ESRF	**Experiment title:** Large gain by iterative redistribution of off-resonant spectral power into resonance	**Experiment number:** HC 2764
Beamline: ID 18	**Date of experiment:** from: 01 Dec 2016 to: 06 Dec 2016	**Date of report:** 01 Mar 2021
Shifts: 18	**Local contact(s):** Rudolf Rüffer	*Received at ESRF:*

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Report:
The experiment was performed as suggested in the proposal. The data measured in this beamtime formed the basis for two publications:

[1] K. P. Heeg, A. Kaldun, C. Strohm, C. Ott, R. Subramanian, D. Lentrodt, J. Haber, H.-C. Wille, S. Goerttler, R. Rüffer, C. H. Keitel, R. Röhlsberger, T. Pfeifer, and J. Evers, *Coherent X-ray-optical control of nuclear excitons*, Nature **590**, 401-404 (2021) (DOI:10.1038/s41586-021-03276-x)

Abstract: Coherent control of quantum dynamics is key to a multitude of fundamental studies and applications. In the visible or longer-wavelength domains, near-resonant light fields have become the primary tool with which to control electron dynamics. Recently, coherent control in the extreme-ultraviolet range was demonstrated, with a few-attosecond temporal resolution of the phase control. At hard-X-ray energies (above 5-10 kiloelectronvolts), Mössbauer nuclei feature narrow nuclear resonances due to their recoilless absorption and emission of light, and spectroscopy of these resonances is widely used to study the magnetic, structural and dynamical properties of matter. It has been shown that the power and scope of Mössbauer spectroscopy can be greatly improved using various control techniques. However, coherent control of atomic nuclei using suitably shaped near-resonant X-ray fields remains an open challenge. Here we demonstrate such control, and use the tunable phase between two X-ray pulses to switch the nuclear exciton dynamics between coherent enhanced excitation and coherent enhanced emission.

We present a method of shaping single pulses delivered by state-of-the-art X-ray facilities into tunable double pulses, and demonstrate a temporal stability of the phase control on the few-zeptosecond timescale. Our results unlock coherent optical control for nuclei, and pave the way for nuclear Ramsey spectroscopy and spin-echo-like techniques, which should not only advance nuclear quantum optics, but also help to realize X-ray clocks and frequency standards. In the long term, we envision time-resolved studies of nuclear out-of-equilibrium dynamics, which is a long-standing challenge in Mössbauer science.

[2] K. P. Heeg, A. Kaldun, C. Strohm, P. Reiser, C. Ott, R. Subramanian, D. Lentrodt, J. Haber, H.-C. Wille, S. Goerttler, R. Rüffer, C. H. Keitel, R. Röhlsberger, T. Pfeifer, J. Evers, *Spectral narrowing of x-ray pulses for precision spectroscopy with nuclear resonances*, Science **357**, 375-378 (2017) (DOI:10.1126/science.aan3512)

Abstract: Spectroscopy of nuclear resonances offers a wide range of applications due to the remarkable energy resolution afforded by their narrow linewidths. However, progress toward higher resolution is inhibited at modern x-ray sources because they deliver only a tiny fraction of the photons on resonance, with the remainder contributing to an off-resonant background. We devised an experimental setup that uses the fast mechanical motion of a resonant target to manipulate the spectrum of a given x-ray pulse and to redistribute off-resonant spectral intensity onto the resonance. As a consequence, the resonant pulse brilliance is increased while the off-resonant background is reduced. Because our method is compatible with existing and upcoming pulsed x-ray sources, we anticipate that this approach will find applications that require ultranarrow x-ray resonances.