Linear and Nonlinear Spectroscopy with Dual Frequency-comb Lasers

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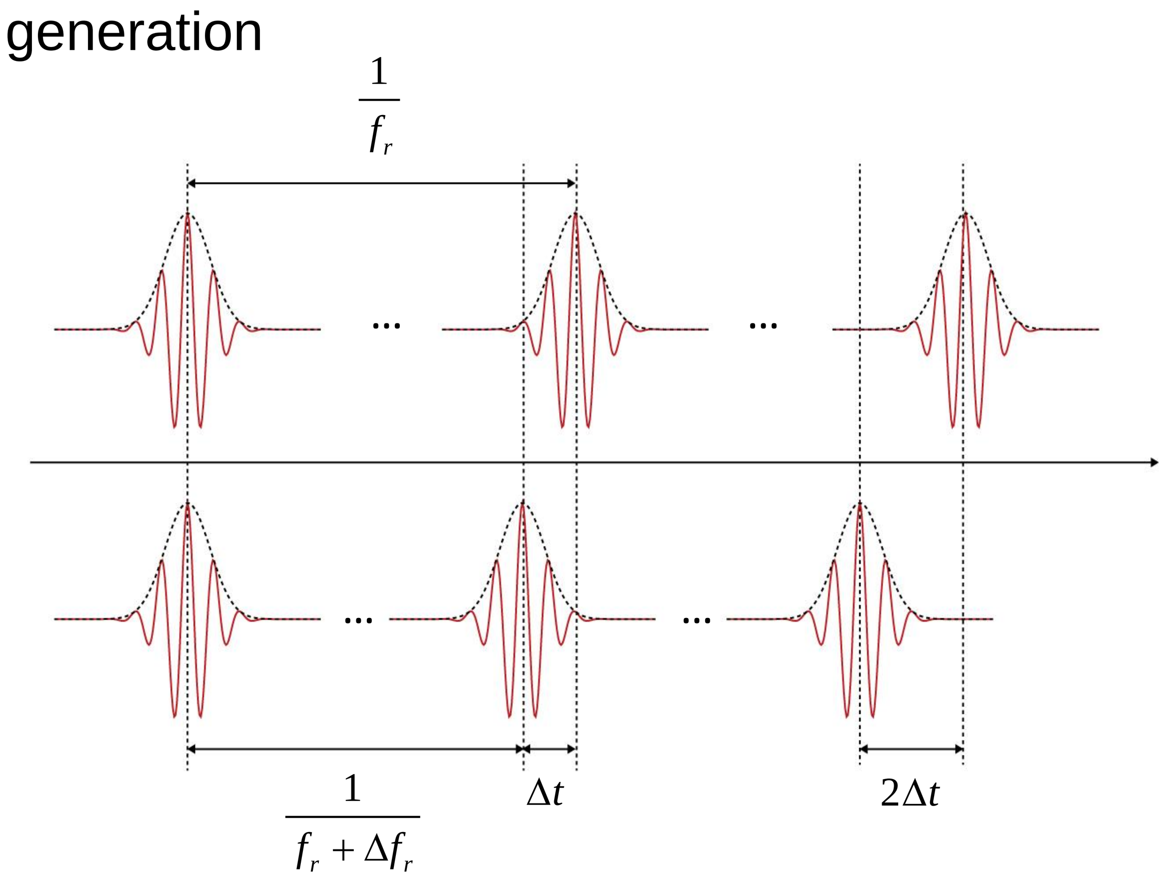
The spectrum of multi-mode laser is composed of equally-spaced narrow lines. The frequency of n-th spectral line of the laser can be expressed as $f_n = n\, f_r + f_{ceo}$, where f_r is the repetition rate, which is determined by the laser cavity length, and f_{ceo} is called the carrier-envelope-offset frequency, which arise from the mismatch between phase velocity and group velocity in the laser cavity. When both f_r and f_{ceo} of a laser are stabilized, the laser is called frequency comb. Based on RF frequency locking techniques, frequency comb has been widely used in physics for precision measurement and gas-phase spectroscopy. In condensed phase, the molecular electronic transition lines is broad as much as tens of THz due to its inhomogeneity and fast relaxation processes. Therefore, the high frequency resolution of frequency comb has not been necessary for condensed phase spectroscopy.

