

Strong Field Tunneling in Systems with Multiple Electrons





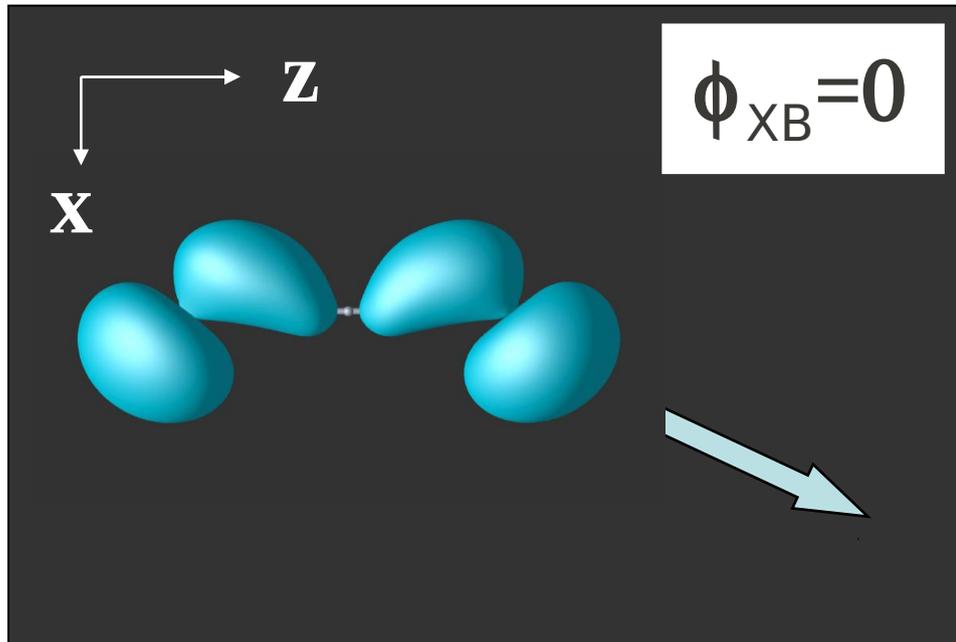
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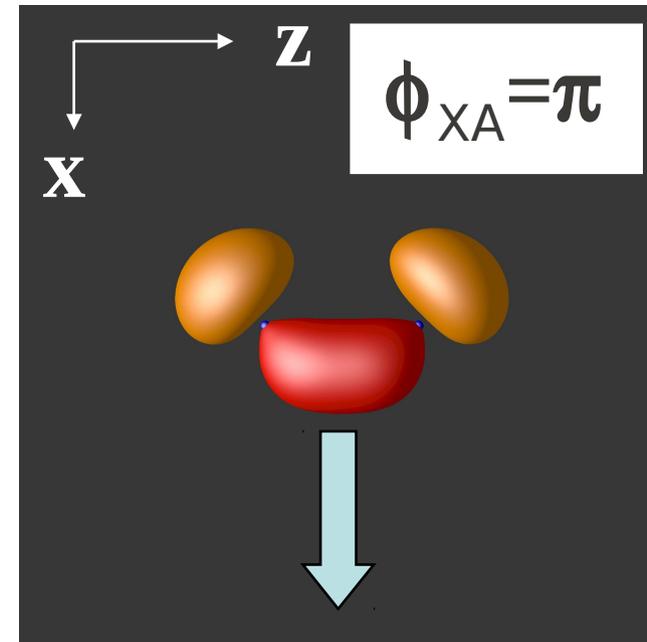
HHG spectroscopy of core rearrangement



**Direction of
electron
escape**

0 phase between channels X and B
after ionization for CO₂

Nature 2009 Smirnova et al



**Direction of
electron
escape**

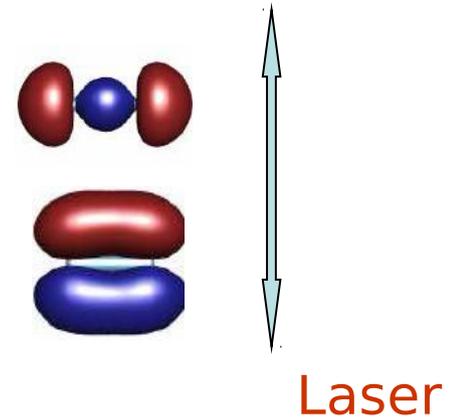
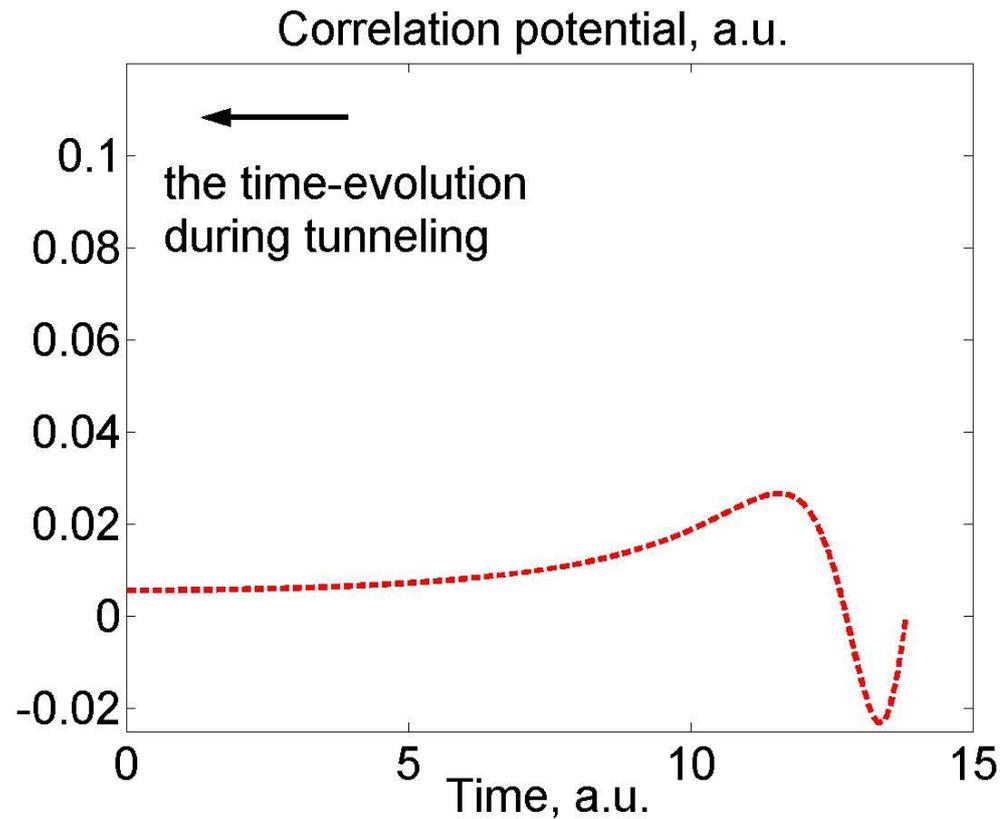
π phase between channels X and A
after ionization

PRL 2010 Mairesse et al

Why do we see what we see?

