. $W_r(E)$ is the energy distribution of the returning electrons, b is the impact parameter, and $a_{e,i} = \sqrt{2/\triangle E}$, where $\triangle E$ is the excitation or ionization energy for RESI or RDI mechanism. $W_d(E)$ is the energy distribution of the directly ionized electrons that account for the single ionization events. In contrast to previous studies [30,33,34], the excited electrons are ionized with an intensity dependent probability P_{OBI}. Because of the low ionization potential of the excited states of alkaline-earth metal ions, over-barrier ionization (OBI) of the excited electrons takes place for the intensities of interest here. For each target, the threshold intensity for OBI is much lower than the intensities used in our experiments (see Table 1 below). Hence, it is a reasonable assumption that the OBI mainly occurs within the half cycle after recollision. We note that the physical picture in the present study is still valid if the OBI lasts for several cycles after recollision. The OBI probability is

$$P_{OBI} = 1 - e^{-\int_{t_r}^{t_r+0.5T} w_{OBI} dt}, \tag{2}$$

where the OBI rate is given by an empirical formula [53]:

$$w_{OBI} = w_{ADK} e^{-\alpha(Z_c^2/I_{p*})(F/k^3)}. \tag{3}$$

Here w_{ADK} is the ADK rate [54], α is a fitting parameter, Z_c is the charge of the ionization products, I_{p*} is the ionization potential of the excited states of singly charged ions, F is the laser electric field, and $k = \sqrt{2I_{p*}}$. Since the values of α in Eq. (3) are not available for alkaline-earth metal ions, we use the values as listed in Table 1 to fit the experimental results.

To obtain $W_r(E)$ and $W_d(E)$, 3D Monte Carlo calculations have been performed. For each target, an ensemble of 2×10^6 trajectories is randomly distributed in the time interval $-\pi/2 \le \omega t_0 \le \pi/2$. Here ω is the laser frequency and t_0 is the tunneling ionization instant of the electron. The electron trajectories at each time step during the laser pulse are calculated by solving $\frac{d^2\mathbf{r_i}}{dt^2} = -\mathbf{F}(t)$,

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Table 1. The values of α used in our calculations for different alkaline-earth metal ions in the excited states. The corresponding threshold intensities for OBI are also shown.

	Mg^{+*} 3p	Ca^{+*} 4p	Sr^{+*} 5p	Ba^{+*} 6p
α	9.1	9.5	8.9	3.0
I_{th} (10^{12} W/cm^2)	12.7	5.8	4.1	2.8

where the laser electric field is $\mathbf{F}(t) = \mathbf{F_0} f(t) \cos \omega t$ and the pulse envelope function $f(t)$ is a constant equal to 1 for the first 10 cycles and reduced to 0 with 3-cycle ramp in the form of $\cos^2$. We collect the trajectories return to the ionic core to obtain $W_r(E)$. The weight of the trajectories is given according to tunneling ionization theory [55,56]. The tunneling exit point is determined by $z_0 = -I_p/F(t_0)$, where I_p is the first ionization potential of atoms. The electron is assumed to have zero initial longitudinal velocity and an initial transverse velocity in the current calculations.

The calculated results are presented along with the experimental data in Fig. 2. For Mg, we only consider the RESI channel because the RDI contribution to NSDI is negligible [56]. For other atoms, both RESI and RDI channels are considered because the impact ionization cross section can not be ignored. From Fig. 2 we find overall good agreement with the experimental results for all the targets. The small discrepancies possibly result from the uncertainties of the experimental cross sections. Our results suggest that unlike Mg, both recollision induced excitation and ionization cross sections are important for NSDI of Ca, Sr, and Ba. This can be clearly seen in Fig. 3 where we show the previously measured excitation and ionization cross sections for different alkaline-earth metal ions as used in our calculations. We also calculate the returning energy distributions for the two intensities located at the left and right sides of the "knee" structure for each target. As seen in Fig. 3, these distributions exhibit features similar to previous studies [30,56], i.e., the rate increases continuously to a peak with a sharp cutoff in the high energy region. Due to the existence of the tunneling exit, the cutoff position is slightly larger than $3.17U_p$ (U_p is the ponderomotive energy). The ionization cross section is about one order of magnitude smaller than the excitation cross section for the range of returning energy distributions of interest for Mg^+. For other ions, however, the ionization cross section is much closer to the excitation cross section especially for higher energies. The total cross section for the energy range of interest becomes higher for the target with lower (the second) ionization potential, resulting in the observed target dependent NSDI ratio in Fig. 2.

Next we show that the relative contribution of RESI or RDI to NSDI can be adjusted by simply changing laser intensity. The ratio between the recollision induced excitation events and the total events leading to NSDI is

$$Rr = \frac{\int dE \sigma_e(E) W_r(E) \frac{e^{-b^2/a_e^2}}{\pi a_e^2}}{\int dE \sigma_e(E) W_r(E) \frac{e^{-b^2/a_e^2}}{\pi a_e^2} + \int dE \sigma_i(E) W_r(E) \frac{e^{-b^2/a_i^2}}{\pi a_i^2}}. \quad (4)$$

Figure 4 shows the calculated results for Ba. The mechanism is similar for other targets. As the intensity is decreased, the RESI contribution to NSDI is clearly enhanced. This faithfully follows the trend of the excitation and ionization cross sections as functions of the returning energy [Fig. 3(d)]. For very low intensities below 8×10^{12} W/cm^2, NSDI mainly happens via RESI, making it a good candidate for studying the dynamics of ionic excited states in NSDI of Ba. The situation is similar for other alkaline-earth metals considering the extremely fast decrease of the ionization cross section for lower returning energies, where the excitation cross section shows a maximum (Fig. 3). In future, it is promising to obtain more valuable information of RESI or RDI mechanism from the differential measurements of photoelectrons from double ionization of alkaline-earth metals.

