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High-resolution spectral fingerprinting with a stabilized femtosecond laser frequency comb

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Abstract: We demonstrate a novel optical spectroscopy technique combining a stabilized femtosecond frequency comb with a spectrometer that resolves individual comb elements. Rapid parallel data acquisition over many terahertz with hertz-level resolution can be achieved.
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The frequency stabilization of the broadband (octave-spanning) frequency comb emitted from a mode-locked femtosecond laser has permitted a wide range of scientific and technological breakthroughs—ranging from the counting of optical cycles for next-generation atomic clocks[1] to measurements of phase-sensitive high-field processes[2]. While the concept of using the frequency comb from a mode-locked laser for high resolution spectroscopy is not new[3-5], developments in broad spectral generation and subsequent stabilization of the carrier-envelope offset frequency provide powerful new tools that have led to recent advances in high-resolution (~10^{-11}) spectroscopy performed directly with the output of the mode-locked laser[6, 7]. In this paper, we combine the advantages of a stabilized Ti:sapphire frequency comb with a virtually-imaged phased array (VIPA) spectral disperser[8] that provides sufficient angular dispersion to spatially resolve the individual modes of the frequency comb[9]. With the introduction of an iodine vapor cell in the beam path, specific modes of the frequency comb are attenuated. This is readily observed in the 2-dimensional image of the frequency comb, thus providing an intuitive and unique spectral fingerprint of numerous absorption lines in a molecular gas. Data covering several terahertz (presently limited by imaging constraints) with ultimate resolution at the 1 Hz level are acquired in millisecond averaging times.

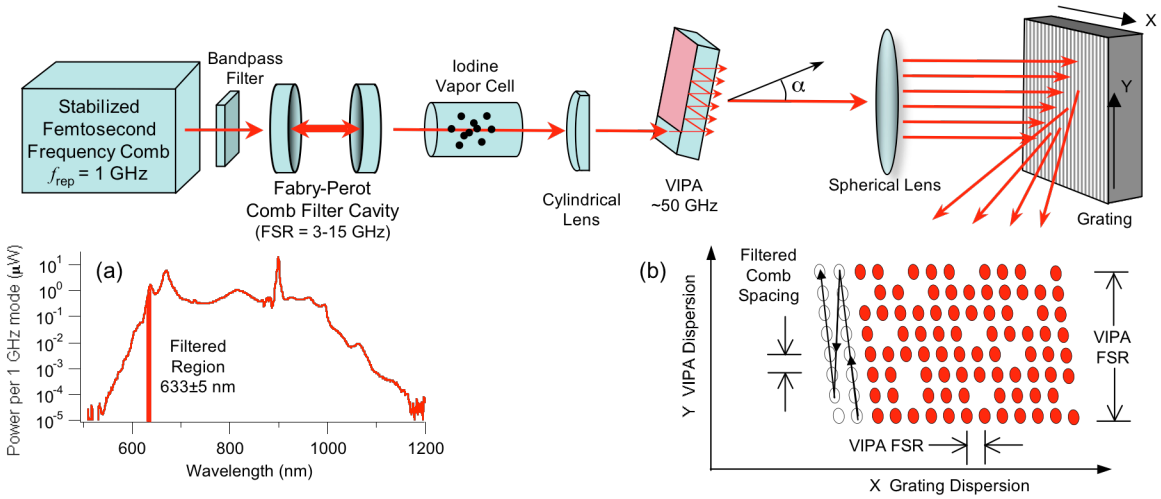

Fig. 1. Experimental setup. A high resolution virtually-imaged phased array (VIPA) disperser is used in combination with a lower resolution diffraction grating to spatially resolve the filtered frequency comb of a frequency-stabilized, broadband Ti:sapphire femtosecond laser. The output spectrum of the laser and the 633 nm region of interest are shown in (a). A vapor cell containing molecular iodine can be moved out of, or into, the beam path to acquire background data or spectroscopic data, respectively. The spectrometer output, as measured on a CCD element, consists of a 2-d array of the frequency comb modes, where each "dot" represents an individual mode (b). Within a column (Y), which is tilted by the grating dispersion, the dots are separated by the cavity-filtered mode spacing (3 GHz in this case). Within each row (X), the dots are separated by the VIPA free spectral range (FSR ~50 GHz in this case). The manner in which successive modes can be indexed and counted is indicated by the arrows in the leftmost two columns. For clarity, not all modes are shown in this cartoon.

