

Supplementary Material: Flow of microgels with different softness at the interface reproduced by a multi-Hertzian model

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REFERENCE SYSTEM

A 2D system of microgels interacting with the square shoulder multi-Hertzian model was investigated under equilibrium conditions as a function of generalized area fraction ζ_{2D} . The structure can be characterized using the radial distribution function, which are depicted in Fig. S1 for a set of reference parameters. Initially, the system is a liquid with a low amount of oscillation in the RDF and a first peak positioned at the diameter $d = 1.0$ of the particles. Starting from ζ_{2D} the system turns solid, indicated by more and higher peaks. Curiously, starting from the second peak each maxima is split, due to the multi-Hertzian nature of the potential. Furthermore, the microgels start to be compressed with increasing generalized area fraction, as can be seen by the shift of the first peak to lower r . Pushing the generalized area fractions beyond 2.50 leads to the first peak being at distance $> d_{out}$ and the cores start interacting. This gives rise to a secondary phase, characterized by core-core contacts, which can be seen as a new peak in the radial distribution function positioned at $r/d = d_{core}$.

Applying an oscillating external shear to these systems, allows the computation of storage, G' , and loss modulus, G'' , which are plotted in Fig. S2. Quite clearly, for $\zeta_{2D} = 0.5 \& 0.75$, the loss modulus exceeds the storage modulus at low angular frequencies, characterizing them as liquids. Increasing the generalized area fraction leads to the glass transition, and, therefore, a higher storage than loss modulus by about one order of magnitude.

AMPLITUDE SWEEPS

Figure S3 depicts Amplitude sweeps of our reference system. We note that especially for liquid systems at low strain amplitude the measured stress signal can be of similar magnitude to that of the general noise, which leads to rather inaccurate results in these regimes. At all area fractions storage and loss modulus appear to be constant up to a strain of 3%, marking the linear viscoelastic regime. Even higher strains lead to an expected decrease in storage modulus.

