## Nonlinear magnetorheology- A new method to describe induced structural arrangements of complex magnetic suspensions

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Heavy and extra-heavy crude oils are framed as the mainstay of the energy market due to their large availability in world reservoirs.<sup>1</sup> However, their well-discussed compositional complexity, which mainly promotes undesirable phase transitions in asphaltene nanoaggregates, induces disruptive and expensive production, transport, and refining. On this basis, adsorptive phenomena involving nanomaterials and asphaltenes have emerged as promising technological alternatives, since outstanding structural changes inside the oil matrices are promoted (e.g.,inhibiting the formation of complex long-range viscoelasticity).<sup>2</sup> We have further studied for the first time, the effect of adsorbed asphaltenes on the magnetic dipolar interactions of dispersed superparamagnetic iron-based nanoparticles in magnetic-modified heavy oils (i.e., mixtures of Fe<sub>3</sub>O<sub>4</sub> kerosene-based ferrofluids and heavy oil), proving by magnetorheology, as well as susceptibility and demagnetizing field concepts, that below certain critical concentrations, adsorbed asphaltenes stimulated "enhanced" positive changes in the viscoelastic properties, which might be interpreted as additional physical attractive forces that can be favorably manipulated by external means. This "enhanced" increase is presumably the result of interdigitation phenomena and magnetic dipolar interactions between the magnetic cores. Although, under the low concentrations (0.1 and 1 wt.%), asphaltene molecules are barely able to saturate the surface of the nanoparticles, which favors interdigitation; both attractive forces could be gradually hindered if the concentration increases, since the aliphatic swelling side chains are more exposed, owing to the formation of multiple asphaltene layers which stimulate a steric repulsive barrier between particles.<sup>3</sup> Despite the discussed hindering phenomena, the sequence of physical processes (SPP) analysis, carried out during the nonlinear viscoelastic characterization, proved that disordered asphaltene aggregates are highly extended and naturally formed in the absence of the magnetic field. However, in the presence of a uniform field, using a controlled rate magneto-rheometer, the formation of interacting structural arrangements was induced (Figure 1), analogous to ferrofluids-magnetically controlled suspensions. In this regard, this research provided a precise extension of the description of the effect of magnetic fields on the microstructural organization of complex magnetically modified fluids using rheology and magnetometry.



Figure 1 Physical interpretation of LAOS responses constituted by the transient moduli plotted parametrically against each other in a temporal Cole–Cole plot. A. In the absence, and B. In the presence of a magnetic field.

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- 2. M. D. Contreras–Mateus, et al., Fuel, 285, 119184 (2021).
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