Brambilla *et al.* Reply: van Megen and Williams (vMW) question [1] our recent claim [2] that dense colloidal hard spheres enter at a large volume fraction φ, a dynamical regime not observed in earlier work [3] and not described by the mode-coupling theory (MCT) of the glass transition. They claim that our results are in contradiction to theirs and suggest that this discrepancy is due to differences in particle size polydispersity.

As shown in Fig. 1, the relative polydispersity of our sample is $\sigma = 12.2\%$. In this range of σ, MCT predicts [4], and our simulations reveal [5], no significant effect due to differential localization of large and small particles, which thus cannot account for our data, contrary to vMW's suggestion.

A second explanation suggested by vMW is that a moderate polydispersity shifts the glass transition to a larger volume fraction, implying that a nonergodic sample might become ergodic if σ increases at constant φ. We have considered this effect. Our simulations [5] show that the effect is quantitatively modest since, for instance, the position of the fitted MCT divergence, φ_c, shifts merely by 0.002 when σ changes from 6 to 11.5% [5]. Taking into account this φ_c shift and uncertainties related to volume fraction determination [3,5], our data are in fact fully consistent with those of Refs. [3,6] up to $\varphi \lesssim \varphi_c$.

However, unlike previous work, we have been able to detect ergodic behavior for samples that have volume fractions above our fitted $\varphi_c \simeq 0.590$, and have discovered that near φ_c, an MCT description of the data breaks down. Since we have allowed φ_c to vary to take polydispersity effects into account, our data cannot be reconciled with MCT in this regime. Indeed, deviations from an algebraic MCT description can only be cured at the expense of using unphysical values of the critical parameters. For example, by imposing $\varphi_c = 0.60$ (instead of 0.59 as in Ref. [2]), we find that the critical exponent γ in the fitted MCT divergence is as high as $\gamma = 4.5$; for $\varphi_c = 0.605$, the exponent is even higher, $\gamma = 6.8$. We made similar observations in our two simulated polydisperse hard sphere models. If these results were solely due to polydispersity, as claimed by vMW, it should be possible to obtain experimental and numerical results with less polydisperse samples, say $\sigma \lesssim 10\%$, that would cover a range of relaxation times comparable to that in our work, but still be fully compatible with MCT. To our knowledge, evidence supporting this scenario is lacking.

Finally, vMW criticize our statement that this new dynamic regime had not been detected in Ref. [3] because crystallization intervened. Indeed, crystallization is not mentioned as an issue in [3], although it did intervene in [6], where a sample with $\sigma \approx 4\%$ was studied. vMW emphasize that the samples with $\varphi > \varphi_c$ in Ref. [3] are not ergodic, even if a larger time window is used: they mention a more recent work [7] where the nonergodic aging dynamics of a sample with $\varphi = \varphi_c + 0.01$ is studied over 5 days. From the fit of the relaxation time $\tau_\alpha(\varphi)$ discussed in [2], we estimate that τ_α grows by a factor $\sim$500 when φ increases from φ_c to $\varphi_c + 0.01$. Assuming a similar behavior for the sample studied in [7], no equilibration is to be expected before several hundreds of days, much longer than the largest waiting time in that work. Similar arguments apply to the sample at $\varphi = 0.583$ in [3].

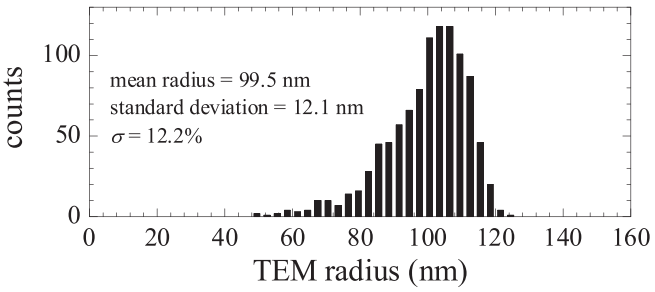

FIG. 1. Particle size distribution as obtained from a sample of 1000 particles imaged by TEM. The average radius measured by TEM is close to the hydrodynamic radius measured by DLS, $a \approx 105$ nm. The particle size reported in Ref. [2] was somehow higher because the solvent viscosity had been underestimated.

To conclude, our data show no discrepancy with earlier work, but explore a broader dynamical range, including an activated regime that has not been accessed before.

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