Alignment-dependent ionization of linear molecules in intense laser fields

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In the past couple of years, several experiments emerged on strong-field ionization from aligned molecules, where the molecular axis is fixed at a particular angle relative to the laser polarization axis. Total ionization yields were measured as a function of alignment angle for ionization from the highest occupied molecular orbital (HOMO) of N2, O2, and CO2 (Fig. 1). The experimental results were modeled using the molecular tunneling theory (MO-ADK), a semi-classical approach that disregards the molecular potential. For N and O2, very good agreement is found between the MO-ADK predictions and the experiment. For CO2, on the other hand, the MO-ADK predicted the maximum ionization yield at an alignment angle of 24 deg, in disagreement with the experimental ionization peak at 45 deg (cf. Fig. 1). The discrepancy between the MO-ADK results and the experiment is attributed partly to inaccurate asymptotic expansion coefficients of the HOMO wave function, which are used as input for MO-ADK. Using wave functions with a better asymptotic form in the MO-ADK analysis results in a maximum ionization yield for CO2 at about 35 deg, still in disagreement with the experiment. By solving the 3D time-dependent Schrödinger equation (TDSE) for the HOMO electron of CO2 within the single-active-electron and the frozen nuclei approximations, the maximum ionization yield for CO2 is found at about 45 deg, in agreement with the experiment. The TDSE results demonstrate an important contribution from the excited states of the molecular potential in strong-field ionization of CO2, and, therefore, explain the breakdown of the molecular tunneling theory.

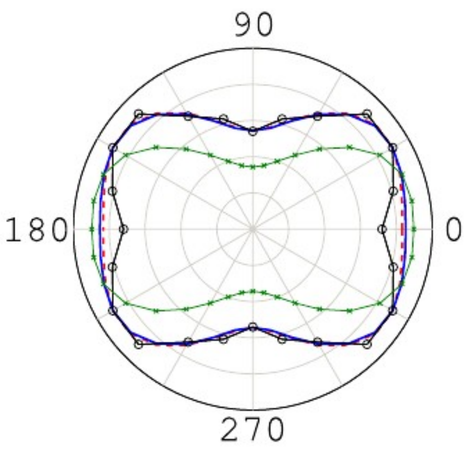

FIG. 1: Experimental (circles) and theoretical (dashed line: 5.6×10^{13} W/cm^2; solid line: 1.1×10^{14} W /cm^2; crosses: MO-ADK) total ionization yield from the HOMO orbital of CO2.