Extending Coherent Control to Complex Systems

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Since the advent of the laser 50 years ago an enduring dream has been to manipulate and control quantum-mechanical phenomena – and thus matter and energy on the atomic and molecular scale. Early attempts to selectively break bonds in polyatomic molecules, guided by intuition, used lasers to pump localized vibrational modes and were thwarted by rapid intramolecular energy redistribution. Subsequently, it was realized that exploitation of constructive or destructive interferences would be the key to quantum control. Demonstrations on simple atoms and molecules in the gas phase quickly ensued. Extensions to complex molecules used modern laser technology that allows sculpting of the phase and amplitude of laser fields in the infrared, optical and ultraviolet spectral regimes. Combining this technology with intelligent feedback has been essential for the control of complex systems – such as large molecules in the solution phase.

In schemes to control molecular dynamics in the solution phase, typically the feedback has utilized visible fluorescence or strong infrared pulses following the laser control pulse. Each of these feedback variables has its limitations. Here we propose that ultrafast x-ray pulses can provide a more incisive and direct probe of electronic dynamics and thus a more general feedback scheme. Single-shot, near-edge x-ray absorption, emission and inelastic scattering spectra on ultrafast timescales, appropriately synchronized to the initial excitation, could be a novel and versatile feedback mechanism for quantum control in complex systems.