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Nonperturbative effects of attraction on dynamical behaviors of glass-forming liquids*

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We investigate systematically the effects of the inter-particle attraction on the structure and dynamical behaviors of glass-forming liquids via molecular dynamics simulations. We find that the inter-particle attraction does not influence the structure, but greatly affects the dynamics and dynamical heterogeneity of the system. After the system changes from a purely repulsive glass-forming liquid to an attractive one, the dynamics slows down and the dynamical heterogeneity becomes greater, which is found interestingly to be associated with larger cooperative rearrangement regions (CRRs). Additionally, the structures of CRRs are observed to be compact in attractive glass-forming liquids but string-like in purely repulsive ones. Our findings constitute an important contribution to the ongoing study of the role of attractions in properties of glasses and glass-forming liquids.

Keywords: attractive glass, dynamics, dynamical heterogeneity, cooperative rearrangement regions (CRRs)

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1. Introduction

Dynamical heterogeneity is considered to be one of the key features of structural relaxation in supercooled liquids and glasses.[1–3] As the temperature of a glass-forming liquid is rapidly lowered, the motion of particles within the liquid becomes spatially and temporally heterogeneous.[2,4–6] While the overall motions within the liquid slow down, particles in some regions exhibit faster dynamics than the rest, and over time these mobile regions can appear and disappear throughout the system.[7] Additionally, particles within the mobile regions are found to move cooperatively, forming spatially extended clusters and strings.[8] For a purely repulsive hard-sphere colloid glassy system, dynamical heterogeneities have been directly observed in experiments by Kasper, Marcus, Kegel, and Weeks.[9–12] They found a non-Gaussian distribution of particle displacements, cage trapping and cage rearrangement motions which are spatially heterogeneous, with groups of particles exhibiting string-like motions. Interestingly, Zhang *et al.*[13] found that dynamical heterogeneities are different between glass-forming liquids composed of particles with attractive interactions versus purely repulsive interactions. Specifically, compared to purely repulsive glass-forming liquids, the attractive glass-forming liquids' dynamics is found to be much more heterogeneous over a wider range of time and length scales, and the associated cooperative rearrangement regions (CRRs) involve more particles. Additionally, the CRRs are observed to be compact in the attractive glass-forming liquids but string-like in the purely repulsive ones. A simulation study by Berthier and Tarjus[14] showed that the dynamics of a standard Lennard–Jones liquid with attraction and that of its purely repulsive counterpart (the well-known Weeks–Chandler–Andersen (WCA)[15] liquid) are quantitatively and qualitatively different. This indicates that the attractive interaction affects the dynamics, which is at variance with the traditional perturbation theory and hence challenges that theory. According to the traditional perturbation theory, the attractive interaction can be treated as a mere cohesive background amenable to perturbative treatment, and hence people for a long time assumed that the attraction affects neither the structure nor the dynamics.[14,16–18] Looking through the literature, so many experiments were performed in a wide variety of colloid and polymer systems concerning the effects of short-range attractive interactions.[13,19–21] Also, there have been many computer simulation studies focusing on properties of glass-forming systems with short-range attractive potentials including, e.g., square well and Asakwa–Oosawa

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potentials.[22–24] Furthermore, it is known that attraction sensitively affects the phase behavior of the attractive system.[25,26] Recent work by Tong and coworkers[27] suggests that attractions affect the structures at the many-body level, which refreshes people's knowledge of the role of attractions in structures in glass-forming liquids. To the best of our knowledge, nearly all previous studies addressed mainly the qualitative difference in two glass-forming systems: one dominated by repulsion (with structural arrest due to caging) and the other by attraction (with structural arrest due to bonding). However, the effects of attraction with different range on the dynamics and dynamical heterogeneities have never been systemically studied.

In this paper, we probe the effects of attractions on the structures and dynamics in the standard model Lennard–Jones glass-forming liquids where the attraction can vary from zero to one by tuning the interaction cutoffs. We find that particles in systems with long-range attractive potentials form more long-lived bonds with their neighbors and exhibit slower dynamics when compared to the system with purely repulsive interactions. Moreover, the dynamics in the attractive systems becomes more heterogeneous and can be associated with larger CRRs. We also find that the CRRs form string-like structures in the purely repulsive systems but compact cluster-like structures in the attractive ones.

