

A DUALITY PROOF OF SAMPLING LOCALISATION IN RELAXATION SPECTRUM RECOVERY

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In a recent paper, Davies and Andersson (1997) examined the range of relaxation times, on which the linear viscoelasticity relaxation spectrum could be reconstructed, when the oscillatory shear data were only known on a fixed finite interval of frequencies. In particular, they showed that, for such oscillatory shear data, knowledge about the relaxation spectrum could only be recovered on a specific finite interval of relaxation times. They referred to this phenomenon as sampling localisation. The purpose of this note is show how their result can be proved using a duality argument, and, thereby, establish the fundamental nature of sampling localisation in relaxation spectrum recovery.

In an oscillatory shear experiment, one makes measurements of the amplitude ratio and the phase lag which are then transformed to their equivalent, *storage modulus* $G'(\omega)$ and *loss modulus* $G''(\omega)$ values (see [8]). The determination of the *relaxation spectrum* $H(\tau)$ from such data reduces to solving the following first kind Fredholm integral equations

$$(1) \quad G'(\omega) = \int_0^\infty \frac{\omega^2 \tau^2}{1 + \omega^2 \tau^2} \frac{H(\tau) d\tau}{\tau},$$

and

$$(2) \quad G''(\omega) = \int_0^\infty \frac{\omega \tau}{1 + \omega^2 \tau^2} \frac{H(\tau) d\tau}{\tau}.$$

The difficulty with solving such integral equations is their ill-posedness, which is directly reflected in the sensitivity of $H(\tau)$ with respect to small perturbations in either or both of $G'(\omega)$ and $G''(\omega)$. Various methods have been proposed to overcome this challenge. Some are quite general and apply to all types of Fredholm integral equations (see [6]), while others specifically address the solution of (1) and (2) (see [3]). Among the latter, the most popular are the algorithms which determine a discrete relaxation spectrum $\{\tau_i, g_i\}$, where τ_i and g_i denote, respectively, the relaxation time and the elastic modulus of the i -th Maxwell mode. In the version in [6], the relaxation times τ_i are assumed to be known and Tikhonov regularisation is utilised to allow a large number N of modes to be fitted. On the other hand, in [3] the algorithm is non-linear,

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because it fits both the elastic moduli and the relaxation times simultaneously. It achieves stabilisation through the non-linear fitting and the restriction of the number N of the modes to be small.

An alternative strategy to that in [1], not yet widely applied in the rheological literature, is to limit attention to the specific information required about the phenomenon of interest, which, in the current context, would correspond to linear (inner product) functionals of the relaxation spectrum; namely,

$$(3) \quad L_\theta(H) = \int_0^\infty \theta(\tau)H(\tau) d\tau,$$

where the function θ , which characterises the form of this linear *solution-functional*, is known. The clear advantage of this approach is that, formally, such linear functionals can often be redefined as corresponding linear (inner product) *data-functionals*, which, in the current context, would take either of the following forms

$$(4) \quad L_{\phi'}(G') = \int_0^\infty \phi'(\omega)G'(\omega) d\omega,$$

or

$$(5) \quad L_{\phi''}(G'') = \int_0^\infty \phi''(\omega)G''(\omega) d\omega.$$

Consequently, once the form of $\phi'(\omega)$ or $\phi''(\omega)$ has been determined for a given θ , the problem of evaluating the solution-functional $L_\theta(H)$ has been reduced to simply performing a smoothing operation on the observational data for the storage and loss moduli, in order to estimate the data-functional $L_{\phi'}(G')$ or $L_{\phi''}(G'')$.

It is shown in [4] how to construct the functions $\phi'(\omega)$ and $\phi''(\omega)$ which correspond to the following solution-functionals

$$(6) \quad L_\theta(H) = \eta_{ab} \approx \int_a^b H(\tau) d\tau, \quad \text{and} \quad L_\theta(H) = g_{ab} = \int_a^b \frac{H(\tau)}{\tau} d\tau, \quad 0 \leq a < b < \infty,$$

which define the *partial viscosity* η_{ab} and the *elastic modulus* g_{ab} over intervals of relaxation times (a, b) .

