Femtosecond enhancement cavity – direct frequency comb spectroscopy and coherent extreme nonlinear optics

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Abstract. To prepare for future experiments on coherent spectroscopy and quantum control in the VUV spectral regions, we have pursued direct frequency comb spectroscopy and generation of phase coherent VUV frequency combs. High-finesse, low-dispersion passive optical cavities are used to enhance the power of femtosecond pulses for extreme nonlinear optics and to increase detection sensitivity for spectroscopy.

1. Introduction

Recent progress in coherent optical spectroscopy has led to the recovery of a record-high quality factor (Q > 2.4 x 10^{14}) for a doubly-“forbidden” natural resonance observed in a large ensemble of trapped ultracold Sr atoms [1]. This unprecedented spectral resolving power impacts fields ranging from precision frequency metrology to quantum optics and quantum information science. Ultrastable lasers, together with optical frequency combs, can now maintain optical phase coherence beyond 1 s and transfer this stability across hundreds of terahertz [2]. As it becomes increasingly challenging to maintain phase coherence beyond multiples of seconds, it is natural that we look beyond the visible domain and consider speeding up the “wheel of precision measurement” to the next level of carrier frequency. We have thus pursued two related experimental directions to address this vision. One is the generation of phase coherent frequency combs in the VUV (50 - 200 nm) spectral domain [3]. In parallel, we have also pursued direct frequency comb spectroscopy [4] to ready ourselves for quantum optics and precision spectroscopy once phase coherent sources become available in VUV.

Both experiments benefit from the use of femtosecond enhancement cavities [5]. These are passive optical cavities with high finesse and low dispersion over a large spectral bandwidth such that incident femtosecond pulse trains can be efficiently coupled inside. Pulse energies can be enhanced by three orders of magnitude to >10 μJ while the original pulse repetition frequency is maintained. This capability permits phase coherent high harmonic generation process to take place at enhanced average efficiency. In addition to the power enhancement aspect, femtosecond cavities effectively increase the interaction length between matter and light, allowing direct frequency comb spectroscopy to acquire linear or nonlinear atomic and molecular signals with dramatically increased sensitivity [6].

2. Interactions between cavity and femtosecond pulse train

We have recently accomplished several important studies on the interaction between a femtosecond-laser based optical frequency comb and a high-finesse, low-dispersion, passive optical cavity. We have achieved direct stabilization of a frequency comb to a high-finesse optical cavity [7]. The resulting frequency/phase stability between the frequency comb and the cavity modes demonstrates a fully coherent process of intracavity pulse buildup and storage. We have also developed a femtosecond comb-based measurement protocol to precisely characterize mirror loss and dispersion [8]. This technical capability has facilitated production of large bandwidth, low-loss and low-dispersion mirrors. In addition, we have studied the nonlinear response of intracavity optical elements, demonstrating their limitation on power scalability [9]. This study has led to the design of novel cavity geometries to overcome this limitation [10]. In summary, we have achieved nearly three-orders of power enhancement inside a femtosecond buildup cavity, resulting in an intracavity pulse train that (1) is completely phase coherent to the original comb from the oscillator, (2) has the original laser's repetition rate (~100 MHz), (3) has a pulse peak energy exceeding 5 μJ, (average power > 500 W), intracavity peak intensity > 10^{13} W/cm^2, and (4) is under 60 fs pulse duration. We also note that this enhancement cavity approach is compatible with a number of femtosecond laser systems, including mode-locked Ti:Sapphire and fiber lasers.

3. Frequency comb in VUV

High-harmonic generation provides a coherent source of vacuum-ultraviolet (VUV) to soft x-ray radiation in a relatively compact system. Using the femtosecond enhancement cavity approach we aim to improve several important aspects of the HHG process, namely average power, system size and cost, and spectral resolution. We have made initial demonstrations that coherent frequency combs in the VUV spectral region are produced from the generation of high-harmonics of the laser without any active amplification or decimation of the repetition frequency [3]. This is accomplished by placing a xenon gas jet at the intracavity focus of a femtosecond enhancement cavity where the peak intensity exceeds 3 x 10^{13} W/cm^2. Since little of the fundamental pulse energy is converted, a femtosecond enhancement cavity is ideally suited for HHG as the driving pulse is continually "recycled" after each pass through the gas target. HHG at high repetition rates improves average power conversion efficiency. Optical-heterodyne-based measurements reveal that the coherent frequency comb structure of the original laser is preserved in the HHG process. These results lead the way for precision frequency metrology at extreme wavelengths and permit efficient HHG using only a standard laser oscillator.

To couple the HHG light out of the cavity, a thin sapphire plate is placed at Brewster's angle (for the IR) inside the cavity. However, the nonlinear response of the intracavity Brewster plate has so far limited the power scalability of the system. To solve this problem, we have designed novel enhancement cavity configurations that will allow us to use more powerful lasers [10]. One of the focusing cavity mirrors has a 200-μm hole drilled in the middle. By using a higher order cavity mode such as TEM01, we are still able to build up sufficient peak power inside the cavity for HHG to work. The generated VUV comb, however, leaks out of the mirror hole due to the significantly smaller diffraction angles enjoyed by the shorter wavelength light beam. This cavity geometry will allow a larger buildup power inside the cavity without any intracavity optics.

4. Highly sensitive, massively parallel, broad-bandwidth, real-time spectroscopy

We have developed cavity-enhanced direct frequency comb spectroscopy utilizing a broad bandwidth optical frequency comb coherently coupled to a high finesse optical cavity inside which atomic or molecular samples are located [6]. Hundreds of thousands of optical comb components, each coupled into a specific cavity mode, collectively provide sensitive intracavity absorption information simultaneously across 100 nm bandwidth in the visible and near IR spectral region. By placing various atomic and molecular species inside the cavity, we have demonstrated real-time, quantitative measurements of the trace presence, transition strengths and linewidths, and population redistributions due to collisions and temperature changes. This novel capability to sensitively and quantitatively monitor multi-species molecular spectra over a large optical bandwidth in real-time provides a new spectroscopic paradigm for studying molecular vibrational dynamics, chemical reactions, and trace analysis. We will continue to develop state-of-the-art laser sources in the infrared spectral regions, possibly even covering the important 3 micron area, to further improve the system sensitivity.

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