2. Simulation details

Our molecular dynamics simulation is performed in a square box with particles interacting via the standard Lennard–Jones potential. The simulation system is composed of a binary mixture of 65% large (A) and 35% small (B) particles with a very strong A–B attraction, which guarantees that it is less prone to crystallization in two dimensions as suggested in Ref. [28]. The total particle number is 1000 and each particle has the same mass m. The inter-particle potential between α and β, with $\alpha, \beta = \mathrm{A}, \mathrm{B}$ is given by

$$u_{\alpha\beta} = \begin{cases} \varepsilon_{\alpha\beta}\left[\left(\dfrac{\sigma_{\alpha\beta}}{r}\right)^{12} - \left(\dfrac{\sigma_{\alpha\beta}}{r}\right)^{6}\right], & \text{for } r \leqslant r_{\mathrm{c}}, \\ 0, & \text{otherwise}, \end{cases}$$

where r is the separation between two particles. α, β, ε, and σ depend on the types of particles under consideration: $\varepsilon_{\mathrm{AB}} = 1.5\varepsilon_{\mathrm{AA}}$, $\varepsilon_{\mathrm{BB}} = 0.5\varepsilon_{\mathrm{AA}}$, $\sigma_{\mathrm{AB}} = 0.8\sigma_{\mathrm{AA}}$, and $\sigma_{\mathrm{BB}} = 0.88\sigma_{\mathrm{AA}}$.[29] $\varepsilon_{\mathrm{AA}}$, σ_{AA}, and m are the unity of energy, length, and mass, respectively. Therefore, the density $\rho = 1.2$ in this study is given in units of $N\sigma_{\mathrm{AA}}^3/L^3$ (L is the scale of the simulation square). We use the traditional standard "truncated and shifted potential" (SP) which sets a cutoff distance to tune the attractive part of the potential. The range of interaction is controlled by the SP cutoff distance, denoted by r_{c}. Specifically, $r_{\mathrm{c}} = 2^{1/6}\sigma$ is equal to the position of the minimum of $u_{\alpha\beta}$ for the WCA model with purely repulsive interactions and the standard LJ model corresponds to the cutoff $r_{\mathrm{c}} = 2.5\sigma$. Therefore, systems with greater values of r_{c} but not less than $2^{1/6}\sigma$ have larger attractive interactions. We run (Nose–Hoover's NVT ensemble)-simulations at $r_{\mathrm{c}} = 1.12\sigma, 1.2\sigma, 1.4\sigma, 1.7\sigma, 2.0\sigma, 2.5\sigma$, respectively. At each state point, the system is first equilibrated for at least $100\tau_\alpha$ (τ_α is the structural relaxation time) before collecting data. In order to obtain reliable results and improve the statistics, we perform a production run for at least $500\tau_\alpha$ and 5 independent runs for each r_{c}. Moreover, we have checked that there are no finite size effects by getting qualitatively similar results in small systems having 1000 particles and large systems (up to 10^5 particles).

Here, we use A particles to analyze the overall dynamics of the systems, and the (few and small) B particles are to a large extent slaves of the structure set by the A particles.

3. Results and discussion

3.1. Static structure

We first discuss the results of the static pair correlations[15,24,30] $g(r)$ for the systems with different inter-particle attractions. Clearly, there are no obvious differences between the structures of the six systems with vastly different cutoff distances r_{c}, as reflected by the same $g(r)$ curves for these systems, see Fig. 1. In addition, we find no evidence of crystallization for all the systems examined. Therefore, attraction has negligible effects on static structures of glass-forming liquids, which is consistent with previous results.[15,24]

