Ramsey-comb spectroscopy
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2014

document version
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Download date: 10. Mar. 2024
The most accurate, absolute measurements that we can perform are time/frequency measurements simply because the physical unit of the second is the most accurately defined SI-unit. Since 1967, the second is defined as the duration of 9,192,631,770 periods of a hyperfine transition frequency in $^{133}$Cs and commercial products are available for distributing this frequency standard at the $10^{-13}$-level in many metrology laboratories around the world. With the help of optical frequency combs (FC) based on mode-locked lasers, this remarkable accuracy can be transferred all the way from the microwave to the optical domain, thus enabling high-precision spectroscopy on atomic and molecular systems. Beyond their use as calibration tools for referencing continuous-wave lasers, FCs can also be employed themselves as the spectroscopy lasers. This very successful technique of direct FC spectroscopy combines high accuracy with high laser intensities and broad wavelength coverage from the pulsed laser oscillators. However, there are a range of applications that require even higher intensities, e.g. for the excitation of very weak transitions or in order to convert the wavelength of the FC to spectral regions inaccessible by laser oscillators.

In order to reach ultrahigh pulse intensities and energies, our group has followed the unique approach of coherently amplifying only selected (two) pulses derived from a regular FC pulse train. Because the average power can be kept low, this approach allows for a significantly higher amplification level than alternative methods based on, e.g., full repetition rate amplification in conjunction with enhancement cavities. While leading to the first high-precision measurement in the extreme ultraviolet wavelength region $<$60 nm, the initial approach nevertheless sacrificed part of the beauty of the underlying FC as only single, isolated transitions could be measured and the accuracy was limited by the maximum delay and phase shifts during the amplification process. To overcome this limitations a novel spectroscopic method has been developed, “Ramsey-comb spectroscopy”, which is introduced and demonstrated in the course of this thesis. Although the Ramsey-comb method still relies on amplified FC pulse pairs, the new ability to coherently change the inter-pulse delay over a wide range enables a fundamentally new way of measuring and
analyzing the spectroscopic signals. We show that the combination of high-energy laser pulses with the accuracy and resolution of FCs provides an extremely versatile tool for high-precision spectroscopy. A more detailed introduction and motivation for producing high peak power FC pulses is given in Ch. 1 along with a short outline of the thesis, whose content is summarized in the following paragraphs.

In order to efficiently discuss the technical and conceptual challenges in the course of this thesis, first some particularly relevant background is reviewed in Ch. 2. Starting from Maxwell’s fundamental equations, the concept of temporal and spectral envelopes for laser pulses is derived and applied to study physical effects such as propagation and dispersion. Going from single pulses to pulse sequences, one of the key elements of the Ramsey-comb method is introduced, the optical FC, including a discussion of how FCs are implemented in practice. Finally, the theory of two different approaches for amplifying laser pulses is introduced: the concept of optical parametric amplification, which enables the phase-coherent amplification of FC pulses, and the more conventional amplification in laser gain media, which is employed to create the high-energy pump pulses for the parametric amplification.

Realizing Ramsey-comb spectroscopy requires a complex experimental laser system for producing high-energy, multi-delay coherent laser pulse pairs. The description of the developed system starts in Ch. 3, which contains a detailed discussion of the pump amplifier system for producing the high-energy pump pulse pairs for the parametric amplifier. First, the home-built, passively mode-locked pump oscillator is described, whose output is adjusted in terms of pulse duration and center wavelength. From the full pulse train, two pulses are selected via programmable pulse pickers at variable delays. Subsequently, these pulses are amplified in a two-stage, ultrahigh-gain pre-amplifier, boosting the pulse energy from less than 100 pJ to the 1 mJ-level. The pre-amplifier is followed by either a flashlamp-pumped or a diode-pumped post-amplifier. Altogether, the developed laser amplifier system is capable of producing 1064 nm pump pulse pairs at the 100 mJ-level with inter-pulse delays of multiples of the cavity round-trip time (~8 ns) of the master pump oscillator well into the microsecond range. Furthermore, the inter-pulse delay can quickly be changed while keeping the optical paths in the amplifier the same, thus minimizing wavefront deviations for the second pump pulse as a function of delay time.

The amplified pump pulse pairs from the system described in Ch. 3 are used to pump an optical parametric amplifier, which in turn selec-
tively amplifies pulse pairs from a synchronized FC oscillator operating at the same repetition rate as the pump oscillator. The performance of the parametric amplifier system, in particular the phase stability of the mJ-level amplified FC pulses, is the main subject of Ch. 4. First, the home-built Ti:sapphire FC is described, which acts as the seed for the parametric amplifier. A technical overview of the parametric amplifier system and the electronic synchronization scheme is then followed by an extensive study of the phase shift induced by the amplifier, both via numerical simulations and direct measurements. It is established that although the amplified FC pulses can exhibit a differential phase shift of a few 100 mrad, this phase shift remains effectively constant when changing the inter-pulse delay. This important outcome makes the experimental system suitable for high-accuracy Ramsey-comb spectroscopy.

Having concluded the detailed description of the developed laser system, Ch. 5 shows the capabilities of Ramsey-comb spectroscopy via a measurement of complex two-photon transitions in atomic rubidium and cesium. Owing to the good signal-to-noise ratio and a counter-propagating excitation scheme to reduce Doppler broadening, Ramsey signals are obtained at more than 40 different macro delays (in steps of 8 ns), which in the case of rubidium is more than four times the lifetime of the upper state. With the help of a novel time-domain fitting algorithm, the excited transition frequencies are determined with an accuracy better than 10 kHz including systematic uncertainties. For the weak transitions in cesium, this accuracy level represents a more than thirty-fold improvement to previous spectroscopic results. This shows that the Ramsey-comb method can significantly outperform traditional forms of spectroscopy on transitions that are too weak to be easily excited with conventional unamplified FCs or continuous-wave lasers.

After its first introduction in the previous chapter, a more comprehensive analytical framework of the Ramsey-comb method is presented in Ch. 6. Starting from Ramsey’s originally derived equation describing excitation with separated oscillating fields, Ramsey signals from different macro delays are combined on a global time axis. In the frequency domain, this combination of individual measurements can be used to derive a “synthetic” excitation spectrum that resembles spectra obtained from traditional direct FC spectroscopy. However, it is shown that as opposed to traditional FC spectra, the spectral analysis is affected by additional interferences that severely complicate the frequency determination if more than one resonance is excited. Fortunately, the time-
domain analysis is found to be much more robust as it relies solely on the
temporal phases of the individual Ramsey scans. This has the additional
advantage of being insensitive towards common spectral line-broadening
effects and (transition-independent) constant phase shifts, due to, e.g.,
the amplification process or the AC-Stark effect from the excitation pul-
ses themselves. Finally, the performance of the presented time-domain
fitting approach is investigated via numerical simulations for different pa-
rameter sets including cases of transition-dependent broadening mecha-
nisms and phase shifts. Within the simulation uncertainties of a few kHz,
no sizeable systematic effects on the fitting results are encountered.

The thesis is concluded by an outlook in Ch. 7, which describes
possible upgrades of the experimental system as well as interesting future
targets for Ramsey-comb spectroscopy such as H$_2$ and He$^+$. 