The basic lemma, which they utilise, formalises the Fourier inversion deconvolution applied by [5] when analysing the electrical properties of polar polymers. Using this Lemma, [4] essentially showed that, formally, the construction of functions such as $\phi'(\omega)$ and $\phi''(\omega)$ reduces, in one way or another, to taking inverse Fourier transforms of functions of the form

$$(7) \quad \Phi(\tau) = \hat{\Theta}(\tau) \cosh\left(\frac{1}{2}\pi\tau\right), \quad \hat{\Theta}(\tau) = \int_0^\infty \theta(\tau) \exp(-i\tau \ell n \tau) d \ell n \tau,$$

where $\hat{\Theta}(\tau)$ denotes the logarithmic Fourier transform of $\theta(\tau)$. For example, $\hat{\Theta}(\tau) = 1$, when $\theta(\tau) = \delta(\ell n \tau)$, the Dirac delta function centred at $\tau = 1$, and $\hat{\Theta}(\tau) = \exp(i\tau)$, when $\theta(\tau) = \delta(\ell n(\tau/t))$. An analysis of this result was then used to establish various

sampling localisation constraints on the recovery of estimates of the relaxation spectrum $H(\tau)$ from oscillatory shear measurements. In particular, [4] demonstrated that the widespread belief that measurements performed on the frequency range $\omega_{\min} < \omega < \omega_{\max}$ determine the relaxation spectrum on the reciprocal range $\omega_{\min}^{-1} < \tau < \omega_{\max}^{-1}$ is imprecise. The correct interval on which the relaxation spectrum is determined is

$$(8) \quad \exp(\pi/2)\omega_{\min}^{-1} < \tau < \exp(-\pi/2)\omega_{\max}^{-1},$$

which is *shorter* than the reciprocal frequency range by 1.36 decades (approximately). These results have been used to construct moving average formulae for the determination of elastic moduli of equation (6) (see [2]).

As explained in [4] the functions $\phi'(\omega)$ or $\phi''(\omega)$ possibly correspond to some type of distribution, and it is the knowledge of their support which gives (8). Due to the importance of (8) for experimental rheology and relaxation spectrum recovery, the question naturally arises as to the exact nature of these and the relaxation spectra to which they pertain. In terms of equation (7), this reduces to examining the properties of objects of the form

$$(9) \quad \kappa_f = \mathcal{F}^{-1}(\Phi(r)) = \mathcal{F}^{-1}(\hat{f}(r) \cosh(\lambda r)), \quad \hat{f}(r) = \hat{\Theta}(r),$$

where \mathcal{F}^{-1} denotes the inverse Fourier transform.

For $\lambda > 0$ (a constant), let

$$(10) \quad \xi_\lambda(r) = \cosh(\lambda r), \quad (r \in \mathbb{R}).$$

Take $1 \leq p \leq 2$ and consider the space

$$(11) \quad F[\lambda, p] = \{f \in L^1(\mathbb{R}) \mid \xi_\lambda \cdot \hat{f} \in L^p(\mathbb{R})\}$$

which is taken to have the graph topology; namely,

$$(12) \quad \|f\|_{F[\lambda, p]} = \|f\|_1 + \|\xi_\lambda \cdot \hat{f}\|_p.$$

Since the Fourier transform of a Gaussian is, up to a factor of modulus one (that is, up to an arbitrary translation), another Gaussian, such functions lie in $F[\lambda, p]$. Their linear span is dense in $L^1(\mathbb{R})$, so $F[\lambda, p]$ is large from a practical point of view.

Given $f \in F[\lambda, p]$, consider the functional κ_f on $L^p(\mathbb{R})$ defined by

$$(13) \quad \langle \kappa_f, g \rangle = \int_{-\infty}^{\infty} \xi_\lambda(r) \hat{f}(r) \hat{g}(r) dr = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \cosh(\lambda r) (f * g)(s) e^{-irs} ds dr.$$

By the Hölder and Hausdorff-Young inequalities,

$$(14) \quad |\langle \kappa_f, g \rangle| \leq \|\xi_\lambda \cdot \hat{f}\|_p \|\hat{g}\|_q \leq \|\xi_\lambda \cdot \hat{f}\|_p \|g\|_p,$$

where $1/p + 1/q = 1$. So this functional is continuous, and thus given by a function in $L^q(\mathbb{R})$ which we again denote by κ_f . Because of the presence of the $\cosh(\lambda r)$ term, one cannot change the order of integration on the right hand side of (13). For each $\varepsilon > 0$, set

$$(15) \quad \langle \kappa_f(\varepsilon), g \rangle = \int_{-\infty}^{\infty} \xi_\lambda(r) \hat{f}(r) \hat{g}(r) e^{-\varepsilon^2 r^2 / 2} dr.$$