Correlation 'pulse' in N_2

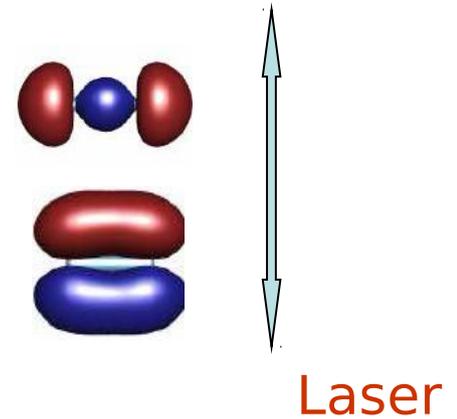
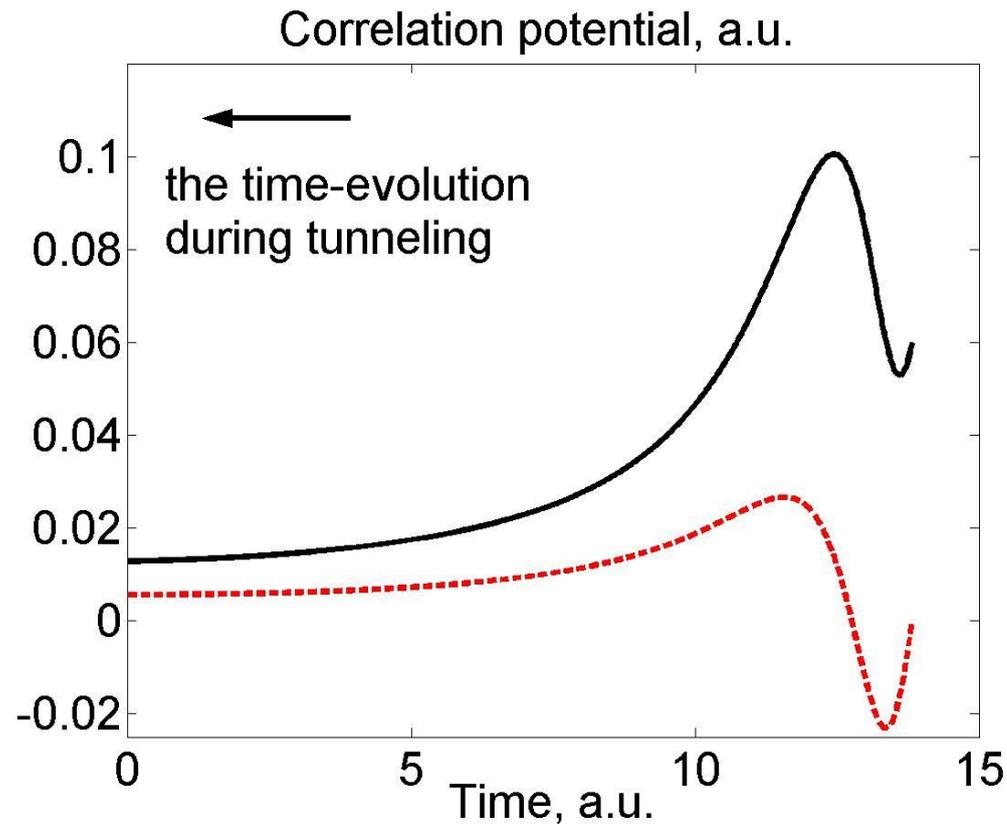
$$\theta=90^\circ$$