Dual frequency-cob (DFC) is the key to combine the frequency comb and conventional spectroscopy for chemistry. As the beginning point of DFC spectroscopy for chemistry, we have proposed DFC absorption spectroscopy and DFC transient absorption experiment. The broad bandwidth and following short time resolution of our DFC system enable us to resolve broad electronic transition character and ultrafast dynamics of molecular systems in condensed phase. The unique properties of DFC, such as fast time-delay scanning, pulse-to-pulse phase-coherence and simultaneous multi-channel data recording, would enable us to measure unprecedented region of chemistry.

Principle

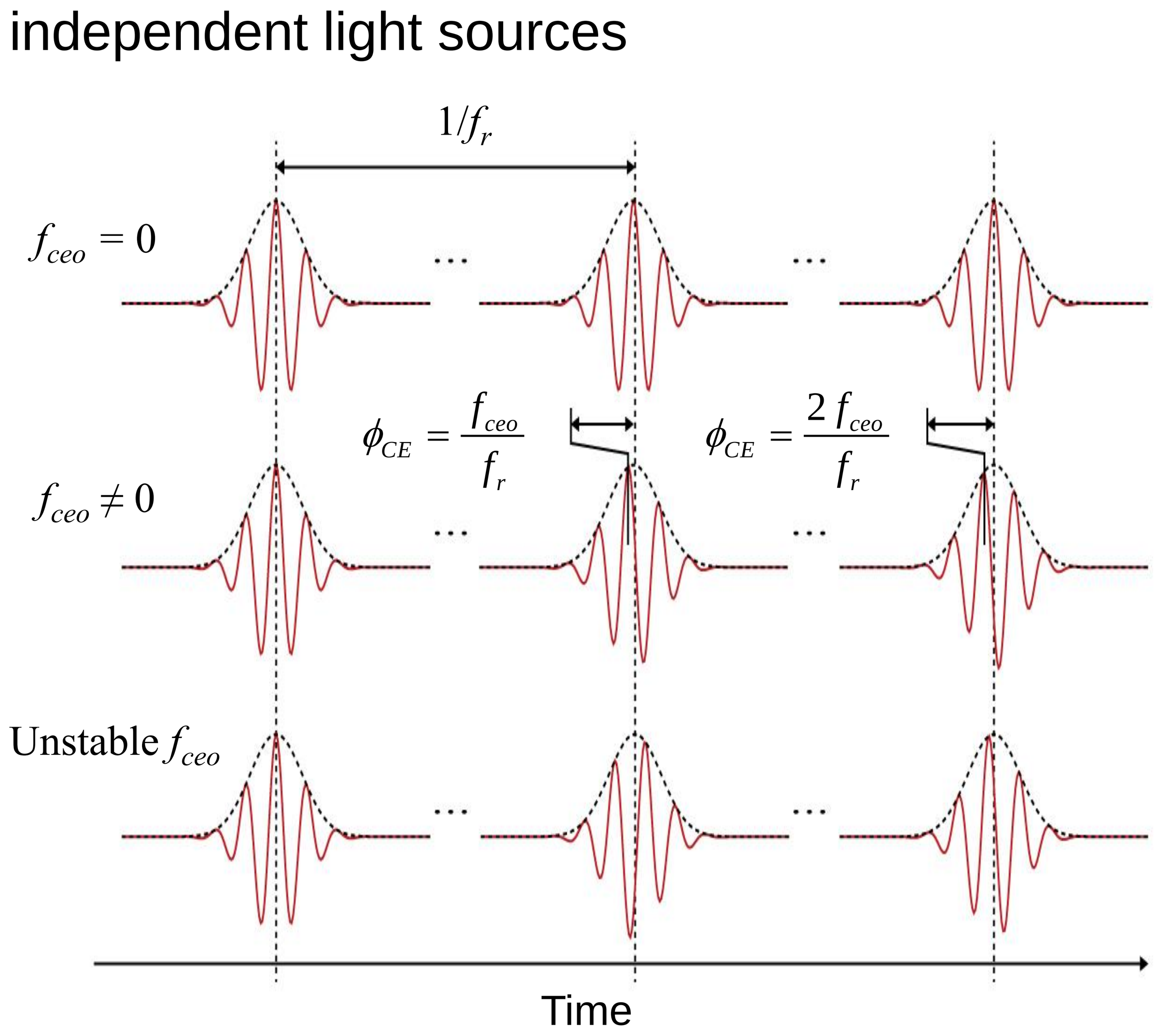
Asynchronous optical sampling (ASOPS) of DFC system : automatic time-delay generation



