Discussion of 004405JOR by M. Laurati et al.

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Question: Roger Bonnecaze: From your understanding of the interplay among interparticle forces, microstructural dynamics, and macroscopic properties of attractive colloidal gels, how would you advise users to formulate the gels to achieve desired properties such as moduli, yield stress, yield strain, and viscosity? In short, how does microscale understanding inform design of these important materials?

Author Response: Marco Laurati: All the discussed talks indeed show that the response of gels to shear depends in a detailed way on the structural organization of the particles, in particular, cluster sizes and network connectivity, but also bond strengths. These are in turn determined by the particle-particle interactions and the route leading to gelation, which is often a nonequilibrium route. Moreover, the application of shear can also influence and modify this structural organization and be used to tune the structure using the control parameters of the shear field. So the important control parameters for designing the materials are the characterization and knowledge of the interactions, but also the determination and control of the processing step of the material. A combined control of these two aspects might lead to the possibility of controlling the design of materials with desired properties.

Question: Emanuela Del Gado: Can you comment on the shape of the LB plots? Do you interpret them in terms of a strain hardening or do you think there is a different explanation?

Author Response: Marco Laurati: The shape of the elastic Lissajous–Bowditch plots indeed may directly reveal an intracycle strain hardening response to a trained observer. This is also confirmed by the additional Chebyshev and Anharmonic Moduli analyses presented in the paper on which we rely to discuss intracycle responses. The latter, taking into account all higher harmonic contributions, indicates an in-cycle strain hardening around and beyond the yield point of the gel.

Question: Ralph Colby: How can the two samples in Fig. 1 have nearly identical $G''(\omega)$ yet very different $G'(\omega)$ when the data are supposed to be linear viscoelastic response and the two moduli need to be the out-of-phase and in-phase parts of the same complex function? Do these data satisfy the Kramers–Krönig relation?

Author Response: Marco Laurati: To accurately apply the Kramers–Krönig relations, a sufficiently extended frequency region has to be sampled. In our measurements, the frequency region is rather limited and in particular does not extend to small enough frequencies, where the frequency dependence of the moduli of the two samples starts are expected to deviate from each other significantly. Indeed, the storage modulus of the gel with smaller polymer concentration starts to decrease fast in this region, indicating the approach to a structural relaxation, while the G' of the higher polymer concentration gel still shows a weak frequency dependence.