Correlation potential for the field-free HOMO and HOMO-1

Laser-dressed correlation pulse N_2

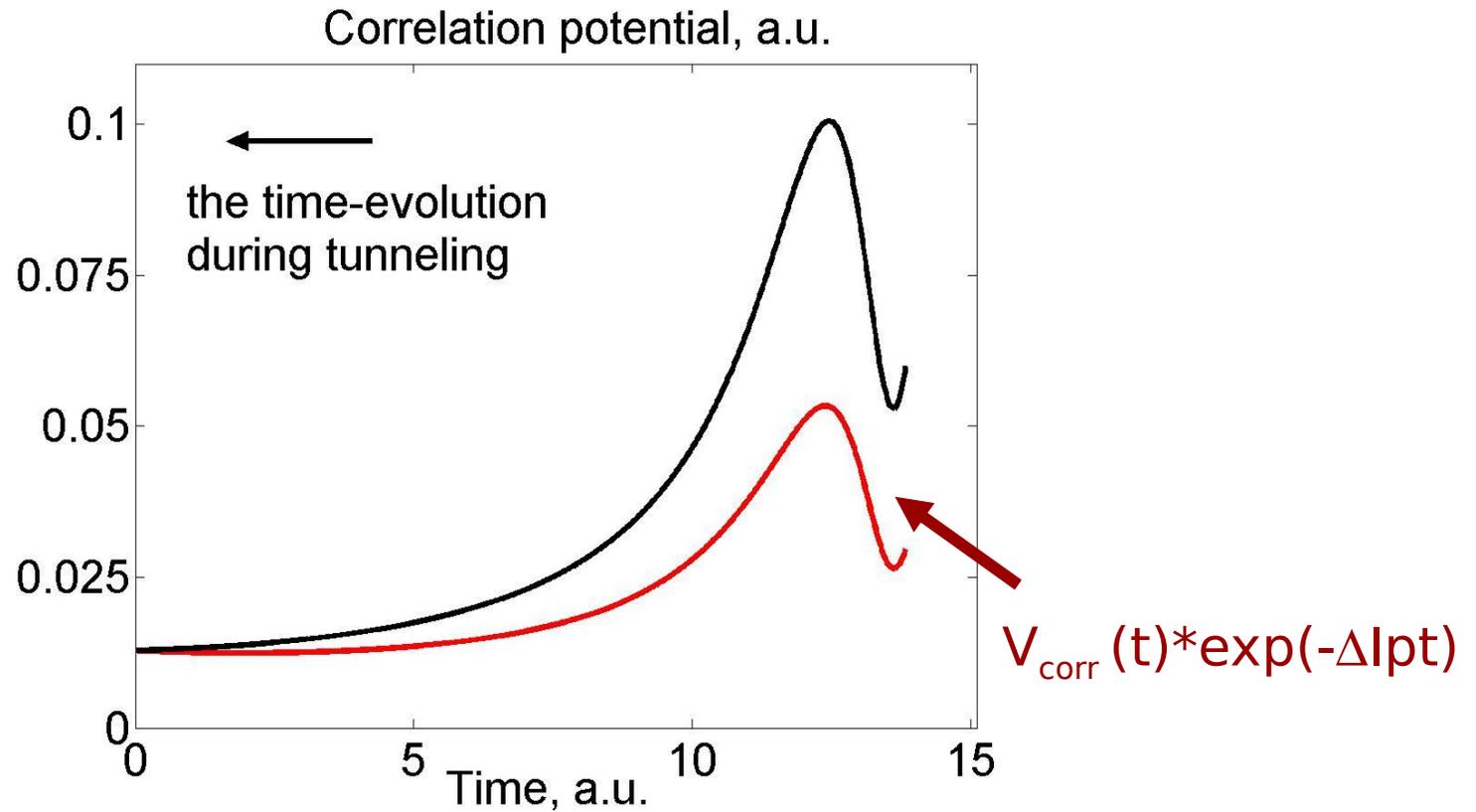
$$\theta=90^\circ$$



Correlation potential for laser dressed HOMO and HOMO-1

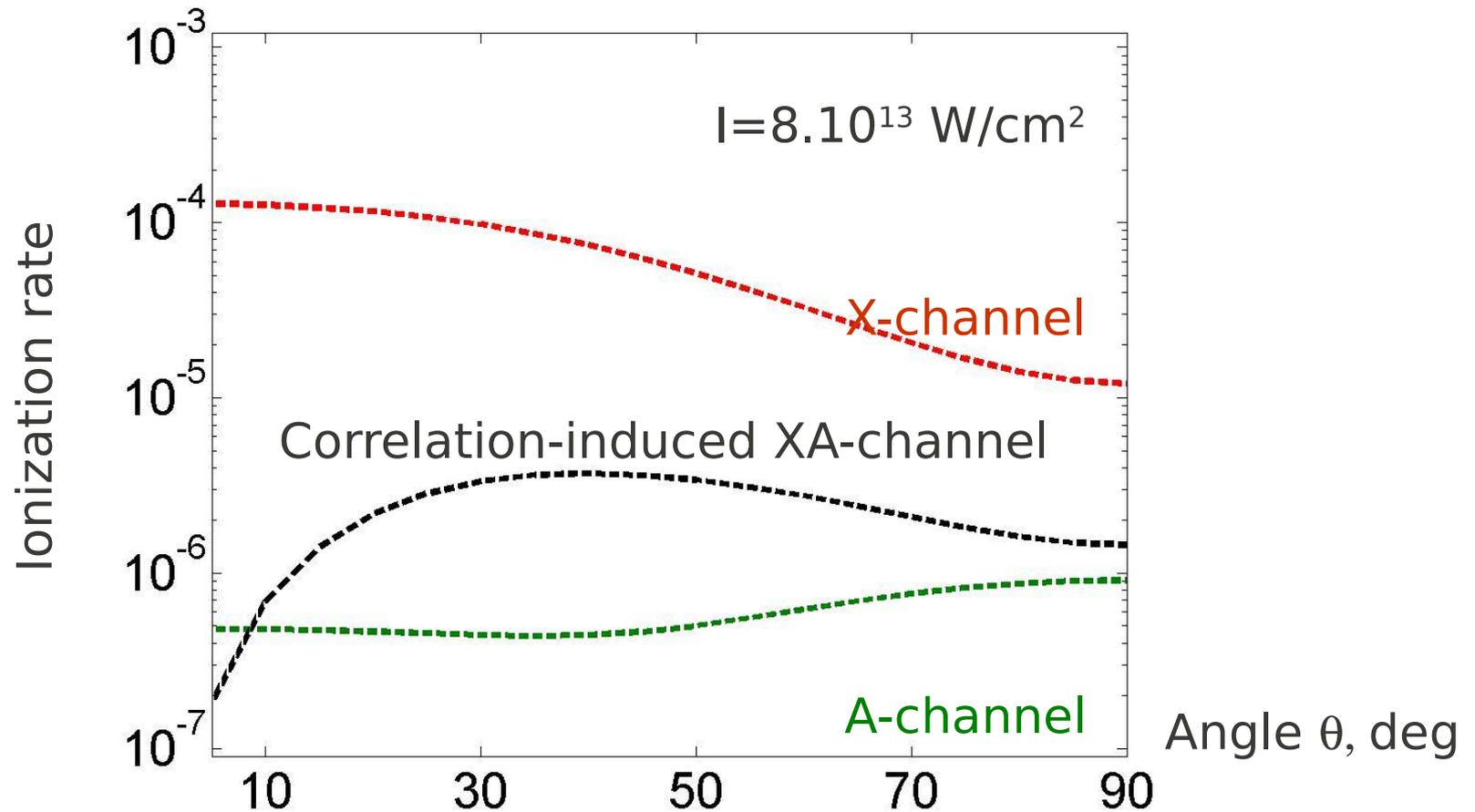
Effect of exponential suppression, N₂

$$\theta=90^\circ$$



$$c_{mn} = \int_{\tau}^0 dt V_{\text{corr}}^{mn}(t) e^{-\Delta I_p t}$$

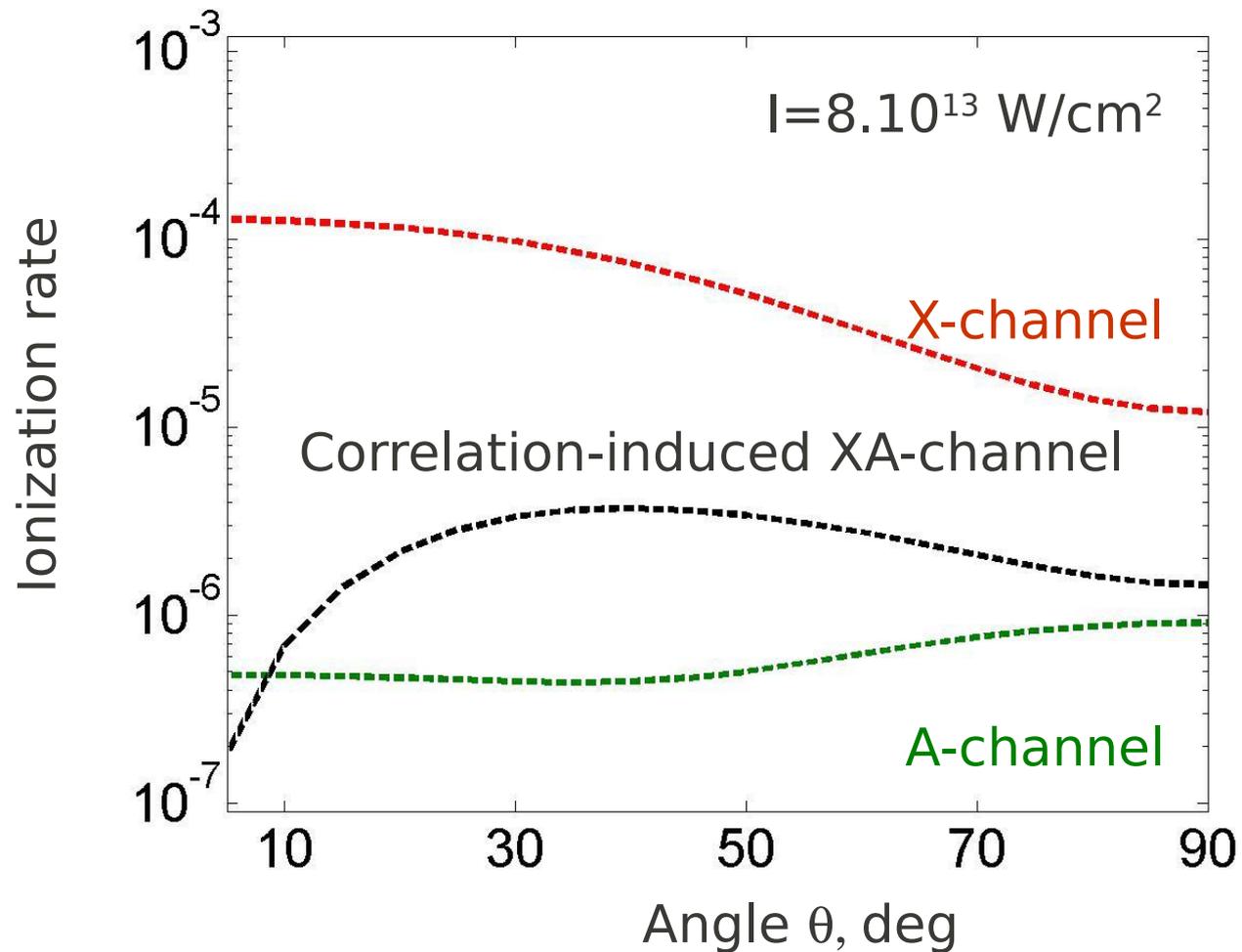
Correlation effects in N₂ molecule



Correlation -induced pathway can dominate ionization to channel A in N₂

Its phase is π relative to direct!!

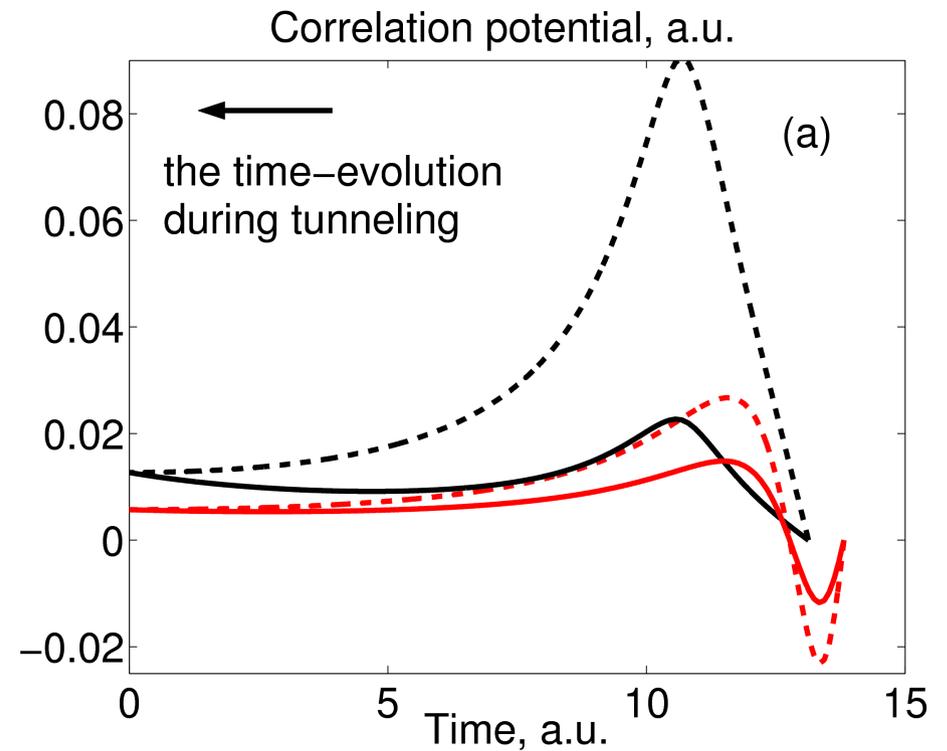