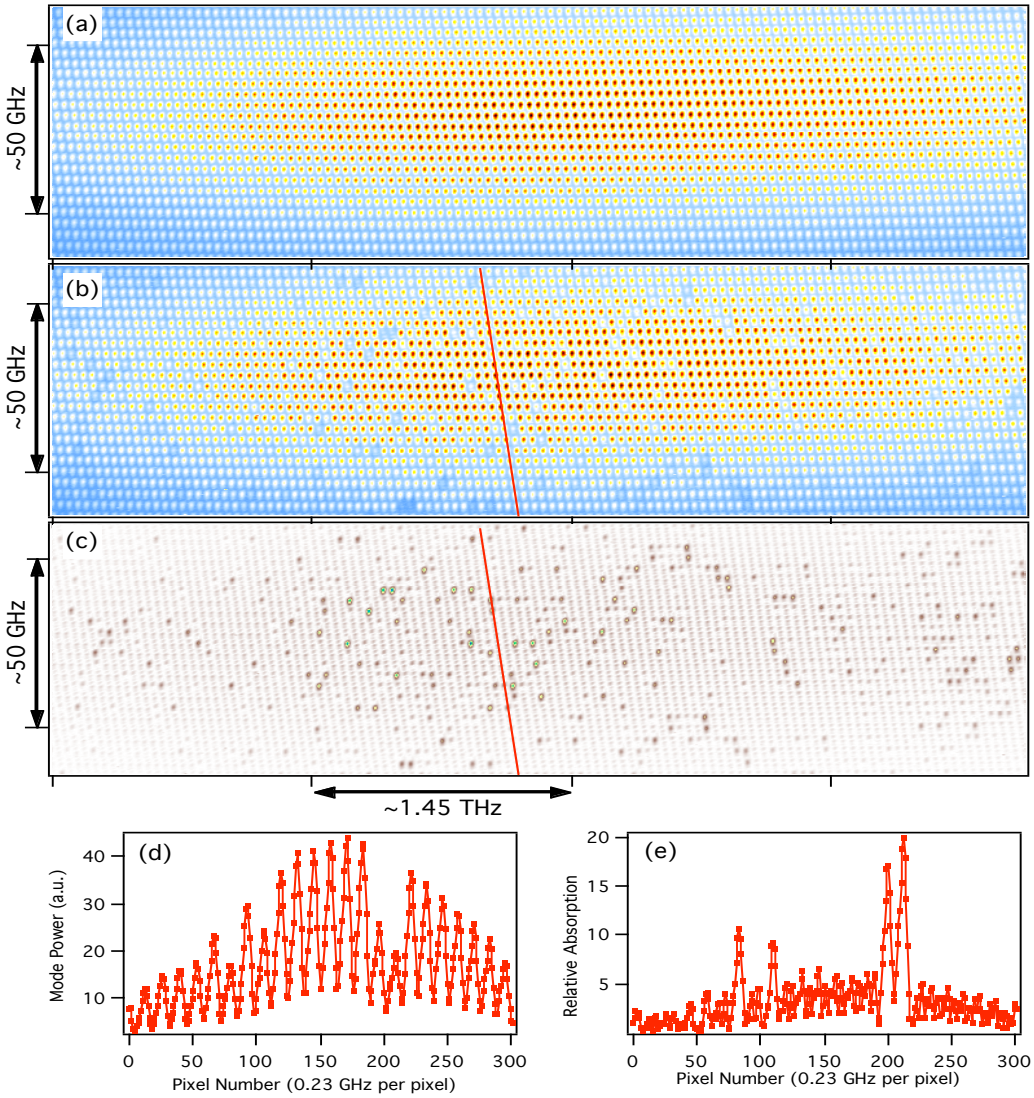

Figure 2: False color spectral-spatial mode images obtained with the high-resolution spectrometer of Fig. 1 showing ~1850 individual modes. (a) Background image with no iodine cell present. (b) Same frame as above, but now with iodine cell present. Note that some modes are missing or noticeably attenuated. (c) The difference of images (a) and (b) with a different false color scale. (d),(e) Line scans along the red lines in (b) and (c), respectively, showing the VIPA resolution of ~1.5 GHz.