By detuning the repetition rates of two independent frequency combs, a time-delay is generated with a increment Δt

$$\Delta t = \frac{1}{f_r} - \frac{1}{f_r + \Delta f_r} \cong \frac{\Delta f_r}{f_r^2}$$

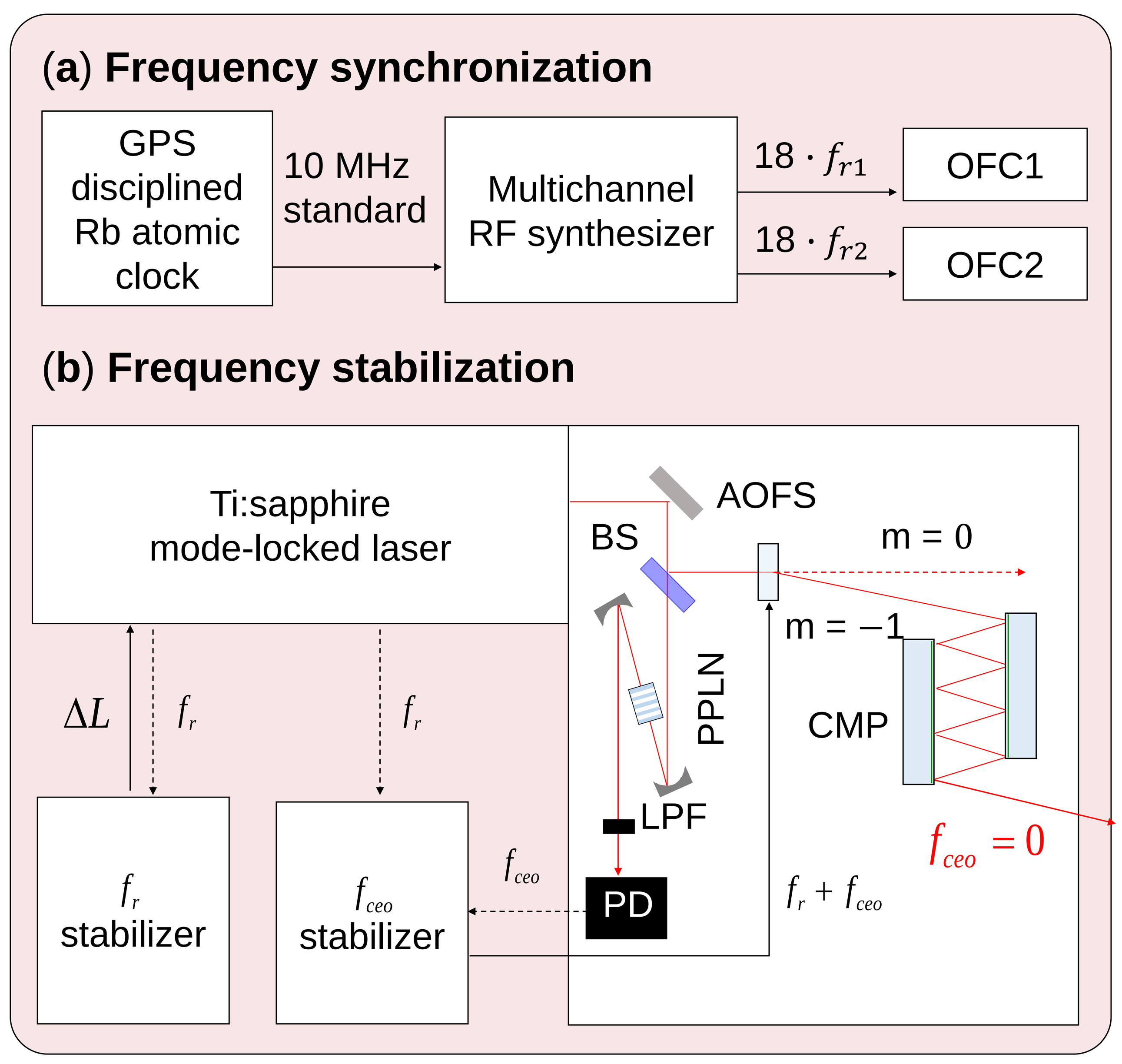
Phase-coherence from f_{ceo} stabilization : the measurement of interference between two independent light sources