Correlation effects in N2 molecule



Dominance of the XA channel in ionization to A is the origin of π phase difference observed in Mairesse et al, PRL 2010

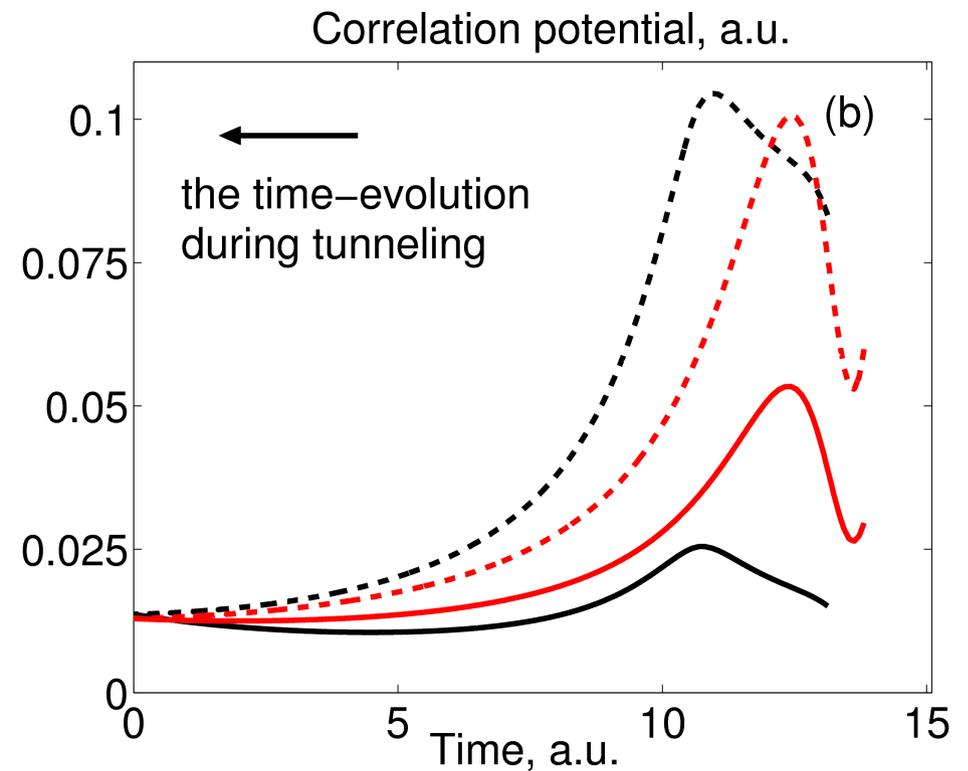
Field Free correlation potential vs time

- CO₂ aligned at 90 degrees, N₂ at 0 degrees – only XA transitions
- Dashed lines=before exponential suppression, solid lines=after exponential suppression



Field Dressed Correlation Potential vs time

- Same plot as before, except calculated between quasistatic states
- After field dressing and exponential suppression, N2 XA channel is more significant than CO2 XA channel – before, it was opposite!