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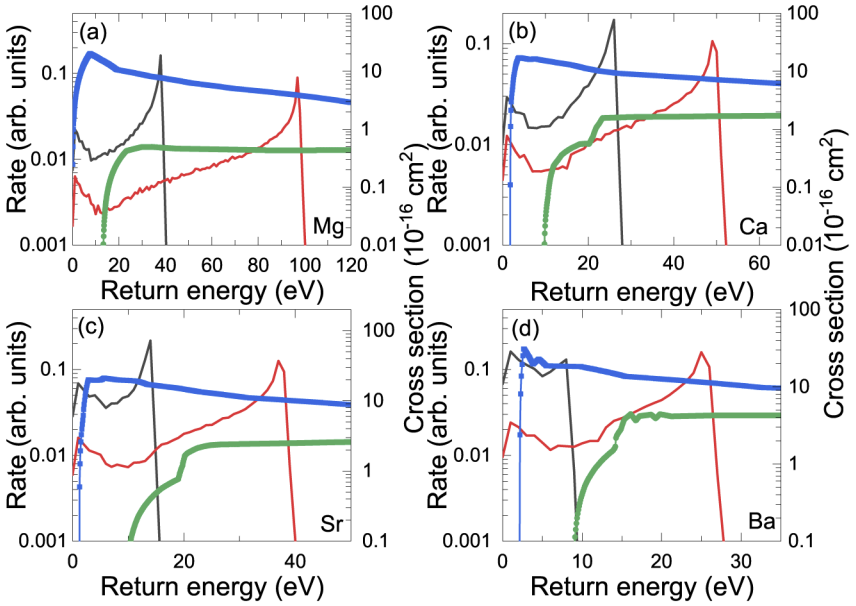

Fig. 3. The excitation (blue dots and lines) and ionization (green dots and lines) cross sections for different alkaline-earth metal ions as used in our calculations. The data of excitation cross sections are taken from the measurements in Refs. [57,58]. The experimental ionization cross sections have been summarized in Ref. [59] (see also the references therein). The returning energy distributions (black and red lines) for the two intensities at the left and right sides of the "knee" region of Fig. 2 are also shown for each target. These intensities are 3×10^{13} W/cm^2 and 8×10^{13} W/cm^2 in (a), 2×10^{13} W/cm^2 and 4×10^{13} W/cm^2 in (b), 1×10^{13} W/cm^2 and 3×10^{13} W/cm^2 in (c), 5×10^{12} W/cm^2 and 2×10^{13} W/cm^2 in (d).

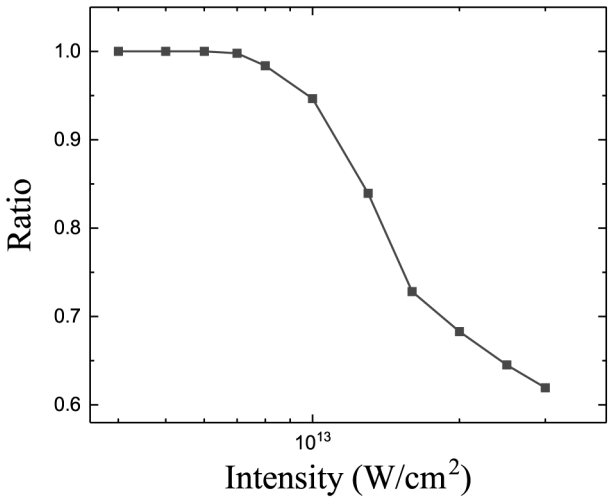

Fig. 4. Calculated relative ratio of the recollision induced excitation events as a function of intensity according to Eq. (4) for Ba.

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So far we have demonstrated that the recollision impact excitation and ionization cross sections play a crucial role in NSDI of alkaline-earth metals. We anticipate that this physical picture is also valid for other atomic targets with complex structures provided that the quasistatic field approximation applies. Looking forward, this provides a general and characteristic knob to control strong-field double ionization on subcycle timescale as the returning electrons can be well controlled by sculpted laser fields such as two-color and phase controlled few-cycle pulses. In turn, ultrafast dynamics of impact excitation and ionization processes is also possibly inferred from the measurements of recollision induced double ionization.

4. Conclusion

In conclusion, we have investigated NSDI of four alkaline-earth metal atoms subjected to intense mid-infrared femtosecond pulses. The measured ratio of doubly and singly charged ions corresponding to NSDI displays a strong dependence on the target. It is found to be decreased when the ionization potential of the atom is higher. The experimental results are faithfully reproduced by 3D Monte Carlo calculations considering recollision impact excitation and ionization cross sections. We show that the observation is due to the change of excitation and ionization cross sections for different alkaline-earth metals. We expect that this mechanism is applicable to other atomic targets with complex structures. It thus offers a general and novel way to control electron correlation and dynamics of ionic excited states in NSDI through steering the recolliding electrons.

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Disclosures

The authors declare no conflicts of interest.

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