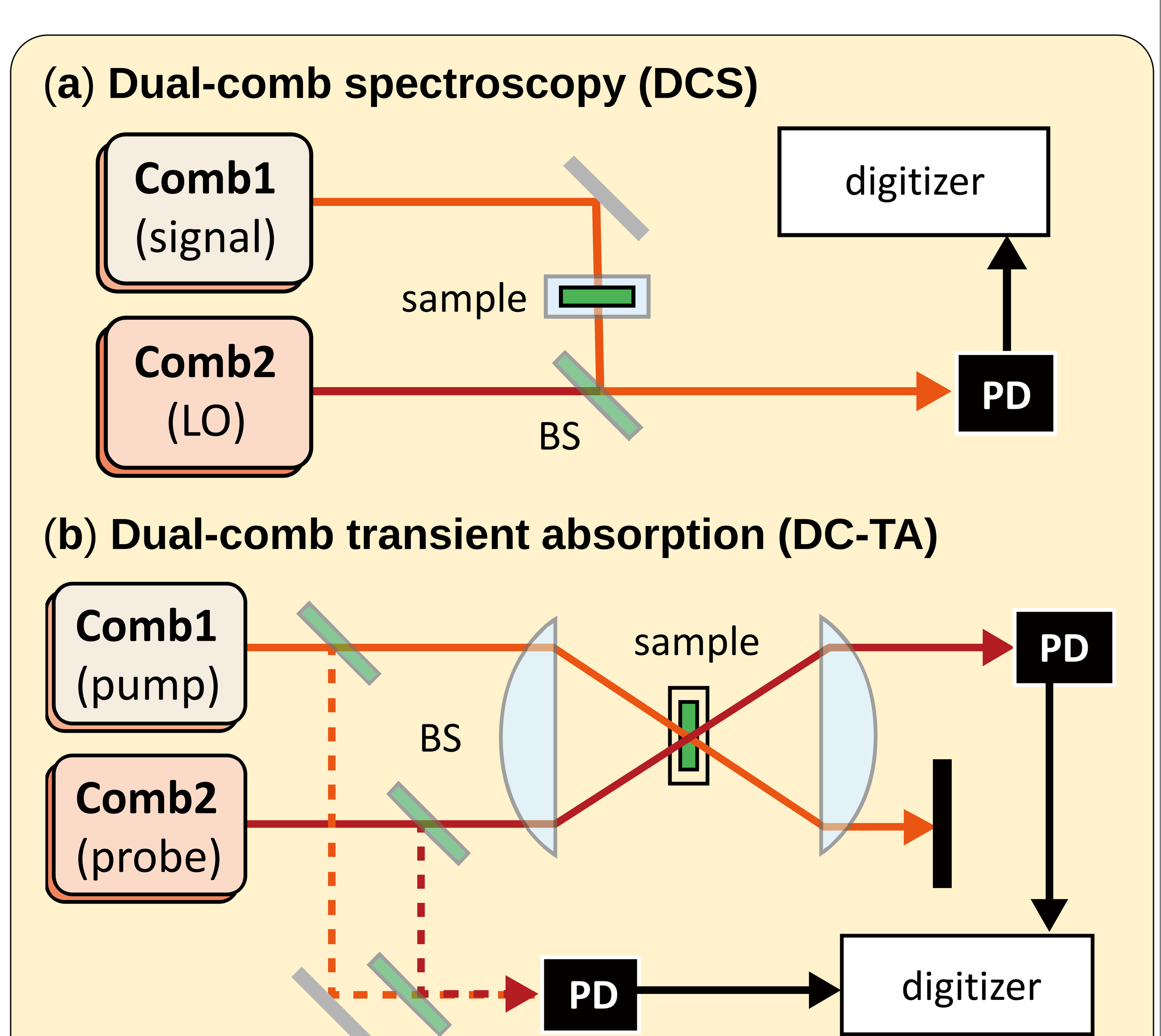
When only f_{ceo} is stabilized, the optical interference in DFC system arises at a down-converted frequency f_{RF}

