# Large Amplitude Oscillatory Shear of the Prandtl Element Analysed by Fourier Transform Rheology

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#### ABSTRACT:

This work contributes to the theory of strain controlled large amplitude oscillatory shear (LAOS) as well as modelling the key properties of type III behavior of Hyun, the decreasing storage modulus and a loss modulus with considerable maximum. The latter two can be modelled with the help of the Prandtl element. Since it is a yield stress fluid, the use of LAOS is necessary to calculate the storage and loss modulus. Furthermore, a condition is presented which has to be met in order to avoid even harmonics. The storage and loss modulus as well as the higher harmonics of the Prandtl element are determined analytically in this work. They are given as mathematical functions which can be discussed conveniently. This allows the identification of characteristic points which are related to material parameters of the Prandtl element and enable a physically motivated material parameter identification. Beside this, it is observed that the yield strain do not coincide with the crossover  $G'(\hat{\gamma}) = G''(\hat{\gamma})$  but with the increasing of the loss modulus and the decreasing of the storage modulus. Thanks to the analytical calculations, it is also obvious that the stress response of yield stress fluids does not necessarily include even harmonics. In this work the steady state stress response of the Prandtl element is also presented as Lissajous plots and Pipkin diagrams to visualise the rheological fingerprint.

#### KEY WORDS:

large amplitude oscillatory shear, Fourier Transform rheology, harmonic analysis, storage modulus, loss modulus, yield strain, Prandtl element, Lissajous plot, Pipkin diagram

## **1** INTRODUCTION

In recent years, strain controlled large amplitude oscillatory shear (LAOS) has been developed into a powerful tool to investigate non-linear material behavior. Therefore, a sinusoidal strain is imposed and the stress response is measured. The latter can be decomposed by a harmonic analysis, the Fourier Transform (FT) rheology [1-11], into the storage modulus G', the loss modulus G" as well as the higher harmonics  $a_k$  and  $b_k$ . These are material functions which tell about the material behavior and help drawing conclusions about model assumptions. Based on these, one can create a material model and is able to do further simulations. The storage and loss modulus can be also used for material classification. For example Hyun et al. [10] presented a classification depending on the LAOS behavior of  $G'(\hat{\gamma})$  and  $G''(\hat{\gamma})$  which consists of four types. Another way to present the characteristic of materials is the visualisation of their stress responses. Here it has become established to plot the Lissajous curves as well as the Pipkin

diagrams, the so called rheological fingerprint [12–14]. Furthermore, sinusoidal loading is an useful tool to investigate the rate-dependency  $\dot{\gamma} \sim \hat{\gamma} \omega$  within a limited range of strain  $-\hat{\gamma} \leq \gamma \leq \hat{\gamma}$  by varying the angular frequency  $\omega$  [3, 10, 11, 15]. Thus, the critical strain  $\gamma_{cr}$ , which describes the limit of linearity to separate the range of small amplitude oscillatory shear (SAOS) from LAOS, can be determined within a high sensitivity investigation [8, 16]. After passing the limit of linearity the Fourier analysed stress response contains higher harmonics. Often the lack of physical interpretation of these higher harmonics is emphasised [17 – 20]. For improvement, new approaches have been suggested like (1) the Fourier Chebyshev analysis of Ewoldt [19] applying the stress decomposition of Cho [18], (2) the method of Klein [21] using sine, square, triangular and sawtooth waveforms as four fundamental basis functions, and (3) the sequence of physical processes technique of Rogers [15]. Further details as well as the state of the art are summarised elsewhere [11, 20, 22–24] making such a discussion redundant at this place.

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Prandtl element three characteristic points were identified by the storage and loss modulus so that the elastic modulus G and the yield stress  $\tau_{y}$  can be determined for example from the yield point: the critical strain (Equation 24) and the preyield-plateau of the storage modulus (Equation 33). The parameters also can be identified by the maximum point of the loss modulus according to Equation 39. Characteristic points were also found in higher harmonics. Thus the root of  $a_{p.k,odd}$ (Equation 45) and each maximum MAX{*b<sub>p.k,odd</sub>*} (Equation 50) can be used in addition to calculate G and  $\tau_{\gamma}$ . In principle, there is only one point that is relevant for the Prandtl element to identify its material parameters. However, to determine material parameters of soft matter whose LAOS behavior can partially modelled by a Prandtl element the knowledge of more characteristic points is useful. Furthermore, it was observed that the critical strain does not have to coincide with the crossover  $G'(\hat{\gamma}) = G''(\hat{\gamma})$  in any case. In case of the Prandtl element the beginning of the postyield results a priori by the constitutive equations. It happens at the critical strain which occurs at the increasing of the loss modulus and the decreasing of the storage modulus. This shows the need to be careful about conclusions that are drawn a posteriori about material systems by experimental analysis methods. Beside this, it was proved analytically that the LAOS stress response of yield stress fluids does not include even harmonics imperatively as it is sometimes considered. Even harmonics vanish in case of the Prandtl element because its stress response fulfil Equation 9. Another presentation of the LAOS behavior of the Prandtl element, its rheological fingerprint, was given by the Lissajous plots and the Pipkin diagrams. They were shown at the end of Section 3.2. Depending on  $\tau_{v}/(G\hat{\gamma})$ , the Prandtl element behave either in Mode 1 or Mode 2. Since they differ significantly from each other, it became clear that a rheological element does not necessarily generate only one characteristic first and second Lissajous plot. In general, it may has several typical Lissajous plots. Both modes have in common that the preyield and postyield parts are separated by sharp corners similarly to the storage and loss modulus. Latter also show a sharp transition from the preyield regime to the postyield at the critical strain because the constitutive equations of the Prandtl element contain a yield function and a yield condition. The sharp transition is expressed by the sudden increase of  $G''(\hat{\gamma})$  and the decrease of  $G'(\hat{\gamma})$ . Thus, independent from the fact that the storage and loss modulus are quantities which integrate the signal over the period according to Equations 5 and 6, they are able to detect the yielding if an amplitude sweep is considered.

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