Effects of an orbital node

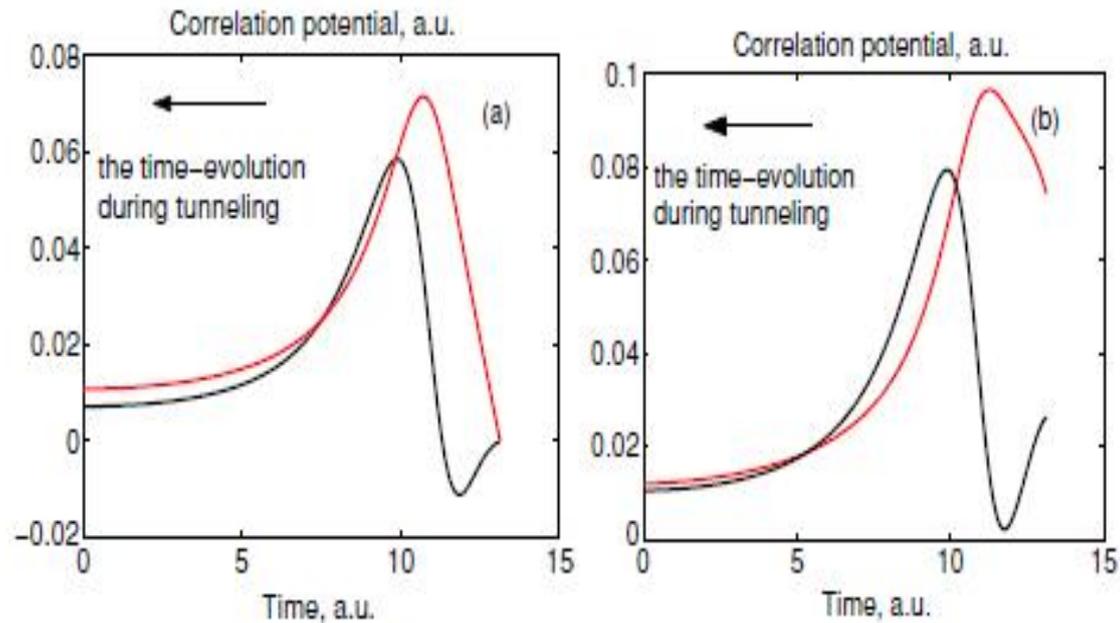
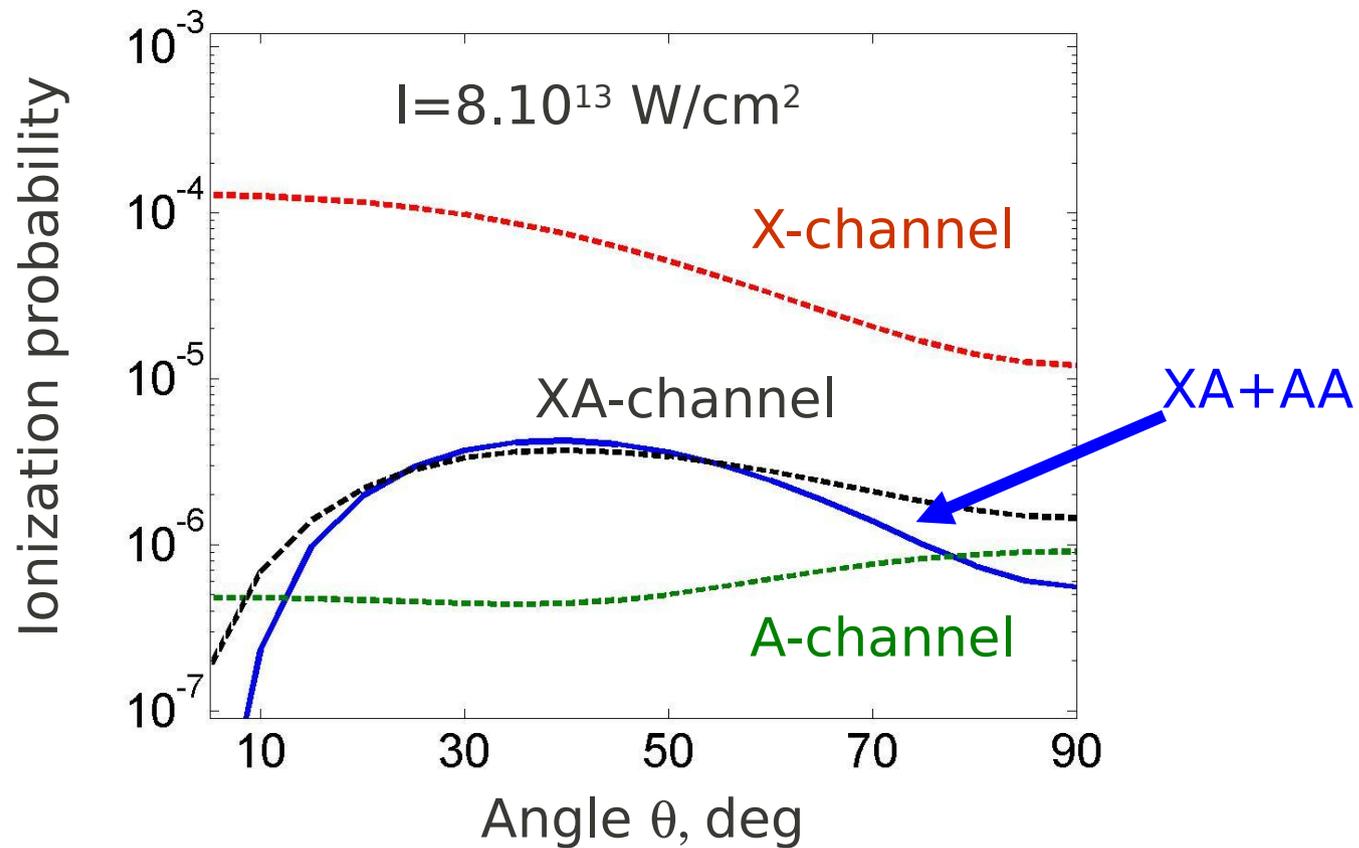


Figure 2. (a) Field-free $V_{AX}^{corr}(t)$ (red, grey in print), $V_{BX}^{corr}(t)$ (black) and (b) laser-dressed $V_{AX}^{corr}(t)$ (red, grey in print), $V_{BX}^{corr}(t)$ (black) correlation potentials versus (imaginary) time for CO_2 aligned at 30° . Ionization starts at $\tau = 13$ au.