$$f_{RF} = f_{opt}\frac{\Delta f_r}{f_r}$$

Experiment



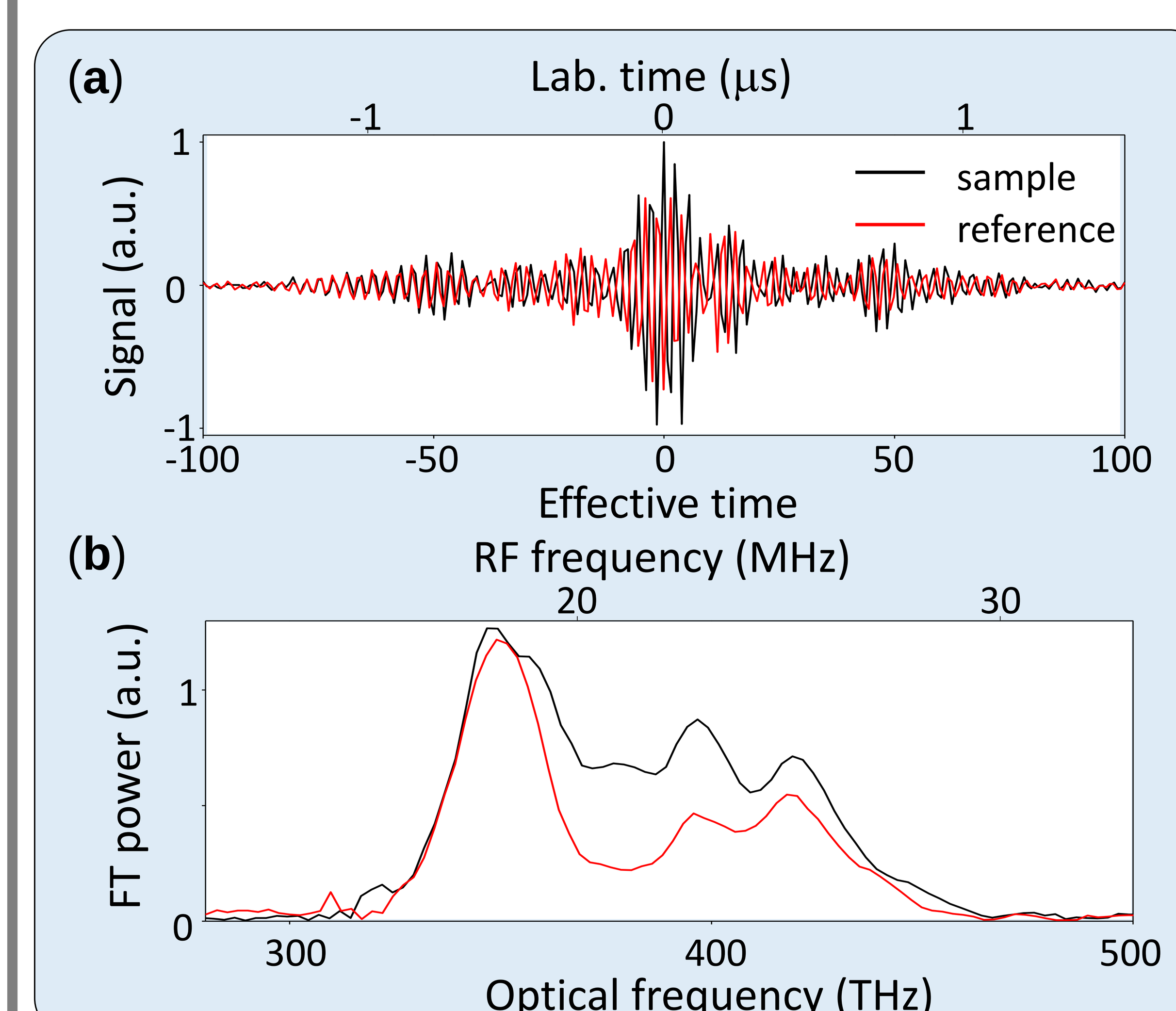
(**a**) The frequency synchronization for DFC system. OFC: optical frequency comb (**b**) The repetition rate of OFC, f_r, is stabilized by controlling the cavity length of a mole-locked laser, while its carrier-envelope-offset frequency, f_{ceo}, is by a feed-forward-loop with an acousto-optic frequency shifter (AOFS). PPLN: periodically-poled lithium niobate, LPF: long-pass-filter



The optical layout of **DCS** and **DC-TA**. LO: local oscillator, PD: photodetector.

