Strong-field ionization of aligned molecules

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Synopsis A framework for studying strong-field ionization of aligned molecules is presented, and alignment-dependent ionization yields are computed for CO_2 and CS_2. Our calculations are in unprecedented agreement with recent experiments, and explain the breakdown of the molecular tunnelling theory and strong-field approximation.

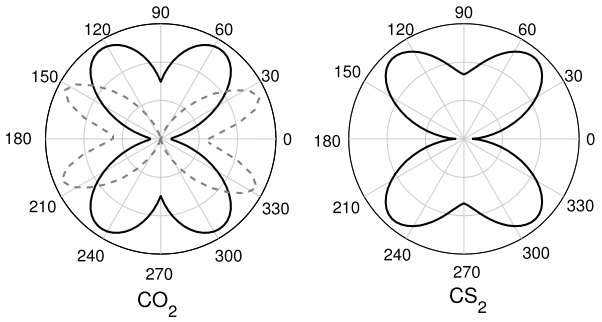

Fig. 1. Ionization yields as a function of the angle β for CO_2 and CS_2. The solid (dashed) line denotes our TDSE (MO-ADK) calculations.

Theoretical studies of strong-field ionization of molecules are impeded by the complexity of the molecular electronic structure. Up till now, full *ab initio* calculations of the alignment-dependent ionization are available only for H_2^+ and H_2. For larger molecules, despite a tremendous amount of experiments, no *ab initio* calculations are available, and the most widely used approaches to explain strong-field processes are the molecular tunneling theory and strong-field approximation. Calculations of alignment-dependent ionization yields based on these theories fail to explain recent experiments: Tunneling theory and strong-field approximation predict the ionization yield to follow the electron density of the initial electronic state, in contrast with observations for the CO_2 molecule. [1]

In [2], we use *ab initio* theory within the single-active electron approximation to investigate the response of polyatomic molecules to intense femtosecond laser pulses. Our approach is grid based, which is the most widely used approach in strong-field physics, and takes input potentials from standard quantum chemistry codes. We only consider the dynamics of the outermost electron (the HOMO orbital): the remaining electrons are accounted for by an effective potential.

The computed ionization yields are shown in Fig. 1 for CO_2 and CS_2 as a function of the angle β. The orientation-dependencies of ionization are generally similar for the two molecules. For CO_2 and CS_2, the ionization yields are largest at $54\pm3^\circ$ and $48\pm3^\circ$, respectively. These results are in unprecedented agreement with recent experiments [1], which predict the ionization yields to peak at about 46°. Our approach is clearly superior to the tunneling theory (cf. the figure) and the strong-field approximation (results not shown here) which predict the ionization yields for CO_2 to peak at about 25°.

References

[1] D. Pavičić, K. F. Lee, D. M. Rayner, P. B. Corkum, and D. M. Villeneuve, Phys. Rev. Lett. **98**, 243001 (2007).

[2] M. Abu-samha and L. B. Madsen (Submitted for publication).

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