

long-ranged electrostatic repulsions [Figs. 1(b), (e), and (h)]. We thus term this state a ‘Wigner glassy state’. At low  $\phi$  and higher  $c_p$ , we see the formation of clusters and term this state a ‘cluster glassy state’ [Figs. 1(a) and (i)]. Meanwhile, increasing both  $c_p$  and  $\phi$  results in a gel which we define through percolation [Figs. 1(d) and (j)], and appears dynamically arrested [Fig. 2(a)]. A typical state diagram for a colloidal system without electrostatic repulsions is schematically shown in Fig. 1(g). We see striking differences in the structure of the state diagram between systems with and without electrostatic repulsions (see also appendix). With electrostatics the ergodic region is very narrow, around  $\phi \sim 0$ , and the glassy states dominate the state diagram. There are three glassy states with very different structures: Wigner and cluster glassy states and a gel state. We also found three types of transitions between these states accompanied by local structural changes.

Before discussing the structure of these three states in more detail, let us consider the dynamics. Mean squared displacement (MSD) measurements, in which the colloids are tracked in two dimensions (2D), are shown in Fig. 2. What is clear is that the (ergodic) fluid we see at low  $\phi$  appears to exhibit diffusive behavior, and the gel appears dynamically arrested, within the accuracy to which we can track the particles (100 nm or  $\sigma/20$ ). The other states show extensive non-diffusive behavior, yet they do not reach a clear plateau on the experimental timescale although we track the particles for up to 20 hours for the cluster glassy state and up to 2 hours for the Wigner glassy state. We are limited in particle tracking at long times due to particle loss (particles diffusing out of our volume), bleaching and drift, along with residual sedimentation. We estimate the cage size as the width of the first and second peaks in  $g(r)$  for the Wigner and cluster glassy states respectively [see Fig. 3(a)]. In the case of the gel the cage size is taken as the bond length,  $0.19\sigma$ . Concerning the cluster glassy state, similar sub-diffusive behavior has recently been seen in computer simulation [19]. We note that such behavior is expected from the fact that clusters can rotate as rigid bodies [see Fig. 2(b)]. This strong decoupling between translational and rotational motion may be unique to a cluster glassy state. In the case of the Wigner glassy state, based upon simulation work in a similar system [26], we believe that our experimental timescales limit full access to relaxation phenomena. In any case, the cage size is not reached [see Fig. 2(a)]. The soft nature of the interactions is another factor which prolongs the sub-diffusive regime and thus makes a rigorous confirmation of nonergodicity very challenging. The non-diffusive behavior is suggestive of the glassiness of these states. Since we do not find a clear plateau