Node in the B orbital causes a sign change in the field-free XB amplitude (left). This causes a minimum in the quasistatic XB amplitude (right).

Total ionization to channel A

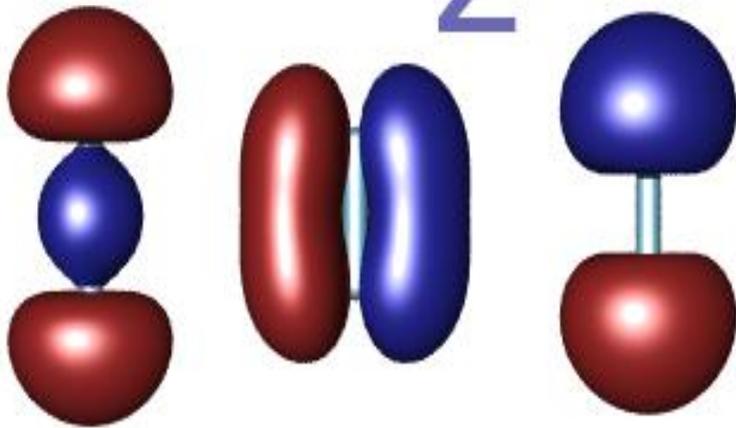
Direct and indirect ionization pathways add destructively



Correlation-induced channel has different angular dependence on molecular alignment than the direct channel!

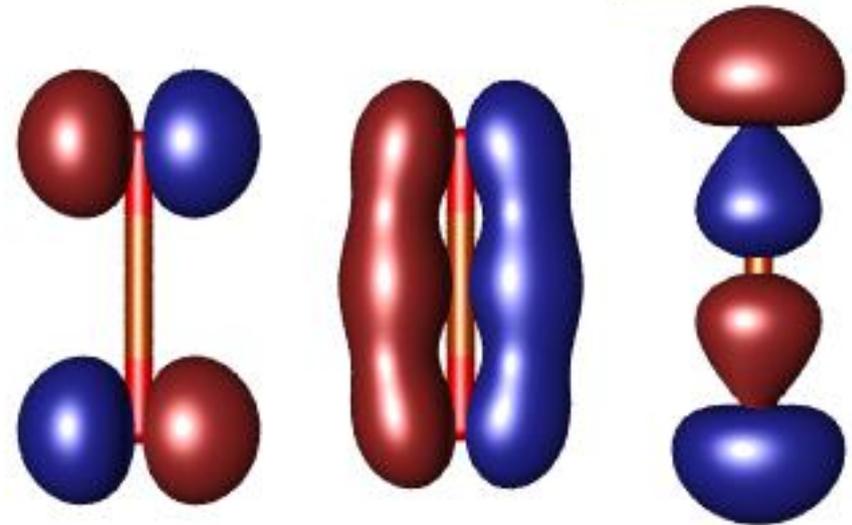
Molecules Considered

N_2



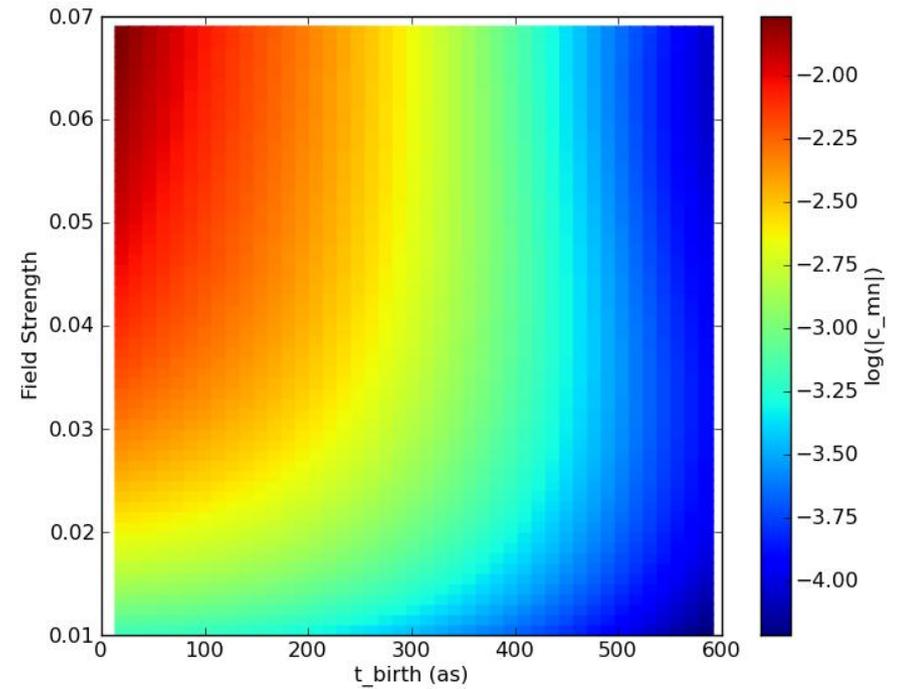
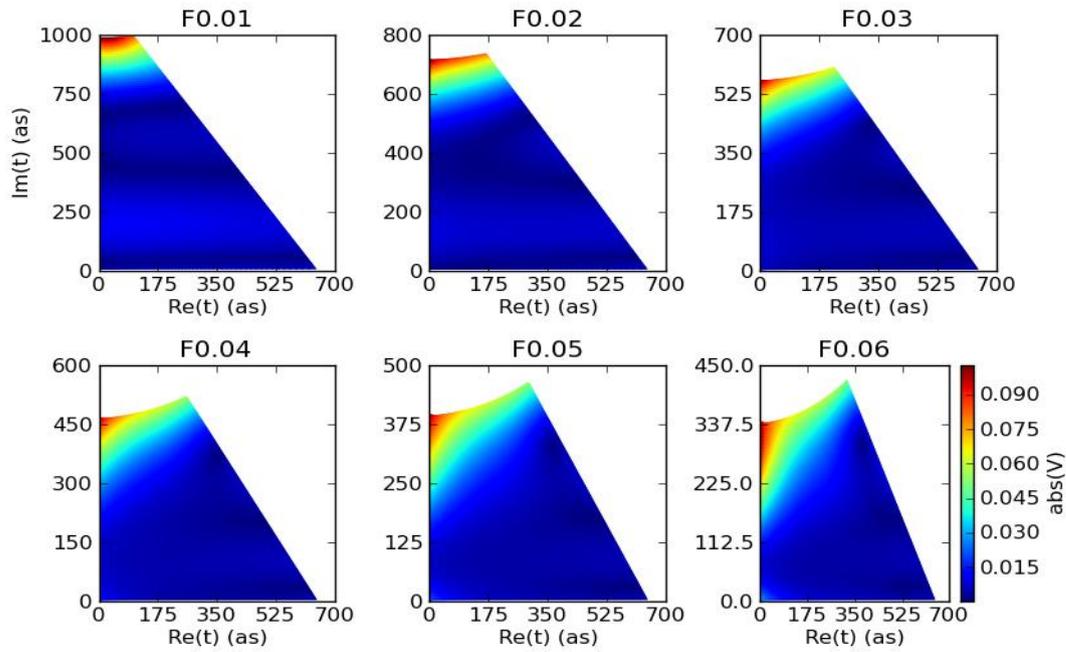
X A B

