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Transient event-locked Fourier spectroscopy

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Abstract

A low-cost method of adding time-resolving capability to commercial Fourier-transform spectrometers with a continuously scanning Michelson interferometer has been developed. It achieves accurate timing by locking the sampling to the transient events and thus eliminates noise and artifacts caused by mirror-speed variations in the interferometer. This acquisition method is accompanied by a novel mathematical algorithm for extracting the spectral information from the data. The method has been used from the far IR to the UV at resolutions up to $0.1\,cm^{-1}$.

Keywords: Time resolved; Fourier; Event-locked

Fourier spectrometers with wide spectral coverage and high resolving powers have become wide spread tools for various types of spectroscopy. Most common are instruments with continuously scanning interferometers. Their time-resolving capabilities are typically very limited. This is partially caused by the limitations of existing methods for time-resolved spectroscopy with such interferometers. The asynchronous [1] and synchronous [2] methods require the transient event under investigation to be repeated at relatively high repetition rates ($\geqslant 1\,kHz$) which also limits their use to short transients. The rapid-scan technique [2] is used for long transients ($\geqslant 0.1\,s$). Transients with lifetimes between 10 μs and 100 ms repeated at low repetition rates, can be studied by the interleaved method [2]. This technique suffers from several disadvantages. We developed the new 'event-locked' method to overcome these problems. The new technique can be used with any continuously scanning interferometer and allows time-resolutions that go beyond the scope of the interleaved acquisition.

A scanning interferometer has the autocorrelation function of the input light as its output. This output is detected and recorded as an interferogram. Numerical techniques with the fast Fourier transform (FFT) as kernel extract the spectral information from the interferogram. This requires the interferogram to be sampled and digitized at even intervals of sufficient density (free spectral range) and for a sufficient scan length (resolution). The subdivision of the scan is typically achieved with the help of a Helium-Neon laser (HeNe) beam passed through the interferometer. Its sequential interference pattern generates the necessary samples requests. For the interleaved method, several consecutive sampling requests are used to obtain interferogram points for several different time delays. While sampling occurs at exactly evenly spaced positions, the time delay of the samples with

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respect to the transient event depends on the speed history of the travelling interferometer mirror. Most transients exhibit a change of the total signal with time. Hence, uncertainty in the time of sampling translates into uncertainty of the sampled signal which causes noise and artifacts. Interleaved experiments [3] show an improvement in S/N with a reduction of mirror speed variations from 2% to 1% (rms), involving elaborate hardware modifications. However, the problem persists. The new approach is to lock the sampling of the interferogram to the transient events instead of the HeNe sampling requests. This assures accurate timing regardless of mirror speed. Speed variations now cause the samples to be unevently spaced, which requires non-traditional analysis techniques [4]. The sampling positions are interpolated from a simultaneously acquired record of the mirror position versus time at the actual time of sampling. The new analysis involves least-squares fits of harmonic functions (sin, cos) to the interferogram. The procedure converges to the well-known Mertz method [5] in the limit of even point spacing and, further, to the Cosine Fourier Transform for zero phase errors. Efficient calculation involves extirpolation (inverse interpolation) and two FFTs.

Without the tight synchronization with the HeNe sampling requests, very flexible set-ups may be designed. Time and spectral resolutions are limited only by the speed and dynamic range of the Analog-Digital Converter. The entire spectral range and resolution of any continuous-scan interferometer may be used. Even the use of free-running sources appears possible. A first implementation with 1 μs time resolution has been used from the far IR to the UV (30–25 000 cm^{-1}) at spectral resolutions up to 0.1 cm^{-1}. Fig. 1 shows a section of the time-dependent emission (forward in 11 μs steps) of Nd^{3+} in the two-site crystal $KLiYF_5$ after short, site-selective excitation with a dye-laser. The good S/N without averaging, at a resolution of 2 cm^{-1}, demonstrates the potential of event-locked Fourier spectroscopy.

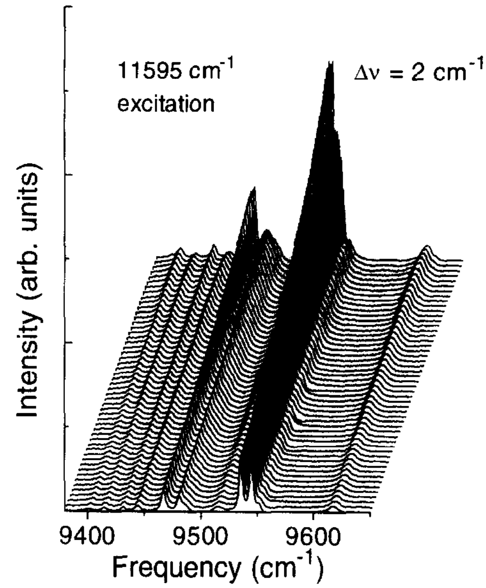

Fig. 1. Time-dependent emission spectra of $Nd^{3+}:KLiYF_5$ demonstrate the potential of event-locked Fourier spectroscopy without averaging (back to front in 11 μs steps).

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