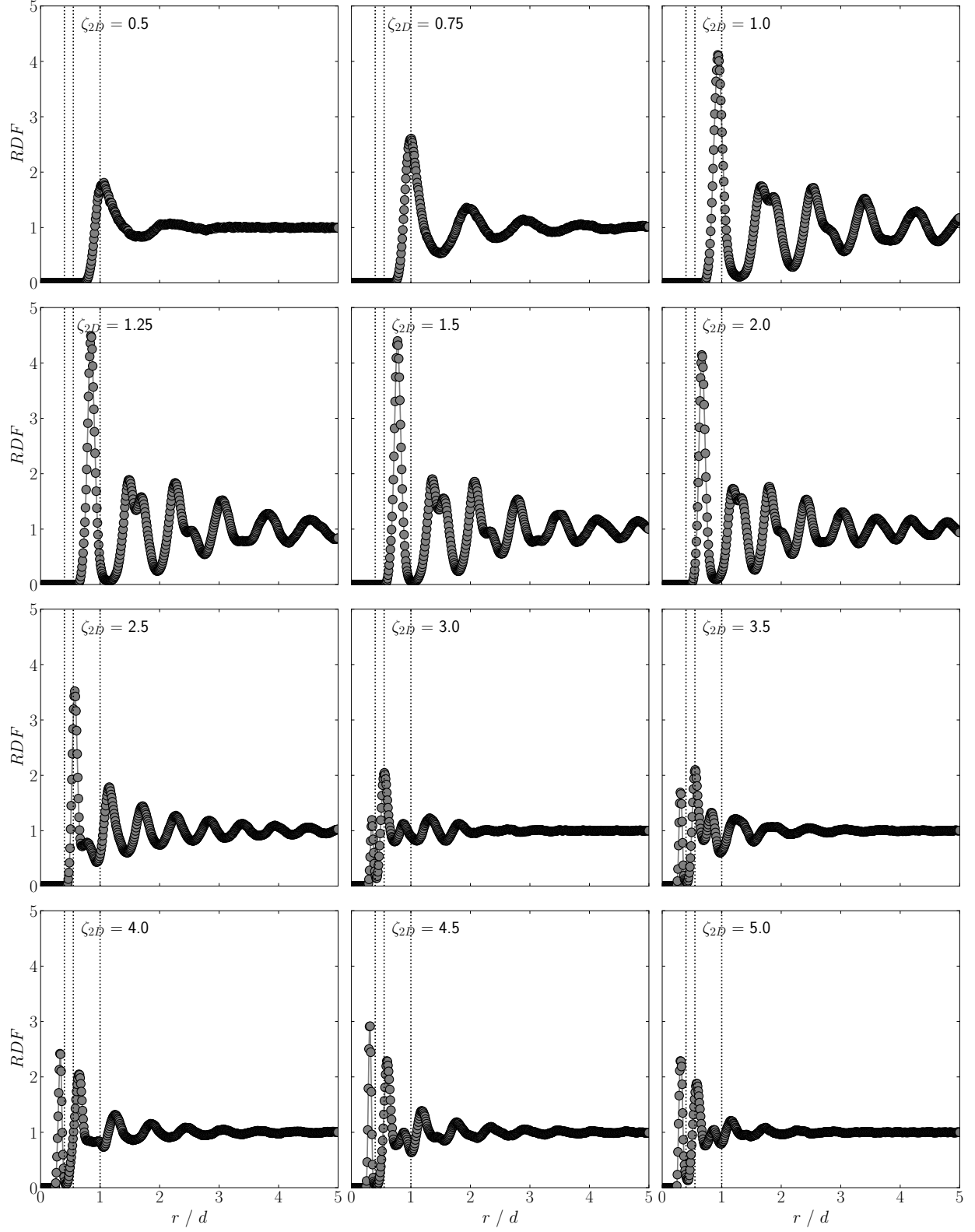


FIG. S1. Radial distribution functions for the square shoulder multi-Hertzian model using the reference parameters $Y = 200k_B T/d^2$, $Y_{dens} = 300k_B T/d^2$ and $d_{core} = 0.3d$ and varying generalized area fractions. Dotted lines mark the position of d_{core} , d_{out} and d .

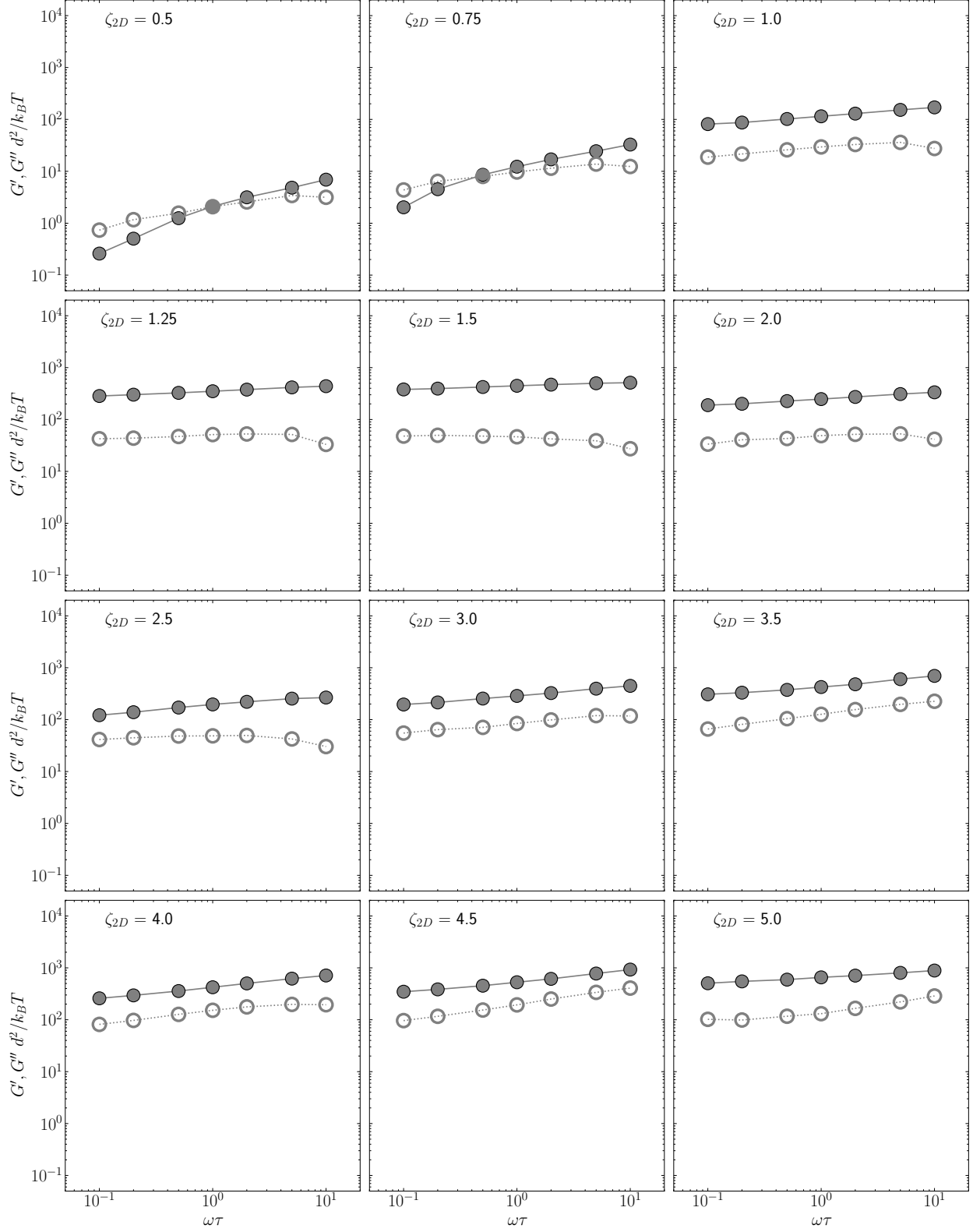


FIG. S2. Storage (full symbols) and loss modulus (empty symbols) for the square shoulder multi-Hertzian model as a function of shear rate using the reference parameters $Y = 200k_B T/d^2$, $Y_{dens} = 300k_B T/d^2$ and $d_{core} = 0.3d$ and varying generalized area fractions. The shear strain is set to $\gamma_0 = 0.03$

