Supplementary Information:

Time-stretch infrared spectroscopy

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Supplementary Note 1: Comparison of signal-to-noise-ratio (SNR) between wavelength-swept spectroscopy and Fourier-transform spectroscopy

We theoretically compare SNR of spectra measured by wavelength-swept spectroscopy (WSS) and Fourier-transform spectroscopy (FTS) and show that the former has better SNR than the latter by a factor proportional to $\sqrt{N}$, where N represents the number of spectral elements. Since the detectable optical power is limited by dynamic range of a detector (or a digitizer or an amplifier), we assume a situation where the same optical power, P [W], is detected with the same detector for both the measurements, leading to the same shot noise and detector noise. We also assume the same relative intensity noise (RIN). In this comparison, we treat shot noise, detector noise, and RIN by following an analytical scheme adopted in ref. S1. For simplicity, we assume intensity profiles of measured spectra are uniform (i.e. flat spectra) and omit the contribution of center-bursts of the interferograms measured in FTS. Under this assumption, the shot noise can be treated as additive noise while it is technically multiplicative[S1]. We also treat RIN as additive noise because its multiplicative low frequency noise (typically less than MHz) does not contribute to the high-speed measurement at a rate of MHz to tens of MHz[S1]. We denote the total noise equivalent power including shot noise, detector noise and RIN as $\sigma(P)$ [W $\mathrm{Hz}^{-1/2}$]. We assume spectral data points N are measured for a single spectrum within the same time period for both the measurements. We also assume the effect of digitization noise can be ignored.

Now, we consider the difference between WSS and FTS: the former measures a spectral element at a single sampling point, while the latter measures N spectral points with N sampling points (note that $2N$ sampling points are needed to have N spectral points in FTS, but for simplicity, here we omit the factor 2, which actually causes an additional disadvantage in SNR). We can estimate that both the measurements generate the same signal intensity because they detect the same amount of optical power P in the same measurement time (we also omit an effect that the beat measurement in FTS can generate smaller signal intensity than that in WSS for simplicity). In WSS measurement, the total noise equivalent power can be described as $\sigma(P)\sqrt{f_\mathrm{s}/2}$, where f_s denotes the spectral sampling rate. On the other hand, in FTS measurement, the total noise equivalent power can be described as $\sqrt{N}\sigma(P)\sqrt{f_\mathrm{s}/2}$ because all the noises from N sampling points contribute to that of a single spectral point, leading to the $\sqrt{N}$ larger noise. This noise disadvantage is universal for any FTS techniques. In conclusion, WSS has a SNR advantage proportional to $\sqrt{N}$ compared to FTS.

We comment on the multiplex advantage (or Fellgett's advantage) of FTS that is known as a SNR advantage in FTS. Briefly speaking, the multiplex advantage originates from the fact that FTS can measure multiple spectral elements at once, while a single-channel grating monochromator measures only one element in a measurement and discards the other elements. Therefore, the former takes N-times shorter spectral acquisition time than the latter, which leads to the SNR advantage of FTS. This multiplex advantage of FTS is not applicable to the comparison between WSS and FTS because both the techniques can measure all the multiple spectral elements in the same period of time by detecting the same amount of light.

Supplementary Note 2: Trade-off relation among SNR, spectral resolution and spectral sampling rate

As other spectroscopy techniques, TS-IR also has a trade-off relation among the SNR, spectral resolution and measurement rate (spectral sampling rate). For simplicity, we assume a uniform spectral intensity over the measured spectral region. The number of spectral elements of a single spectrum, N, is described as

$$N = \frac{f_\mathrm{s}}{f_\mathrm{r}},$$

where f_s denotes the spectral sampling frequency, which corresponds to the number of sub-pulses in a second generated by FACED, and f_r the repetition frequency of the laser pulses. The spectral resolution, $\delta\lambda$, is described as

$$\delta\lambda = \frac{\Delta\lambda}{N} = \Delta\lambda\frac{f_\mathrm{r}}{f_\mathrm{s}},$$

where $\Delta\lambda$ denotes the spectral bandwidth of the measured spectrum. Then, the SNR can be described as

$$SNR = \frac{P}{\sigma(P)\sqrt{f_\mathrm{s}/2}} = \frac{\sqrt{2}P}{\sigma(P)}\sqrt{\frac{\delta\lambda}{\Delta\lambda f_\mathrm{r}}},$$

Where P denotes the optical power of the detected pulses, $\sigma(P)$ the total noise equivalent power including shot noise, detector noise and RIN. We see in the equation that the SNR decreases when measuring a larger spectral bandwidth, higher spectral measurement rate, and higher spectral resolution, as expected.

Supplementary References

S1. Newbury, N., Coddington, I. & Swann, W. Sensitivity of coherent dual-comb spectroscopy, *Opt. Express* **18,** 7929-7945 (2010).