Coherent control of internal conversion in strong field molecular ionization

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We demonstrate coherent control over internal conversion during strong field ionization with a shaped few cycle laser pulse. We make use of an ultrafast pulse shaper to produce few cycle phase locked pulse pairs with independent control over the phase and delay between pulses. The laser pulses drive resonance enhanced strong field ionization of a homologous series of halogenated methanes (CH_2IBr, CH_2BrCl and CH_2I_2) which is probed with velocity map imaging of the resulting photoelectrons. The photoelectrons label different states of the molecular cation, which are correlated with separate intermediately resonant states of the neutral molecule. Varying the relative phase between the two pulses controls the relative yields for different ionic states. This control can be related to interference in different neutral states of the molecule, which are coupled via non-Born Oppenheimer terms in the molecular Hamiltonian. Our measurements are interpreted with the help of electronic structure calculations, and demonstrate the preservation of molecular phase in non-adiabatic transitions between electronic states.