Collinear phase-modulated femtosecond pump-probe experiments using a low repetition-rate laser system

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Synopsis A 5 kHz repetition-rate laser system is used to perform phase-modulated pump-probe spectroscopy on rubidium vapor. We confirmed the applicability of high-frequency lock-in modulation with such low repetition-rate lasers and show that much higher modulation frequencies than laser repetition-rates can be used without losing performance. This undersampling technique can be advantageous for any other lock-in measurements with low sampling rates by shifting the lock-in demodulation to a higher frequency for more efficient noise filtering.

The helium nanodroplet isolation (HENDI) technique is a well-established powerful method to perform spectroscopic studies at very low temperatures. Due to the low target densities in doped droplet beam experiments, coherent time-resolved spectroscopy of such systems has remained a challenging task. In this context, we are investigating the phase-modulation technique established by Marcus et al. [1]. The combination of continuous acousto-optical phase-modulation with lock-in detection greatly improves the signal-to-noise ratio (SNR) and the sensitivity in this scheme [2]. However, the method was thought to be suitable only for high repetition-rate laser systems (> 200 kHz) to be able to sufficiently sample the imparted acousto-optic modulation.

Recently, we have successfully implemented this technique in a pump-probe scheme with femtosecond laser pulses at 5 kHz repetition-rate. As a simple model system, we investigated the $5^2S_{1/2} \rightarrow 5^2P_{1/2}$ transition in a rubidium vapor. We found, what seems at first very counterintuitive, that much higher modulation frequencies than laser repetition-rates can be used without losing performance (see Fig. 1). This effect, which we call phase-synchronous undersampling, shows promise for the implementation of the phase-modulation scheme in even lower repetition-rate XUV laser sources by shifting the carrier frequency far away from the low frequency noise spectrum. Besides this application, the phase-synchronous undersampling scheme can be transferred to any other lock-in measurement where it is desirable to demodulate at higher frequencies than the sampling rate in the experiment.

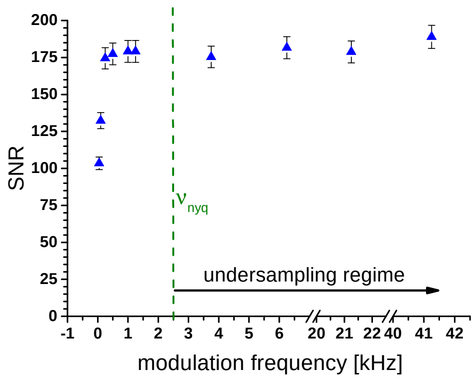

Figure 1. Dependence of the SNR in the pump-probe measurements with the 5 kHz repetition-rate laser on the modulation frequency. The dashed line represents the Nyquist frequency (ν_{nyq}), which is given by half the laser repetition-rate. For modulation frequency higher than ν_{nyq}, the acousto-optic modulation is sampled by less than two laser shots per period. We call this the undersampling regime. Surprisingly, the SNR does not show a decrease when reaching this regime.

References

[1] A. H. Marcus *et al.* 2006 *J. Chem. Phys.* **125** 194303

[2] L. Bruder *et al.* 2015 *Phys. Chem. Chem. Phys.* **17** 23877

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