CO_2

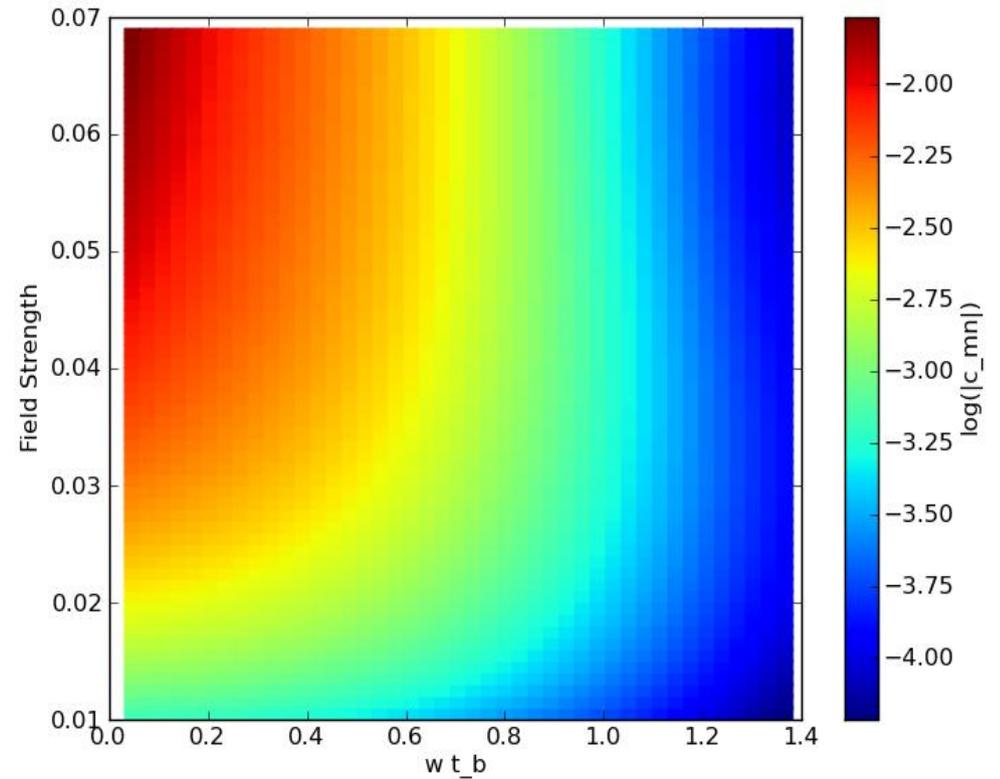
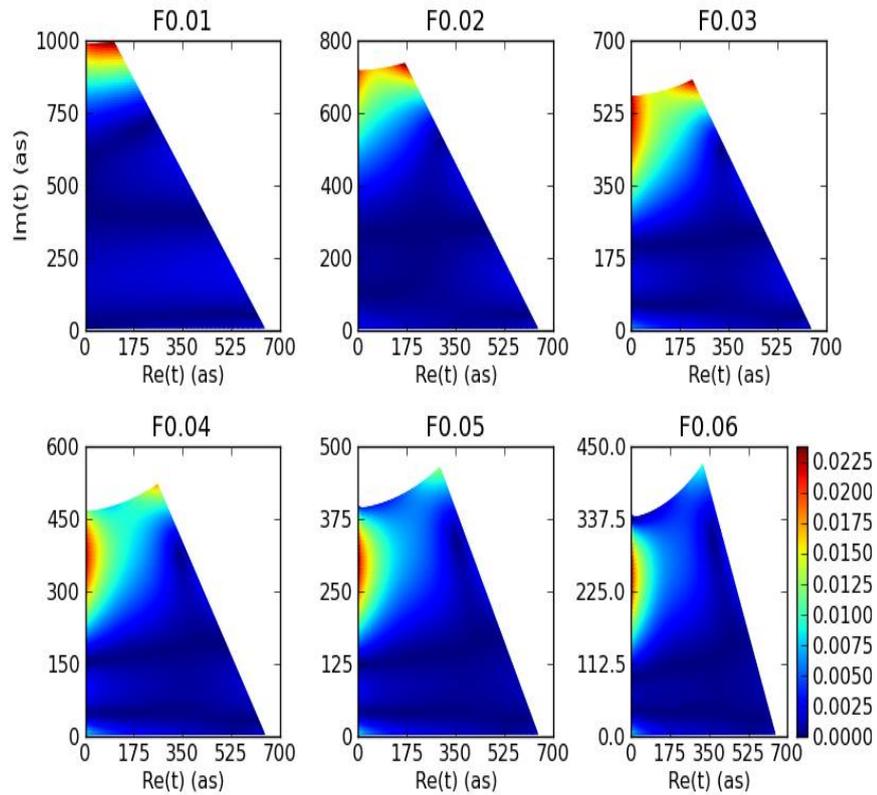


