Brambilla *et al.* Reply to a Comment by J. Reinhardt *et al.* on "Probing the equilibrium dynamics of colloidal hard spheres above the mode-coupling glass transition"

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G. Brambilla et al. Reply to a Comment by J. Reinhardt et al. questioning the existence of equilibrium dynamics above the critical volume fraction of colloidal glassy hard spheres predicted by mode coupling theory.

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Brambilla et al. Reply: Reinhardt et al. [1] (RWF) use mode-coupling theory (MCT) to analyze a subset of our data [2] and question our claim that dense colloidal hard spheres enter at large volume fraction φ a dynamical regime not described by MCT. To reach this conclusion, RWF fit intermediate scattering functions (ISFs) obtained by light scattering to the outcome of MCT calculations for a monodisperse system of hard spheres. By freely adjusting the short-time diffusion coefficient D_s , and w, the parameter fixing the relative contribution of self and collective dynamics to the signal, they reproduce well the short-time decay of the data to a plateau. More crucially, to reproduce also the long-time decay, RWF need to adjust, for each experimental volume fraction φ considered, the volume fraction φ^{mct} of the corresponding theoretical curve. Since the shape of the ISF does not change much with φ , this analysis is nearly equivalent to adjusting the typical relaxation time $\tau_{\alpha}(\varphi)$, which we had done more simply by fitting the data to a stretched exponential form [2].

RWF's MCT analysis differs from ours when they then estimate the location, φ_c , of what we claim is an avoided MCT transition. If MCT predictions were an appropriat representation of our data, the fitted $\varphi^{mct}(\varphi)$ should be a linear function of φ , with the critical density φ_c estimated from $\varphi^{mct}(\varphi_c) = \varphi_c^{mct}$, with $\varphi_c^{mct} = 0.5159$. RWF obtain $\varphi_c = 0.595$, although deviations from linearity are evident in their Fig. 1b. Indeed, we find that the value of φ_c determined according to this procedure decreases systematically from 0.595 to 0.590 when the upper limit of the fitting interval varies from $\varphi = 0.5908$ to $\varphi = 0.5852$, indicating that the relation $\varphi^{mct}(\varphi)$ is not linear. In the absence of an unambiguous criterium for selecting the 'best' φ_c from RWF analysis, it is mandatory to compare the experimental $\tau_{\alpha}(\varphi)$ to the MCT prediction, $\tau_{\alpha} \sim (\varphi_c - \varphi)^{-\gamma}$. In Fig. 1a we show that with the values $\varphi_c = 0.595$ and $\gamma = 2.46$ obtained by RWF, the fit deviates from the data in a systematic manner for all φ . Thus, RWF's MCT analysis reproduces experimental



FIG. 1: (Color online) a): Comparison of the experimental decay time of the ISF, τ_{α} , to that predicted by a MCT fit, $\tau_{\alpha,\text{fit}}$. Systematic deviations are observed using RWF values, both when considering the full set of data (triangles) or the subset analyzed in [1] (circles), while a genuine MCT regime exists in our analysis (crosses). b): τ_{α} vs. $(\varphi_c - \varphi)^{-1}$, for various choices of φ_c with critical law fits to the data (lines), with an exponent γ shown in labels. Crosses correspond to $\varphi_c = 0.59$, $\gamma = 2.6$ as in [2], while solid triangles correspond to $\varphi_c = 0.595$, but with $\gamma = 3.4$, inconsistently with [1].

ISFs but fails to accurately determine φ_c .

In Fig. 1b, we show a log-log plot of τ_{α} vs. $(\varphi_c - \varphi)^{-1}$, where the MCT critical law becomes a straight line of slope γ , thus allowing for a more stringent test of an MCT description. We find again that an absolute determination of φ_c is ambiguous as γ and φ_c are correlated fitting parameter evolving from ($\gamma = 2, \varphi_c = 0.585$) to ($\gamma = 6.4, \varphi_c = 0.605$). In particular, we determine $\gamma = 3.4 \pm 0.1$ for the RWF preferred value $\varphi_c = 0.595$, while they advocate $\gamma = 2.46$. In our work [2] we had used Fig. 1b to determine the best pair (φ_c, γ) that fits

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our data. We imposed $\gamma = 2.6$, as obtained from MCT theoretical calculations (the precise value depends of the specific approximation used in the theory) and deduced $\varphi_c = 0.59$. As shown in Fig. 1a this choice opens a genuine "MCT regime", which is absent in RWF's analysis. We are then left with ISFs fully decaying to zero for *seven* samples above φ_c , with significant

- deviations of τ_{α} with respect to the divergence predicted by MCT [2, 3]. This motivated us to interpret these significant deviations from MCT predictions as the observation of a different, activated dynamical behavior entered by colloidal hard spheres above the divergence predicted by MCT.
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- [2] G. Brambilla et al., Phys. Rev. Lett. 102, 085703 (2009).
- [3] D. El Masri et al., J. Stat. Mech. P07015 (2009).