Triply charged carbon dioxide molecular ion: Formation and fragmentation

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In an experiment involving detection of a photoelectron and up to three photoions from CO_2 in coincidence, we observe the triple ion coincidence $C^+:O^+:O^+$. Moreover, we observe double coincidences between doubly charged cations and singly charged cation pairs $C^{2+}:O^+$, $O^{2+}:C^+$, $O^{2+}:O^+$. These ion triplets and pairs arise from fragmentation of the triply charged molecular ion CO_2^{3+}. Other ion pairs—viz., $C^+:O^+$, $O^+:O^+$, $O^+:CO^+$—arising from the doubly charged molecular ion CO_2^{2+}, are also observed. From an analysis of the coincidence pattern we postulate four decay modes of the CO_2^{3+} ion. Kinetic energy release in the channel leading to $C^+:O^+:O^+$ is measured, and its distribution is postulated to have four contributing precursor states.

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Small molecules like N_2, CO, CO_2, or H_2O, when stripped of a few electrons, are usually unstable and fragment, forming neutral and charged fragments. While stripping of several electrons is most easily accomplished by ion bombardment and intense laser fields, single-photon absorption leading to multiple-electron emission is a rare process. Fragmentation of highly charged cations of N_2, CO, and CO_2 formed by charged-particle impact or intense laser fields has been observed in several experiments [1–6]. The energetics of a two-body fragmentation in which no neutral fragments are formed is often explained by the Coulomb explosion model, which is usually found to be inadequate to explain experimental values of the kinetic energy released (KER) in the reaction [7]. Experimental verification of the model necessitates detection of the charged particles in coincidence with each other and measurement of their kinetic energies. For triatomics, the fragmentation pattern is far more complex. Besides two-body fragmentation, three-body fragmentation and two sequential two-body fragmentations are also possible. The complete kinematics of a three body fragmentation can be directly deduced only when all three fragments are ions. If one of the fragments is neutral, the kinematics can only be deduced indirectly, provided the momenta of the two ionic fragments are completely determined. The only previous report of coincident detection of three fragments from a triatomic molecular ion is that of H_2O^{3+}, formed by 350 keV H^+ impact on H_2O, by Werner *et al.* [8]. Tian and Vidal have observed ion pair coincidences in triple and quadruple ionization of CO_2 by electron impact [4]. Using the triple-ion-coincidence momentum imaging technique, Muramatsu *et al.* [9] have reported the observation of the static Renner-Teller effect in CO_2.

We report here the formation of CO_2^{3+} by photoionization of CO_2, and its fragmentation via four distinct channels leading to the end products (i) $C^+:O^+:O^+$, (ii) $C^{2+}:O^+$, (iii) $O^{2+}:C^+$, and (iv) $O^{2+}:O^+$. The first of these channels is detected in a three-particle coincidence and the last three as two-particle coincidences. Data were recorded at a few energies in the range 40–320 eV, spanning the oxygen *L* edge and the carbon *K* edges. We focus mainly on the data at 200 eV, which is off the shell energies of both oxygen and carbon. Of the photon energies at which data were recorded, the triple-ionization yield was found to be maximum at 200 eV, after normalization for photon flux and target density. The results presented here are based on this data set alone, to emphasize the nonresonant character of the triple-ionization process. Moreover, the photon flux was highest at this energy, leading to better counting statistics.

The experiment was performed at the Indus-1 synchrotron at Indore. This is a 450-MeV second generation storage ring, with radiation flux peaking at 202 eV (6.1 nm). The experimental station is an improved version of the one described in an earlier paper [12]. The photon beam was monochromatized by a toroidal grating monochromator. A polyamide filter was used to block the second-order diffracted beam from reaching the target, while simultaneously eliminating additional gas load on the grating chamber. The monochromatized photon flux ranged between 10^{10} s^{-1} and 10^{11} s^{-1} with energy resolution $E/\Delta E \approx 300$. The beam spot at the target is 1 mm^2. An effusive beam of CO_2 intersected the photon beam. The number density of the target gas was 10^{12} cm^{-3}. Photoelectrons and photoions formed in a small interaction volume ($\sim$2 mm^3) were extracted in opposite directions by an electric field of strength 300 V/cm, while maintaining the interaction volume close to ground potential. The extraction field, the photon beam, and the molecular beam were mutually orthogonal, with the extraction field parallel to the photon polarization axis. This electric field formed the first acceleration stage of a two-stage ion time-of-flight spectrometer. After the 10-mm-long first stage, ions entered a 6-mm-long second acceleration stage, where the field was 580 V/cm, followed by a 63-mm field-free drift. Ions were detected by a 40-mm-diam channelplate. The large detection area ensures that the lightest singly charged ion C^+ is not lost if its energy is less than 30 eV. Electrons were detected by a channeltron placed just beyond the extraction stage. A 3-mm aperture in front of the channeltron reduces the geo-

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