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Attosecond dynamics of strong-field multielectron excitation in small molecules: the case of carbon dioxide

STANLEY SMITH, DMITRI ROMANOV, Temple University, XIAOSONG LI, University of Washington, H. BERNHARD SCHLEGEL, Wayne State University, ROBERT LEVIS, Temple University

— The electron dynamics of CO₂ interacting with a short IR (1.63 eV) three cycle pulse was theoretically investigated, as manifested by the instantaneous dipole oscillations. The pulse envelope shape (Gaussian or trapezoidal) is shown to control the residual dipole oscillations spectrum (which excited states are populated) and amplitude (or relative population in each state). The carrier envelope phase (for either envelope shape) changes markedly the excited state amplitudes. The windowed Fourier transform was used to extract the attosecond electron excitation dynamics during the pulse. This analysis shows the nonlinear excitation process to be much more complicated than the traditional excited state ladder climbing. In particular, (i) electron transfer plays a major role in the nonlinear excitation dynamics, and (ii) the two pronounced spectral peaks at ~5 and ~8.5 eV do not correspond to any single-photon dipole-allowed transitions. The electron response in the directions orthogonal to the molecular axis was also simulated and, as expected, this response was smaller in magnitude than the response along the molecular axis.