X A B

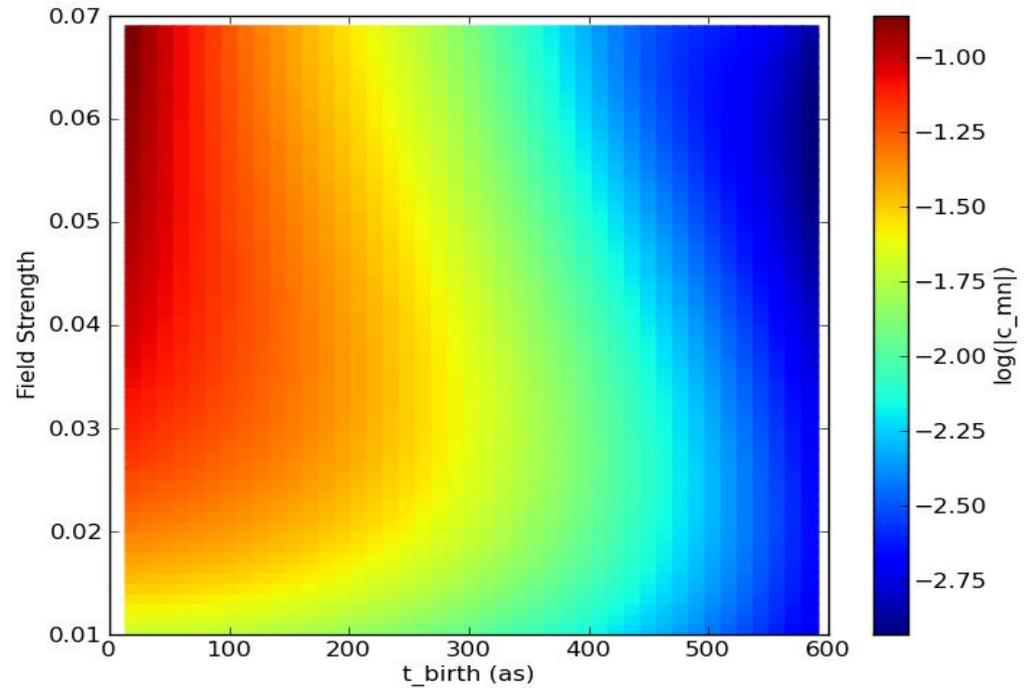
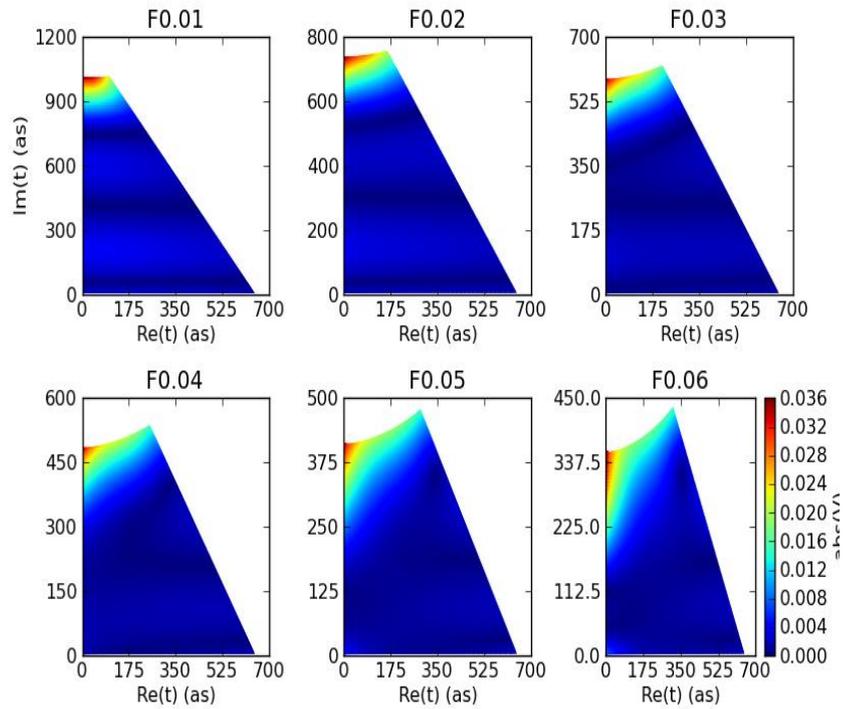
CO2, XA, 60 degrees



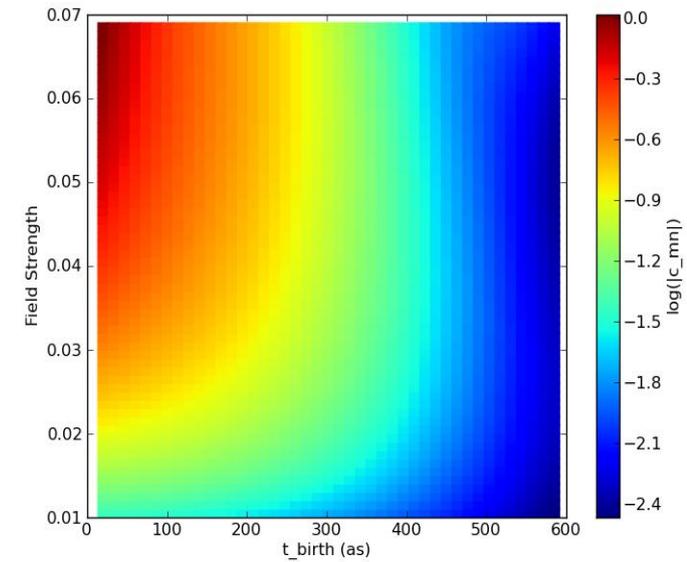
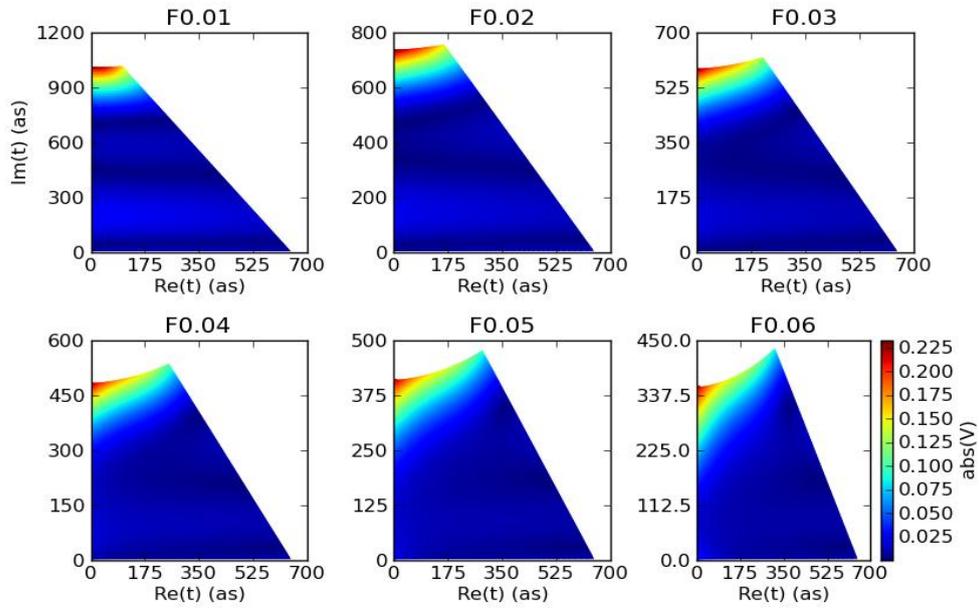
CO2, XB, 60 degrees



N2, XA, 30 degrees



N2, XB, 30 degrees



Conclusions

- Strong Field Ionization is a much more dynamic process than previously realized.
- The correlation potential pulse occurs in complex time, and depends on both the electronic structure and orientation of the molecule and the parameters of the driving laser.
- The indirect pathway is an alternate means for producing excited states of the ion, and is not subject to the full exponential suppression of the direct pathway.

Error vs. time step: Least Action vs. Short Iterative Lanczos