The experimental setup is shown in Figure 1. The frequency comb is produced with a broadband Ti:sapphire laser having a repetition rate f_{rep} =1 GHz. Both the carrier-envelope offset frequency (f_0) and f_{rep} are stabilized to a low-noise microwave frequency standard, such that the frequency of each element of the comb may be absolutely determined with fractional uncertainty at or below $\sim 2 \cdot 10^{-13}$ for averaging times of 1 second and longer[10]. Locking the comb to an optical reference would provide resolution at the 10^{-15} level (i.e. ~1 Hz)[11]. The output of the Ti:sapphire laser spans roughly 600-1100 nm, so at present we use an optical bandpass filter to restrict the spectrum to 10 nm of bandwidth around 633 nm (see Fig. 1(a)). Within this 10 nm bandwidth, a Fabry-Perot cavity (finesse ~300) consisting of two spherical mirrors is used to further filter (thin) the frequency comb, by locking the free spectral range (FSR) of the cavity to a harmonic of the pulse repetition rate. For the measurements presented here, the Fabry-Perot in transmission increases the mode spacing of the comb to $3f_{rep}$, with near-unity transmission efficiency for the resonant modes. This increase in mode spacing is required to better match the mode spacing to the resolution of the spectrometer that follows. For the few nanometers of spectrum transmitted through the bandpass filter it is not necessary to consider the dispersion of the Fabry-Perot cavity. However, with the careful design/characterization of the Fabry-Perot mirrors significantly larger bandwidths (~100 nm) can be resonant in such cavities[12].

With 3 GHz mode spacing, the frequency comb output from the Fabry-Perot filter cavity can be spatially/spectrally separated and resolved using a high-resolution spectrometer based on a VIPA[8]. The VIPA is essentially a plane-parallel solid etalon, where the input beam (focused to a line) is injected at an angle through an uncoated entrance widow on the front face. The remainder of the front face is coated with a high-reflective dielectric coating, while the back face has a dielectric coating with 96% reflectivity. The multiple reflections within the VIPA etalon interfere in such a way that the exiting beam has its different frequencies emerging at different angles α. As with all etalons, the VIPA has a free spectral range (~50 GHz in this case), roughly determined by its thickness and index of refraction. The result is that for an input with spectral bandwidth greater than 50 GHz, the output orders are spatially superimposed on each other. To overcome this difficultly, Xiao and Weiner introduced additional dispersion, as provided by a conventional grating, in the spatial direction orthogonal to that of the VIPA[9]. In such a case, it is required that the grating provide spectral resolution better than that of the VIPA's free spectral range, which we achieve for visible light with a 2400 line/mm grating used at a large angle of incidence.

The output of the VIPA/grating spectrometer is imaged onto a CCD camera, resulting in the array of "dots," representing the individual comb modes spaced by 3 GHz. A background image acquired without the iodine vapor cell in the beam path is shown in Fig. 2(a). Approximately 1850 individual modes can be clearly resolved within the ~5.6 THz bandwidth captured on the CCD. In the vertical direction of this image, the data repeats every 50 GHz (VIPA FSR). This is evident in Fig. 2(b), which is an image acquired with the iodine vapor cell inserted in the beam path. The iodine cell is at room temperature and multi-passed to yield an equivalent length of ~2 m. As clearly seen, numerous modes are attenuated. Subtracting Fig. 2(b) from 2(a), yields Fig. 2(c), which shows the absorptive features as peaks. Indexing and counting of the numerous modes follows the scheme presented in Fig. 1(b). Absolute frequency calibration can be achieved by using the previously measured absorption lines or with a frequency-stabilized laser. Given the particular phase-locked values of f_0 and f_{rep}, the absolute frequency of each mode (or equivalently each CCD pixel) can be determined. While the VIPA spectrometer resolution is presently ~1.5 GHz (see line scans of Fig. 2(d) and 2(e)), spectroscopic features down to the linewidth of the comb lines can be distinguished. This would provide an ultimate resolution at the ~1 Hz level when the comb is locked to a suitable optical reference. Indeed, we have observed changes in the pattern of Fig. 2(b) when f_0 or f_{rep} are changed such that the optical modes shift by an amount on the order of the 500 MHz Doppler-limited linewidth. Thus, multiple images taken at different values of f_0 or f_{rep} allow the various absorptive features to be completely mapped.

While the straightforward absorption spectroscopy performed here is sufficient for strong transistions, the high resolution of this approach would benefit from the enhanced sensitivity of broadband cavity ringdown spectroscopy as recently demonstrated by Thorpe, et al[12]. In fact, the ability to spatially isolate and detect the individual modes of the stabilized frequency comb will enable the means to implement in a massively scalable and parallel manner almost any of the powerful CW laser spectroscopic techniques that have been developed. Moreover, these same tools will provide new and unique approaches to arbitrary waveform generation[13] with control at the level of an optical radian, quantum control with ultrahigh spectral resolution, and increased security in novel spread-spectrum optical communication schemes.

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