Induced Dipoles, Stark Shifts, and Wave Function Dynamics in a Strong Laser Field

B C Walker[1], E C Jones[1], A U Rehman[1], J Wisely[1], and K Szalewicz[1]

[1]*Physics and Astronomy, University of Delaware, 104 The Green, Newark DL, USA.*
Contact Phone: +13027400257
Contact Email: bcwalker@udel.edu

Molecular dynamics in strong laser fields observed over the years include alignment, pendulum states, dissociative ionization, stabilization, wave packet evolution on multiple electron states, multiple fragmentation channels, enhanced ionization, frustrated tunneling ionization, rescattering nonsequential ionization, and Coulomb explosion. The explanations for these employ the approximations of separable nuclear and electron wave functions and a treatment of the electron states as either field-free, or superposition of field-free, states. Molecular tunneling ionization in a strong laser field, for example, uses a field-free approximation for the molecular bound state [1].

Advances in computational speed and techniques now allow insight into the motion of wave functions in response to external fields. The changes in these wave functions on attosecond time scales are profound and challenge the approximations routinely used to date. We report on the molecular response of CO, CO^+, and CO^{2+} to strong external fields beyond the field-free approximation. The impact of the field on the bound wave function can be seen in Fig. 1. The HOMO for CO^+ with no external field is shown in Fig. 1(a). The HOMO in an external field of 0.15 $E_h a_0^{-1} q^{-1}$ (corresponding to the saturation intensity in the experiment) is shown in Fig. 1(b). Though the HOMO is still in a sigma state, a glance at Fig. 1(a,b) shows the reorganization of the charge is significant. The charge density (i.e. $\psi^*_{HOMO}\psi_{HOMO}$) in the field minus the charge density with no external field is plotted in Fig. 1(c). One can estimate visually from the figure that approximately one-half of an electron migrates 3 bohr during a 500 attosecond, quarter-cycle of the field oscillation.

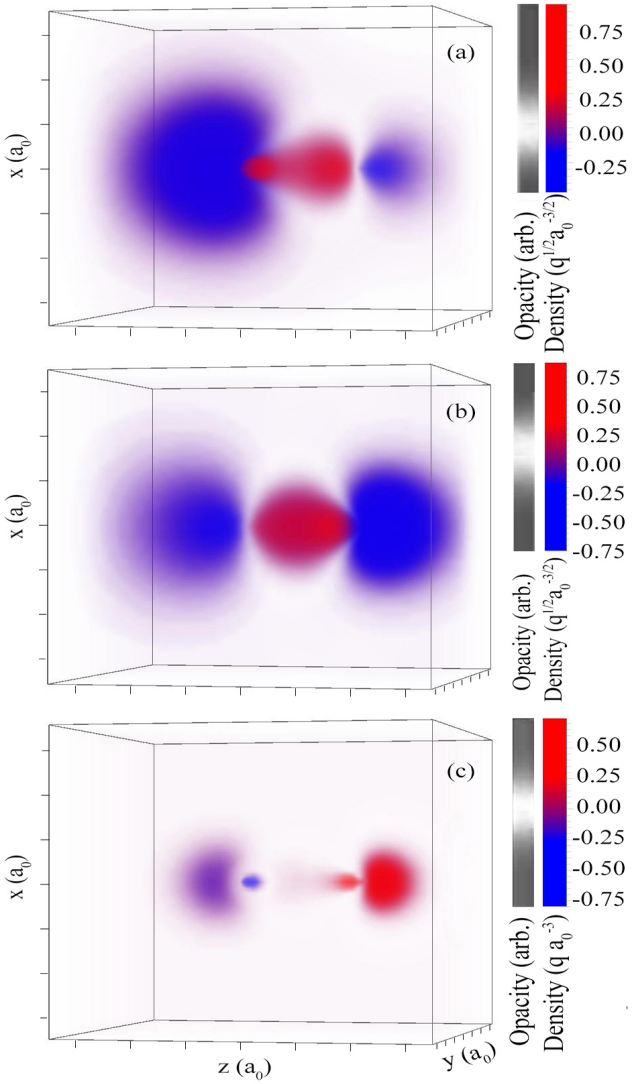

Figure 1: Calculated HOMO for CO^+ with no external field (a) and a field of 0.15 E_h a_0^{-1} q^{-1} oriented from oxygen to carbon. (b) (intensity of 8×10^{14} W cm^{-2}). Carbon is positioned on the left in the figure and oxygen is on the right. The difference in the charge distribution for the HOMO in CO^+ is shown in (c) as the field on minus the field free case. Legends on the side of the figures correspond to the HOMO and charge density in atomic units

The time-dependent evolution of the charge migration in the bound states due to the influence of the sinusoidal external field can be visualized with movies of the wave functions for peak fields at the experimental saturation intensities. The ionization rates for CO, CO^+, and the final dissociated states of CO^{+2} are presented as a function of the external

field intensity using the polarized, and Stark-shifted electronic states in the external field. The calculated ionization rates and dissociation dynamics will be compared to the measure yields [2].

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References

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