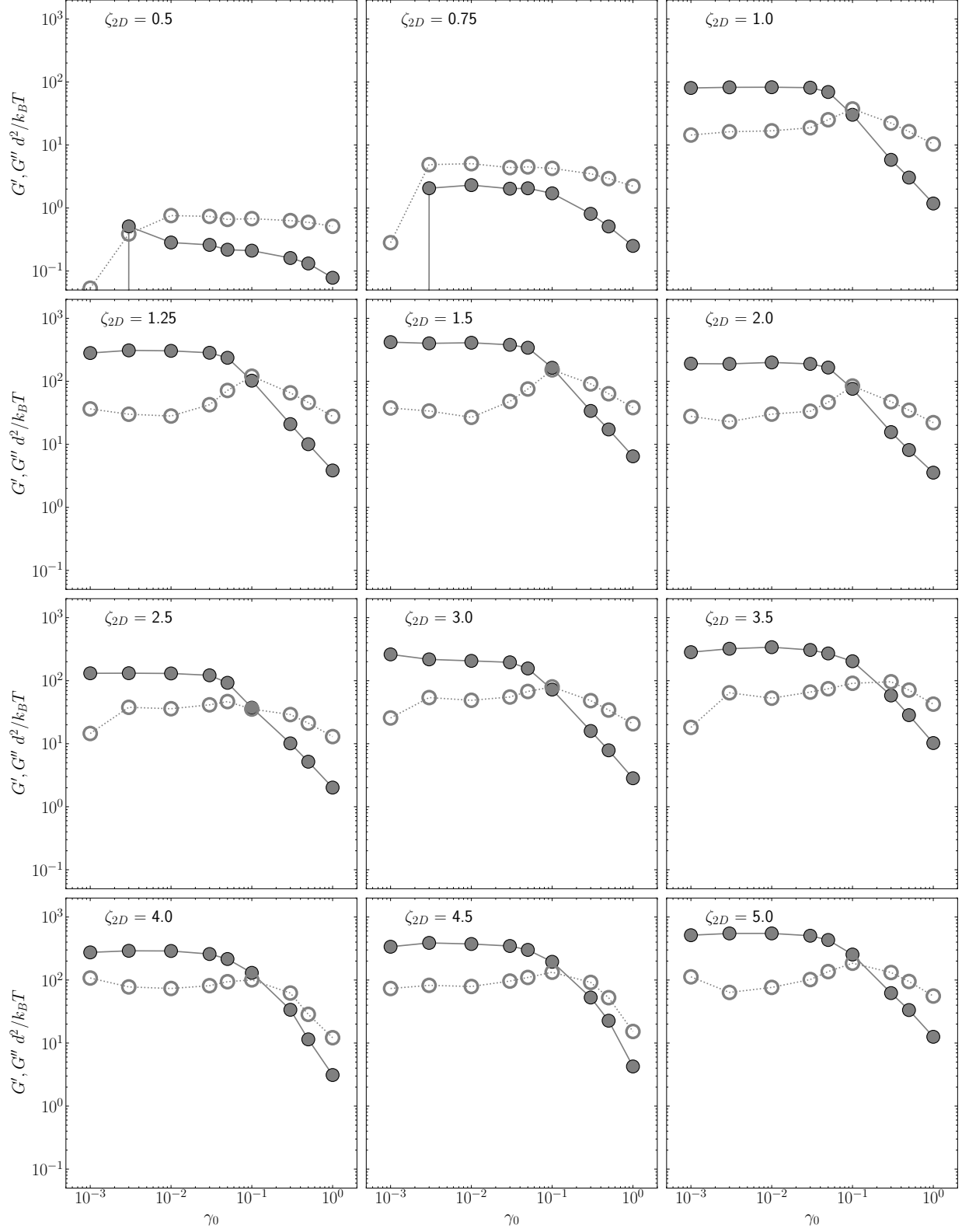


FIG. S3. Storage (full symbols) and loss modulus (empty symbols) for the square shoulder multi-Hertzian model as a function of strain amplitude using the reference parameters $Y = 200k_B T/d^2$, $Y_{dens} = 300k_B T/d^2$ and $d_{core} = 0.3d$ and varying generalized area fractions. The shear rate is set to $\omega\tau = 0.1$.