Nonlinear nuclear optics: first experimental results and future challenges

Andreas Kaldun - Stanford University and SLAC National Laboratory, California, USA

Abstract: In the first nonlinear nuclear excitation experiment in August 2017, we saw evidence of extremely narrow-band (5neV) x-ray photons by two-photon excitation of the iron isotope $^{57}$Fe $I=1/2$- to $3/2$- nuclear transition at about 14.4 keV using the broadband (50eV) femtosecond FEL pulses (see Fig.1). As shown in the experimental results in Fig. 2, the second-harmonic photons are produced by delayed emission in an enriched $^{57}$Fe foil. The delayed emission with its 0.1 ps lifetime was measured directly in the time domain with nanosecond resolution. In my talk, I will present the measured parameter space for an enhancement of the nuclear excitation by a resonant electronic bridge process and the formerly unknown cross-section for the electronically enhanced excitation process that we determined to be $2 \times 10^{-57}$ cm$^4$ s [7]. Further, I discuss how our findings will enable high-flux & non-linear Mossbauer spectrosc. at high-rep. rate FELs.

Background: Mossbauer spectroscopy has found a broad array of applications in biology, chemistry, materials science, quantum optics and precisions measurement. That the high brightness of FELs could potentially be used to convert broad band FEL radiation into narrowband Mossbauer radiation through two-photon absorption was noted by Sebastian Doniach more than a decade ago [1]. Such a process requires the short pulse and high-flux of the FEL and is interesting both from a fundamental science and applications perspective. So far experiments using X-ray FELs have been studied to study only multiphoton electronic nonlinearities at hard x-ray wavelengths, including both two-photon absorption [2] and nonresonant second harmonic generation [3] and two-photon Compton scattering [4]. Expanding nonlinear optics to nuclear resonances has not only a general significance for x-ray spectroscopy, our implementation also substantially advances spectral engineering and control at x-ray energies [5] and is a first step towards exploring nuclear lambda systems and resonant nuclear Raman spectroscopy.