Optimal Fourier Transforms for Probing Oscillatory Rheology of Networks

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Numerous approaches have been proposed for time-resolved oscillatory rheometric protocols that can provide both time- and frequency-resolved measurements on an aging, curing or crosslinking gel system. Previous approaches include multiwave superposition techniques, random/white noise sequences, short-time Fourier transforms and repeated step strain pulses. Here we revisit a common audio signal processing sequence known as the exponential chirp, which offers a number of benefits including (*i*) a frequency-independent strain amplitude and (*ii*) a continuously-varying phase. This chirp sequence enables the linear viscoelastic properties of a 'mutating' (or time-evolving) gel to be rapidly determined over several decades in frequency in $\sim 30-100$ s and has been claimed to be an optimal Fourier transform sequence [1, 2]. However, closer investigation of high-resolution calibration data on a model polymer network shows that regardless of choice of the time-frequency bandwidth parameter, measurement precision can be severely compromised at the highest and lowest frequencies by 'leakage' of material information into side-lobes of the chirp power spectrum. Taking inspiration from the audio sequences used by bats in echolocation [3], we illustrate how these inaccuracies can be resolved through convolution of the chirp sequence with a carefully-chosen windowing function or envelope. The resulting Optimized Windowed Chirp (or OWCh) function provided to the rheometer consists of an exponential chirp multiplied with a Tukey window with window width parameter r and has the form shown in Figure 1:

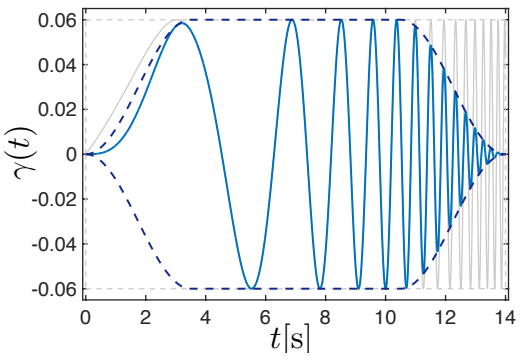

FIG. 1. An Optimized Windowed Chirp (OWCh) function of constant imposed strain amplitude containing frequency information from $0.3 \leq \omega \leq 30$ rad/s.

The optimized waveform/window function can be readily encoded in Matlab and then used to drive the motor of an ARES controlled-strain rheometer. Fast Fourier transformation of the input strain $\tilde{\gamma}(\omega)$ and measured stress response $\tilde{\sigma}(\omega)$ allows direct computation of the linear viscoelastic modulus across a spectrum of frequencies as $G^*(\omega) = \tilde{\sigma}(\omega)/\tilde{\gamma}(\omega)$ in an acquisition time of order $T \simeq 2\pi/\omega_{\min}$.

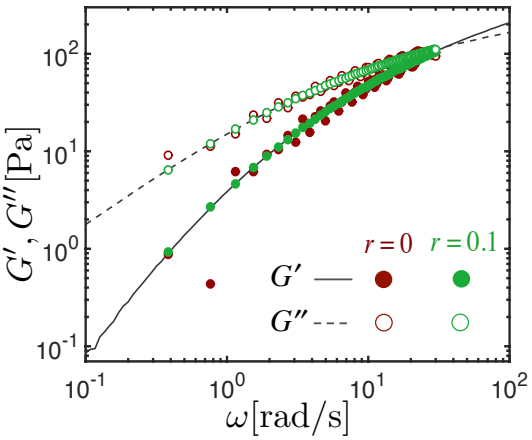

FIG. 2. Use of Optimized Windowed Chirp (OWCh) functions with window width r to extract the linear viscoelastic spectrum of an entangled viscoelastic PIB polymer solution.

Numerical computations and experimental measurements show that the error magnitude can, in fact, be reduced exponentially with window width r through correct selection of the window function, combined with implementation of careful signal conditioning protocols. We present experimental measurements on a semi-dilute entangled polymer solution (cf. Fig. 2), a worm-like micellar fluid and a time-evolving cross-linked biopolymer gel, to show that this approach can indeed rapidly and accurately extract the entire linear viscoelastic spectrum of a time-evolving complex material in less than 15 s, the time typically required to obtain the complex modulus at a single low frequency.

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