Changing the order of integration on the right hand side of this last equation is valid, and a short calculation yields

$$(16) \quad \langle \kappa_f(\varepsilon), g \rangle = \sqrt{2\pi} \int_{-\infty}^{\infty} \frac{1}{\varepsilon} \exp\left(\frac{1}{2\varepsilon^2}(\lambda^2 - s^2)\right) \cos\left(\frac{\lambda s}{\varepsilon^2}\right) (f * g)(s) ds.$$

In the limit as $\varepsilon \rightarrow 0$, the integrand will tend to zero dominatedly on $|s| > \lambda + \delta$ for any $\delta > 0$.

Now

$$(17) \quad \left| \langle \kappa_f - \kappa_f(\varepsilon), g \rangle \right| \leq \int_{-\infty}^{\infty} \xi_\lambda(r) |\hat{f}(r) \hat{g}(r)| \left(1 - \exp\left(-\frac{1}{2}\varepsilon^2 r^2\right) \right) dr.$$

Since $\xi_\lambda \cdot \hat{f} \in L^p(\mathbb{R})$, $\hat{g} \in L^q(\mathbb{R})$, and $1 - \exp(-\varepsilon^2 r^2 / 2) \rightarrow 0$ and is bounded by 1, it follows immediately from the dominated convergence theorem that $\langle \kappa_f - \kappa_f(\varepsilon), g \rangle \rightarrow 0$ as $\varepsilon \rightarrow 0$.

This establishes that, for any $\delta > 0$,

$$(18) \quad \langle \kappa_f, g \rangle = \sqrt{2\pi} \lim_{\varepsilon \rightarrow 0} \int_{|s| < \lambda + \delta} \frac{1}{\varepsilon} \exp\left(\frac{1}{2\varepsilon^2}(\lambda^2 - s^2)\right) \cos\left(\frac{\lambda s}{\varepsilon^2}\right) (f * g)(s) ds.$$

Hence, this limit exists with value equal to the limit of $\langle \kappa_f(\varepsilon), g \rangle$, and moreover is independent of $\delta > 0$.

As a consequence, it follows that $\langle \kappa_f, g \rangle$ is independent of the values of $(f * g)(s)$ for $|s| > \lambda$, with the form of κ_f depending on the choice of f . In particular, consider the functions $f_{[a,b]}$ that have support in the interval $[a, b]$. Suppose further that g vanishes on $[a - \lambda, b + \lambda]$. When $|s| \leq \lambda$ and $t \in [a, b]$, then $t - s \in [a - \lambda, b + \lambda]$, so that $g(t - s) = 0$. Then

$$(f_{[a,b]} * g)(s) = \int_a^b f(t) g(t - s) dt = 0$$

for $|s| \leq \lambda$. But, this means that $\langle \kappa_{f_{[a,b]}}, g \rangle = 0$ for all such $g \in L^p(\mathbb{R})$, whatever their behaviour outside $[a, b]$. This can only happen if the support of $\kappa_{f_{[a,b]}}$ is contained in $[a - \lambda, b + \lambda]$.

There is an alternative approach for the case $p = 2$. On taking $f \in F_{[\lambda, 2]}$ with support in $[-b, b]$, it follows that \hat{f} is an entire function satisfying $|\hat{f}(z)| \leq \text{const} \cdot e^{b|\text{Im } z|}$, so that

$$|\hat{f}(z) \cosh(\lambda z)| \leq \text{const} \cdot e^{b|\text{Im } z| + \lambda|\text{Re } z|} \leq \text{const} \cdot e^{(b+\lambda)|z|}.$$

Since $\xi_\lambda \cdot \hat{f} \in L^2(\mathbb{R})$, it follows from the Paley-Weiner theorem [7] that κ_f has support in $[-b - \lambda, b + \lambda]$.

In this way, it is established that, for suitable data functionals of compact support, and modulo a logarithmic change of variable, functions such as $\phi'(\omega)$ and $\phi''(\omega)$ will have compact support.

As a direct result, the sampling localisation of [4] has been placed on a rigorous footing.

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