Supporting information for "Field intensity dependence of the dissociative multiple ionization of argon dimers in strong femtosecond laser fields"

I. THE EFFECT OF LASER POLARIZATION

The two fragmental ions produced by the dissociative double ionization channel $Ar_2^{2+} \rightarrow Ar^+ + Ar^+$ are Ar^+ ions, both of which are indistinguishable by the detector. The cold-target recoil-ion momentum spectrometer (COLTRIMS) system artificially labels the ions as ion 1 and ion 2 based on their arrival times at the detector. The ion detected first is labeled ion 1, whereas the second ion is labeled ion 2. According to the law of conservation of momentum, ion 1 and ion 2 initially move in opposite directions. Ion 1 flies directly toward the detector, whereas ion 2 first moves away from the detector before being redirected toward the detector by an accelerating electric field. Consequently, in the direction of acceleration electric field of momentum spectrometer (taken to be the z direction), ion 1 has only positive momentum, and ion 2 has only negative momentum.

To obtain the complete momentum distribution of the fragmental ion Ar^+, we plot the momentum distributions of ion 1 and ion 2 together, as shown in Fig. 1. The complete momentum distribution is plotted over the laser polarization plane (xoz plane), for which the x axis represents the direction of the molecular beam and the z axis represents the direction of acceleration electric field of momentum spectrometer. Figure 1(a) and 1(b) show situations when the laser polarization is in the x and z directions, respectively, with both measured at a laser intensity of 260 TW/cm^2. The momentum spectra exhibit white horizontal lines near $P_z = 0$, indicating missing data due to the dead time of the detector. This occurs when two fragmental ions have the same initial momentum in the vertical z direction and arrive at the detector at the same time. This data gap leads to inaccurate counts that ultimately affect the experimental results concerning the laser polarization. For instance, a circularly polarized laser produces fewer counts than a linearly polarized laser.

II. EXPERIMENTAL RESULT

Kinetic-energy release (KER) distributions of Ar^+ ion pairs from the $Ar_2^{2+} \rightarrow Ar^+ + Ar^+$ channel were plotted on a logarithmic scale in Fig. 2(a) and 2(b). To eliminate the influence of the detector dead time on the fragment ion count, experimental measurements were made in the two polarization directions: the x direction in Fig. 2(a), and the z direction in Fig. 2(b). Both plots reveal a distinct three-peak structure, where peak A corresponds to a KER value of 3.8 eV, peak B to 5.4 eV, and peak C to 7.4 eV. The KER spectra at various different laser intensities were normalized using the maximum number of events. Figure 2(a) and 2(b) demonstrates that the relative distribution of released kinetic energy changes with laser intensity, significantly affecting the relative intensities of peaks B and A, which decrease as the laser intensity increases. The pathway corresponding to peak B contributes more to double-ionization dimer dissociation at low laser intensities. The results obtained under both laser polarization conditions are consistent. However, Fig. 2(a) shows that the variation in results is not significant at laser intensities of 390 TW/cm^2 and 970 TW/cm^2.

For peak C, the results in Fig. 2(a) do not exhibit a clear physical regulation. We speculate that this phenomenon may be attributed to two causes: (1) there are fewer dimer dissociation events, and the data collection time is not long enough, and (2) low momentum resolution of the COLTRIMS system in the x direction. We present the experimental results displayed in Fig. 2(b) in the main text. Figure 2(c-g) shows the sum momentum distribution of the fragment ion pair Ar^+/Ar^+ in the laser polarization direction at the five different laser intensities plotted in Fig. 2(b). The laser intensities are 260 TW/cm^2, 330 TW/cm^2, 470 TW/cm^2, 810 TW/cm^2, and 1020 TW/cm^2, respectively; the peak structures A and B are indicated by red dotted dash lines and green solid lines, respectively. For comparison, we also provide the momentum distribution of synchronously collected Ar^{2+} ions (blue dotted line) in the laser polarization direction that were derived from the double ionization process of the Ar atomic monomer. The momentum distribution of Ar^{2+} ions along the laser polarization direction shows significant modulation with changes in laser intensity. It exhibits a characteristic "double hump" structure at 260 TW/cm^2 and 330 TW/cm^2, indicating nonsequential double ionization caused by rescattering. At increased laser intensities, the momentum distribution of Ar^{2+} ions changed to a Gaussian distribution with the peak at zero. This suggested that ionization of the two electrons occurs near the maximum value of the oscillating field, which is a typical sequential tunneling ionization. It is worth mentioning that the full width at half maximum (FWHM) of the Gaussian distribution corresponding to the blue dotted line in Fig. 2(e) is wider than that in Fig. 2(f) and 2(g), indicating that the two ionization mechanisms of non-sequential and sequential double ionization work together under this light intensity.

