Energetics of High Charge State Ionization: An Example From Carbon Monoxide

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We present yields and molecular fragment energies for the ionization of carbon monoxide in a laser field with intensities spanning from 10^{14} W cm^{-2} to 10^{17} W cm^{-2}. The goal of the project is to study the energetics of ionization in a strong and ultrastrong laser field. During this increase in intensity, the charge state progresses from the removal of only one- or two-electrons to the ionization of all electrons beyond the 1s electrons for the ions.

The experiments measure the ion yields and ion energy spectra. The data is collected over several orders of magnitude in signal range to constrain the evaluation of agreement with theory rigorously. Energy and angular resolved ion fragment distributions are also collected. The experiments use a TEM$_{00}$, 40 fs, terawatt CPA laser system. These laser pulses enter an ultra-high vacuum time-of-flight ion spectrometer designed for collections with energies from 0 eV to 200 eV. ^{12}C, ^{13}C, ^{16}O, and ^{18}O isotopes are used to eliminate m/q degeneracy. The highest ion energies observed for C^{n+} and O^{n+} ($1 < n < 3$) extend to 100 eV. Counterintuatively, for C^{m+} and O^{m+} ($4 < m$) the highest ion energies observed decrease to < 60 eV.

The molecular calculations use a complete active space self-consistent field method with molecules in a quasi-static strong external field (CASSCF+SF). CO$^+$ ion dynamics are solved with the time-dependent Schrödinger equation (TDSE) using the field-dependent CASSCF+SF solutions that follow the classical pulsed laser field. The agreement between the calculated HOMO tunneling ionization yields with the observed experimental yields indicates that, while multiphoton ionization and non-sequential ionization are present, tunneling is the primary ionization rate for the interaction at saturation. The kinetic energy of the ion wave function is assumed to be unchanged during the ionization process, i.e., the field energy from the interaction is assumed to be removed by the outgoing electron and the molecular electronic wave function is left in the lowest energy state for the external field at the 'instantaneous' ionization event.

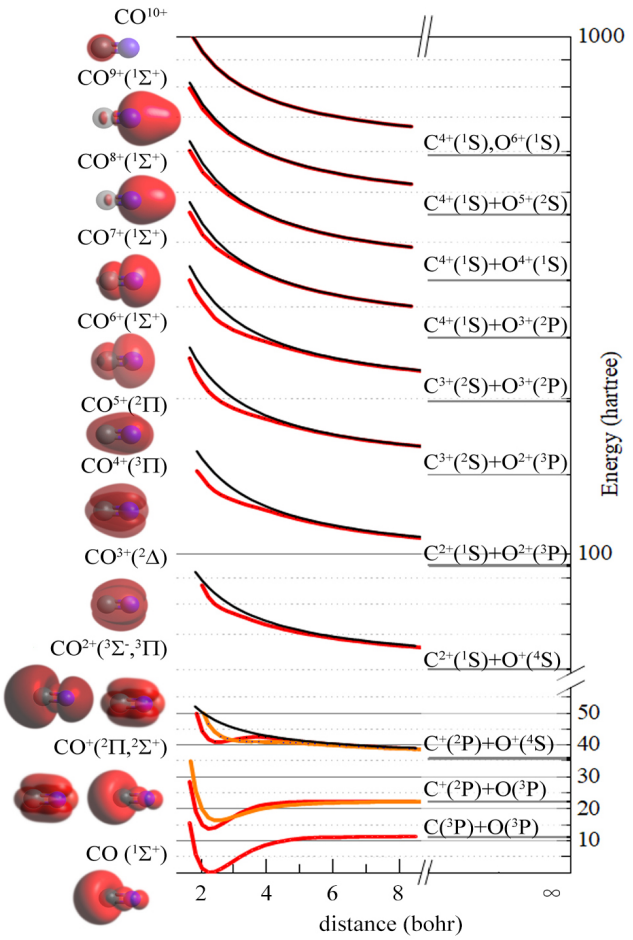

Figure 1: Calculated potential energy as a function of the intramolecular separation for CO^{n+}. The lowest energy configuration (red line) is shown for all species except CO$^+$ and CO^{2+} where the next highest energy bond (orange). For multiply charged molecular fragments, the Coulomb repulsion potential energy curve (black line) is also shown for the most stable configuration of the asymptotic atomic ion states, which are identified with their charge and term at an infinite intramolecular distance. For reference, the highest occupied molecular orbital (HOMO), and HOMO-1 for CO$^+$ and CO^{2+}, are shown to the left of the corresponding potential curve calculated for an intramolecular separation of 2.13 bohr (112 pm) with no external field

With this approximation, the experimental ion energies cannot be explained without invoking recollision ionization.

In summary, we present experimental and theoretical ion yield and ion fragment energy calculations at intensities beyond 10^{17} W cm^{-2} for the progression of the ionization up to CO^{10+}, leaving only the core 1s states of ionic carbon and oxygen remaining. The mechanisms involved include Coulomb explosion, recollision non-sequential ionization, and the time-dependent rise of the ultrafast laser pulse envelope as ionization proceeds over orders of magnitude in intensity.

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