Chapter 7

Summary and outlook

In this thesis, we have joined together the study of cold atoms, which are well established as an ideal sample for spectroscopy with the field of ultrafast lasers, which have recently emerged as a powerful tool for frequency metrology.

We have used a wide-bandwidth, phase-stabilized femtosecond laser as a novel and efficient tool to perform Direct Frequency Comb Spectroscopy (DFCS) of one-photon and two-photon transitions in laser-cooled ^{87}Rb atoms. To begin with, we have studied the dominant sources of systematic shifts and tried to reduce their effects as much as possible. We have then measured the absolute atomic frequency for several $5S_{1/2} \rightarrow 5D_{3/2,5/2}$ two-photon transitions in ^{87}Rb. The two-photon spectroscopy is carried out efficiently despite the low laser power, because we take advantage of intermediate 5P states that are near-resonant with a comb line. We have demonstrated that the absolute optical frequency of the previously unmeasured $5S_{1/2} \rightarrow 7S_{1/2}$ resonances can be determined unambiguously from the comb structure used in the experiment. Lastly, we have shown that DFCS can be successfully applied to one-photon processes as well, by measuring $5S_{1/2} \rightarrow 5P_{1/2,3/2}$ transitions both directly and indirectly, through their resonant enhancement of the 5S-5D two-photon transitions. Detailed dynamics of population transfer driven by a sequence of pulses were uncovered and taken into account for the indirect measurement of the 5P states. Although the current experiments involve one- and two-photon transitions, the advantages of DFCS should also apply to multiphoton excitations. In general, DFCS permits global, high-resolution spectroscopy of all the atomic transitions within the comb bandwidth.

After we successfully implemented DFCS for the study of several one- and two-photon transitions, there is still room left for improvements. Firstly, the resolution of DFCS can be enhanced by locking the femtosecond laser to a cavity, which has been shown capable to reduce the linewidth to below 100 Hz [85], rather than the current 300 kHz. Secondly, a larger signal can be obtained by using a laser with a higher repetition rate; for example, a 1-GHz laser with the same average power and spectral width could increase the signal up to a 100-fold. This is a result of the quadratic increase of the excited state population versus the accumulated number of pulses, where the higher f_r and hence the shorter pulse interval allow for more pulses to be 'added' during the atomic coherence lifetime. Additionally, the power is distributed among fewer comb lines for this $f_r \sim 1$ GHz case, and the decreased congestion of lines would reduce optical pumping effects. Achieving simultaneous one- and two-photon resonance conditions should still be possible. Using a 1-GHz laser provides more latitude in identifying the mode number of the comb component that is resonant with each optical transition. The larger mode spacing makes this identification easier, because it only requires knowledge of the optical frequencies with a precision of a few hundred MHz. One practical consequence of these results is a method to control both degrees of freedom of the femtosecond comb directly by an optical transition in cold atoms.

Another interesting application of the demonstrated pulse accumulation effect is using ultrafast pulse trains for laser cooling of atoms (e.g. hydrogen, deuterium, antihydrogen) that require coherent ultraviolet light not easily accessible by conventional laser sources [86]. Toward this goal, an extension of frequency comb metrology to the deep-ultraviolet spectral region (where cw lasers are not readily available) was recently achieved using an amplified train of phase-controlled pulses from a femtosecond laser [78]. The peak power of these pulses allowed for efficient harmonic upconversion and thus, the 4th harmonic of the femtosecond laser was used for two-photon spectroscopy in krypton. Even more recently, phase-coherent frequency combs in the vacuum-ultraviolet spectral region were demonstrated in our lab, without active amplification of the standard femtosecond laser [87]. Its output was stabilized to a completely passive optical cavity, with a gas jet at the intracavity focus, generating a phase-controlled frequency comb in the extreme ultraviolet region which preserves the original repetition frequency. This technique, in combination with DFCS, opens the door to more accurate atomic optical clocks that operate on resonances with ultrahigh, vacuum-ultraviolet or extreme-ultraviolet, frequencies. These clocks would have a very high stability, proportional to their transition frequency.

For general coherent control experiments, pulse accumulation (when enabled by long coherence times) can complement spectral amplitude and phase manipulations, leading to improved efficiency in population control with the added spectral resolution and longer experimental timescales due to maintaining phase coherence for many consecutive laser pulses. The precise and phase-coherent pulse accumulation may prove particularly useful in efficiently populating atomic Rydberg states for quantum information processing. Multiple ultrafast lasers with optical spectra independently tailored for different spectroscopic features could be phase coherently stitched together [9,88] to further extend the utility of this approach.