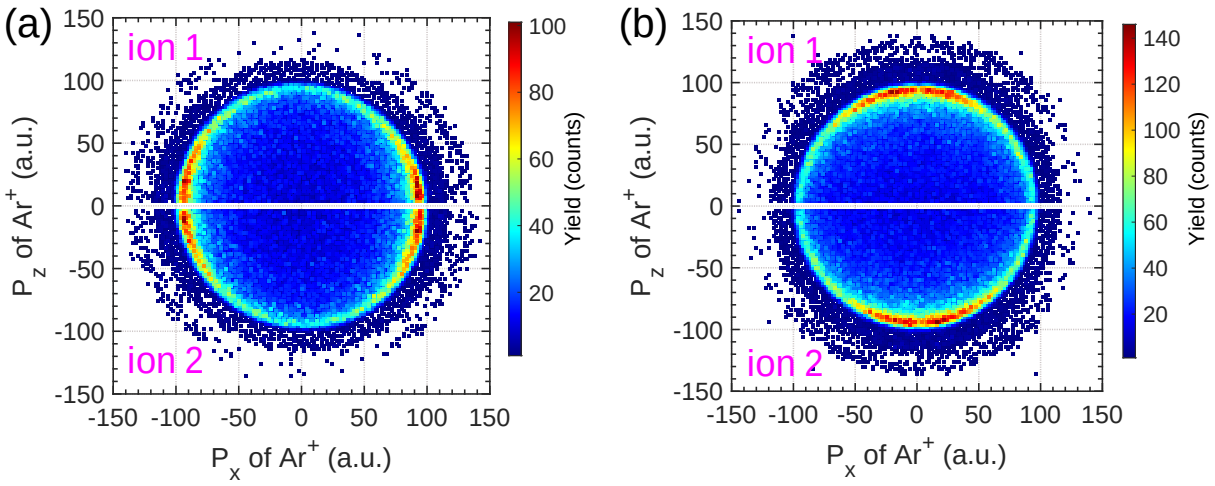

FIG. 1. Momentum distribution of fragmental ions produced in the breakup channel $Ar_2^{2+} \rightarrow Ar^+ + Ar^+$ in the xoz plane; here x represents the direction of the molecular beam and z the direction of acceleration electric field of momentum spectrometer. In panels (a) and (b), the laser polarization is in the x direction and z direction, respectively; the laser intensity is 260 TW/cm^2.

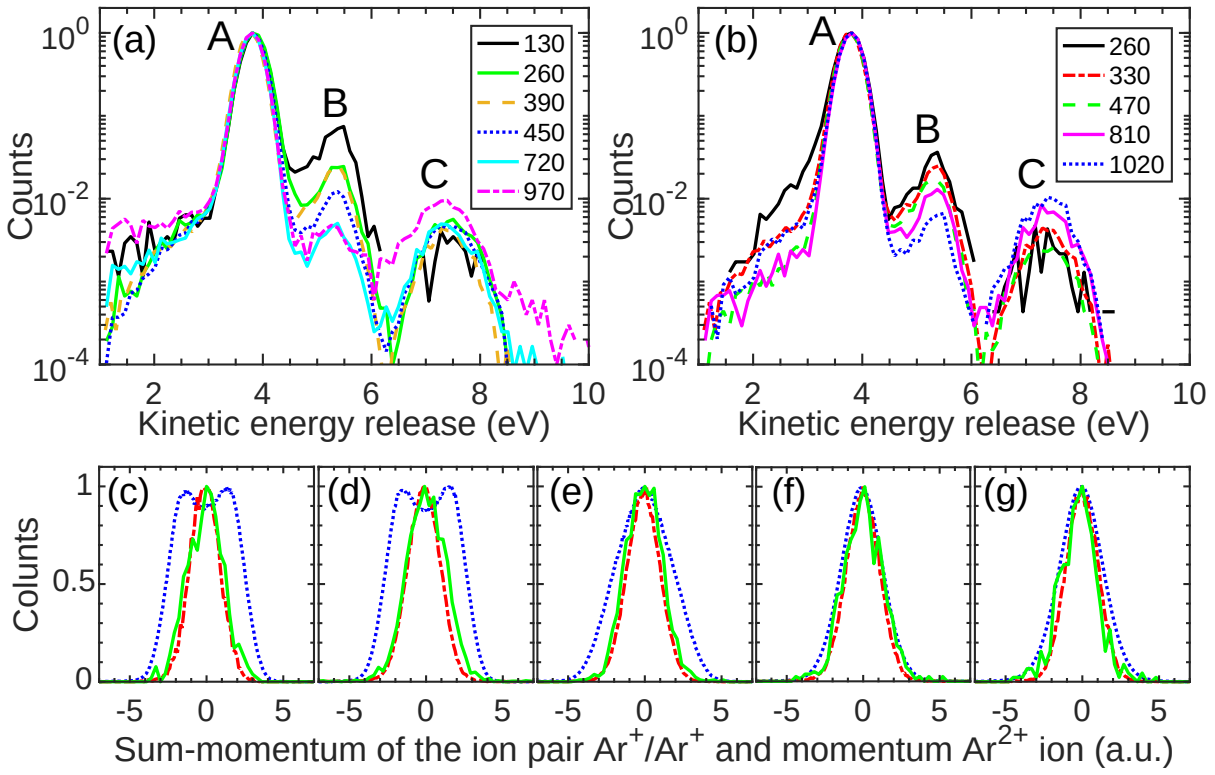

FIG. 2. The KER distributions from the dissociative double ionization channel of Ar dimers under different laser intensities: (a, b) the laser polarization is along directions x and z, respectively. (c–g) The sum momentum distributions of the fragment ion pair Ar^+/Ar^+ in the laser polarization direction under several laser intensities plotted in (b); specifically, the laser intensities are (c) 260 TW/cm^2, (d) 330 TW/cm^2, (e) 470 TW/cm^2, (f) 810 TW/cm^2, and (g) 1020 TW/cm^2, where the red dotted dash line corresponds to peak A and the green solid line to peak B. The blue dotted line is the momentum distribution of synchronously collected Ar^{2+} ions in the laser polarization direction that derives from the double ionization process of the Ar atomic monomer.