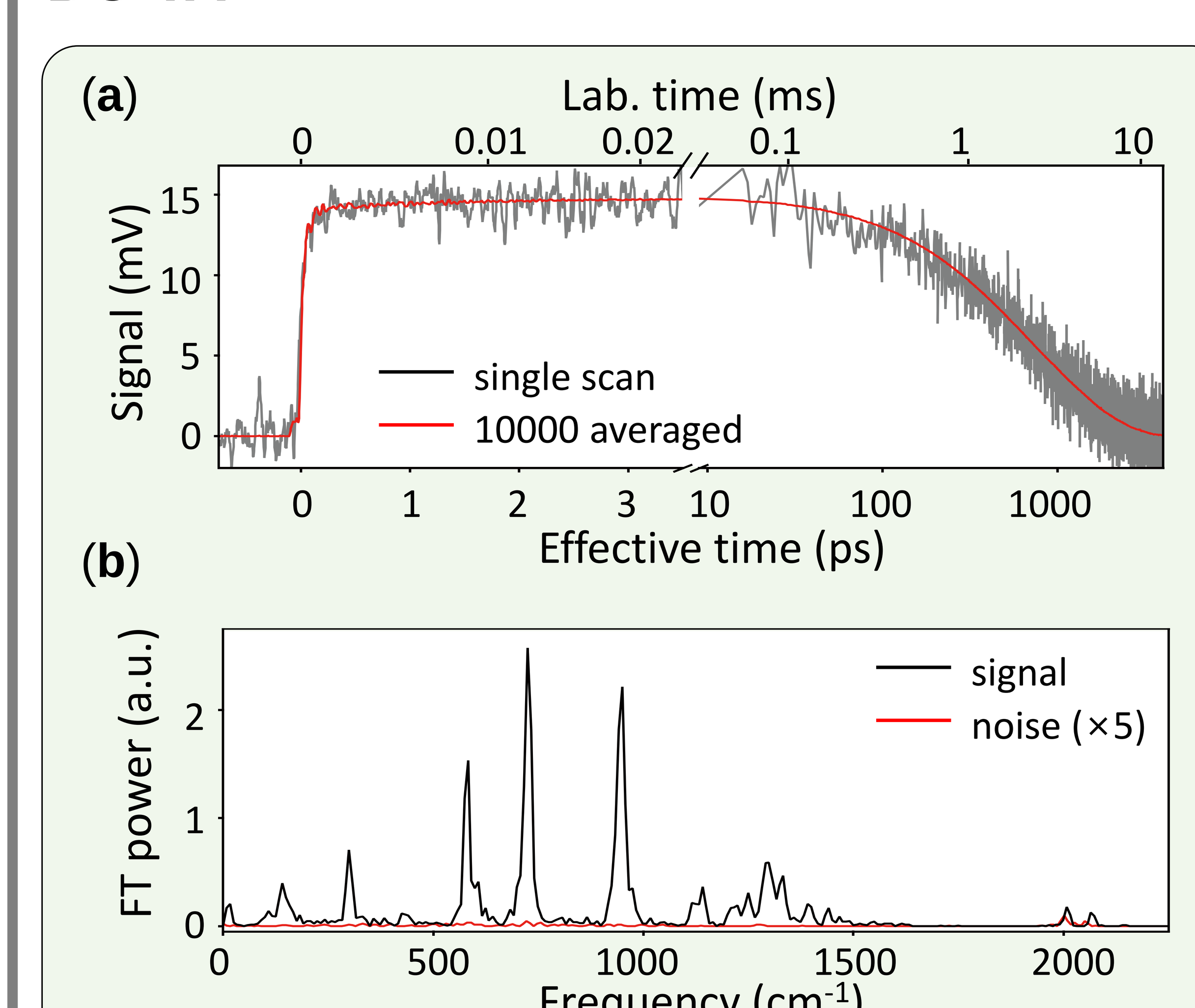
Results

DCS



(**a**) The single scan interferogram between Comb1 and Comb2 with (black) and without sample (red). The sample was IR144 ethanol solution. (**b**) The Fourier transform of (a).

DC-TA



(**a**) The single scan (black) and the ten thousand averaged (red) DFC transient absorption data of IR125 ethanol solution. (**b**) The coherent vibrational spectrum of IR125 ethanol solution. The earlier part , 100 fs to 3 ps, of averaged signal in (a) was Fourier transformed. The noise spectrum was measured at the time window of -5 ps to -3 ps.