

Single- and double emission from multi-electron systems: How to compute IR double emission and solve the CO₂ mystery

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Abstract

By a new ab initio approach we solve two long-standing problems in strong-field multi-electron systems: fully differential double-emission spectra at IR wave length, where we quantitatively reproduce the measured e-2e ratio and provide an experimentally accessible map of correlated vs. uncorrelated emission in the two-electron energy plane, and the notorious discrepancy between experiment and theoretical prediction for the alignment dependence of strong field CO₂ ionization, which is dominated by dynamical exchange.

Using a combination of newly developed methods [1, 2, 3, 4] we solved several key problems in strong laser-matter interaction such as (1) the computation of precision photo-electron spectra at elliptic polarization (attoclock, [5, 6]), (2) accurate fully differential (angle-resolved) double emission spectra from the He atom and from H₂ with polarization parallel to the molecular axis, (3) single photo-emission from the CO₂ molecule in dependence of molecular orientation. In all cases we can provide dependable accuracy estimates.

Fig. 1 shows near-IR double emission calculations with accuracies of 5% together with energy-dependent correlation. The SVD-based correlation measure allows a systematic experimental distinction of correlated from uncorrelated emission.

Fig. 2 shows the first ab initio computational reproduction of the single to double ionization yield ratio at IR wave length.

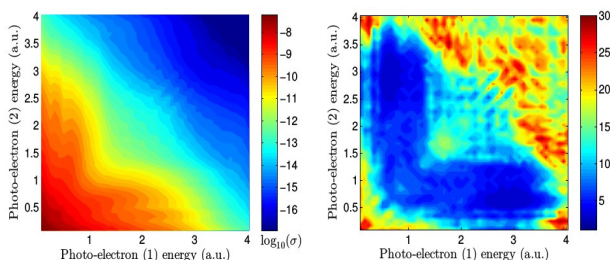


Fig.1. Double emission spectra of He by a 3 cycle 700nm pulse at intensity 3×10^{14} W/cm² (left) and angular correlation (right).

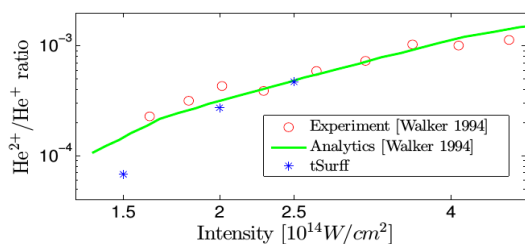


Fig.2. e-2e ratio at 780 nm wave length, experiment (red), fit (green) and ab initio (tSurff, blue).

In the haCC approach [4] for computing emission from molecules, strong field dynamics is combined with quantum chemical structure (COLUMBUS, [7]) including full interchannel coupling and exchange. We find that including exchange is crucial: it explains the observed orientation dependence of ionization of CO₂. Multi-electron effects can clearly be detected, but have no influence on the peak emission angle at 45°. Fig. 3 shows our results and explains the smaller angles reported in literature, Ref. [8].

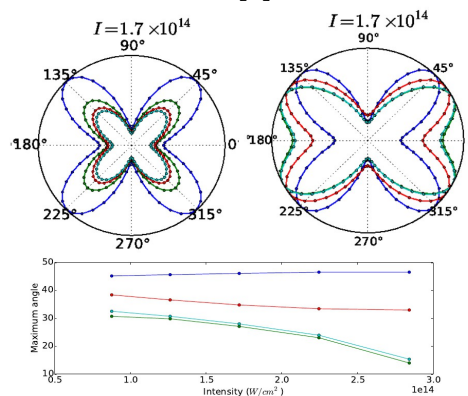


Fig 3. Alignment-dependence of field ionization of CO₂: multi-electron effects reduce the overall yield, but have no influence on angle of peak emission (blue to cyan, left top panel). Lack of anti-symmetrization produces the smaller angles reported in literature (right top panel, red and green). In the full calculation (blue) the emission angle is largely independent of intensity (lower panel).

References

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