Probing Vibronic Coherence in Charge Migration of Molecules Using Strong Field Sequential Double Ionization

C. H. Yuen [1], C. D. Lin
[1] *J. R. MacDonald laboratory, Kansas State University, Manhattan KS, 66502, USA*
iyuen@phys.ksu.edu

We propose a novel scheme for probing vibronic coherence in charge migration in molecules utilizing strong field sequential double ionization. To demonstrate the feasibility of this approach, we perform full simulations of a pump-probe scheme employing few-cycle intense infrared pulses for N_2 and O_2. We predict that the vibronic coherence between the pumped states will be directly imprinted in experimental observables such as kinetic energy release spectra and branching ratios of the dissociative dications. Our simulations are based on the recently developed DM-SDI model [1, 2], which is capable of efficiently accounting for molecular orientations and enabling direct comparison with experimental results. Our findings strongly encourage the use of this probing scheme in future charge migration experiments.

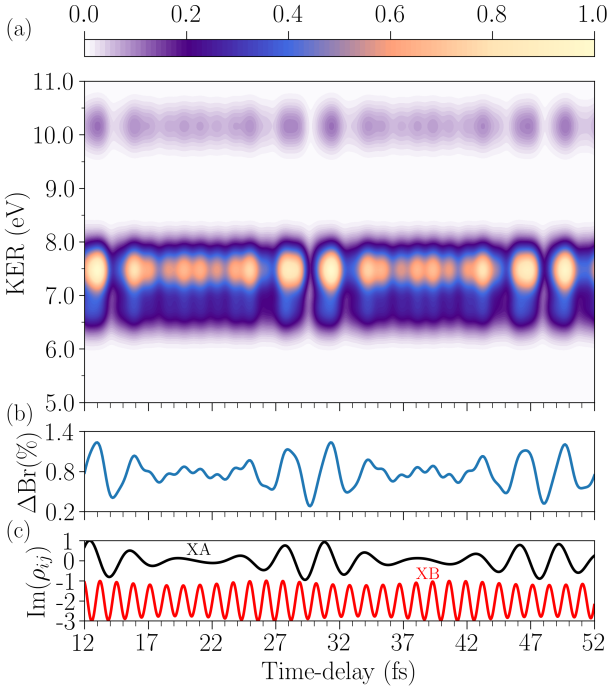

Figure 1: Simulated KER spectra for $N^+ + N^+$(a) as a function of pump-probe delays, subtracted by the respective probe-only signal. (b) The branching ratio of $N^+ + N^+$ over their respective total dication yield as a function of pump-probe delay, subtracted by the probe-only ratio. (c): Imaginary part of the angular-averaged off-diagonal density matrix elements between the $X^2\Sigma_g^+$ and $A^2\Pi_u$ states (black) and the $X^2\Sigma_g^+$ and $B^2\Sigma_u^+$ states (red) of N_2^+.

References

[1] C. H. Yuen and C. D. Lin, Density-matrix approach for sequential dissociative double ionization of molecules, Phys. Rev. A 106, (2022), 023120.

[2] C. H. Yuen *et al.*, Modeling the sequential dissociative double ionization of O_2 by ultrashort intense infrared laser pulses, Phys. Rev. A 107, (2023), 013112.