Coherent x-ray-optical control of Mössbauer nuclei with zeptosecond timing stability

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Some 150 years ago, Bunsen and Kirchhoff pioneered spectroscopy by exciting chemicals with a flame. Despite its significance, from a modern perspective, their approach only involves uncontrolled excitation and passive observation. Since then, laser technology has led to a revolution in light-matter interactions, culminating in the possibility of controlling quantum dynamics using light. As a result, near-resonant light fields have become the primary tool to control electron dynamics. Such control requires a stabilization of the light's phase to fractions of the oscillation period of its field. For transitions in the visible range (E ~ 1 eV), this period is of order h/E ~ 4 fs, thus motivating attosecond science.

Modern x-ray facilities strive to continue this development even to hard x-rays, which is the realm of resonances in Mössbauer nuclei that feature transition energies in the ~10 keV range. However, current experiments with nuclei resemble the approach by Bunsen and Kirchhoff, with excitation using fluctuating x-ray pulses and passive observation. Coherent control of nuclear dynamics has remained impossible, because of the short oscillation period h/E ~ 400 zs, which poses extreme stability requirements on the few-zs level (1 zs = 10^{-21} s).

In this talk, I will show how to overcome these decisive factors. For this, we developed and implemented a method to shape x-ray pulses delivered by modern x-ray sources into tunable double pulses with the required few-zs stability. The double-pulse generation is based on our recent approach to shape the spectra of x-ray pulses using suitable piezo motions [1]. In our experiment, we exploited the relative phase of the two x-ray pulses to coherently control the dynamics of a nuclear target. Our results unlock coherent x-ray optical control for nuclei, and pave the way for a number of promising techniques and applications.