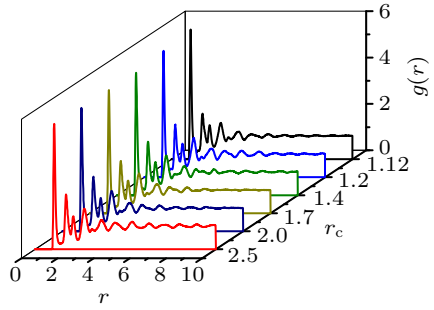

Fig. 1. Static pair correlation functions $g(r)$ as a function of r with different cutoffs for A particles at temperature $T = 0.35$ and density $\rho = 1.2$. black: $r_{\mathrm{c}} = 1.12\sigma_{\mathrm{AA}}$; blue: $r_{\mathrm{c}} = 1.2\sigma_{\mathrm{AA}}$; olive: $r_{\mathrm{c}} = 1.4\sigma_{\mathrm{AA}}$; dark yellow: $r_{\mathrm{c}} = 1.7\sigma_{\mathrm{AA}}$; navy: $r_{\mathrm{c}} = 2.0\sigma_{\mathrm{AA}}$; red: $r_{\mathrm{c}} = 2.5\sigma_{\mathrm{AA}}$.

3.2. Dynamics

Turning to the dynamics, we study how the motion of particles changes as the attraction increases approaching the glass transition. We present first the time dependence of the mean-squared displacement (MSD) for A particles. Figure 2(a) shows that as the attraction increases, particles move more slowly, as expected. All the MSD plots show a characteristic "cage trapping" plateau initially, because the particles

are "caged" by their nearest-neighbor particles.[31–36] Then the MSD curves go up, which is due to rearrangements of the cages, allowing the particles to move to a new location. Notice that for the WCA cutoff ($r_c = 1.12\sigma_{AA}$) with no attraction, the plateau is higher in height but shorter in time if compared with those in the attractive glass-forming systems with ($r_c > 1.12\sigma_{AA}$). Moreover, as the attractive interaction increases (r_c from $1.12\sigma_{AA}$ to $1.7\sigma_{AA}$), the plateau gradually descends, indicating that the cage becomes tight and the displacements of particles decrease drastically in systems with stronger attractions. But for $r_c > 1.7\sigma_{AA}$, there are no visible differences, which is consistent with the results in the viscous regime.[37] This is not surprising in view of the fact that the interactions are very small at such large distances. Therefore, our observations suggest that the dynamics almost does not change when the cutoff distance of potentials changes from $1.7\sigma_{AA}$ to $2.5\sigma_{AA}$. This also implies that it is accurate enough to take $r_c = 1.7\sigma_{AA}$ instead of the conventional $r_c = 2.5\sigma_{AA}$ when studying the dynamical properties in a standard Lenard–Jones potential in the future.

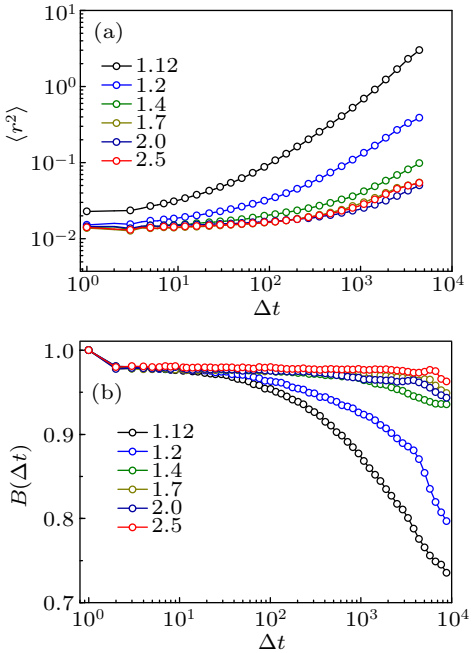

Fig. 2. (a) Log–log plot of the mean-squared displacement (MSD) of the A particles as a function of time in the KABLJ systems with different values of r_c ($T = 0.35$, $\rho = 1.2$). The black: $r_c = 1.12\sigma_{AA}$ (purely repulsive); blue: $r_c = 1.2\sigma_{AA}$; olive: $r_c = 1.4\sigma_{AA}$; dark yellow: $r_c = 1.7\sigma_{AA}$; navy: $r_c = 2.0\sigma_{AA}$; red: $r_c = 2.5\sigma_{AA}$. There are no visible differences for $r_c > 1.7\sigma_{AA}$. (b) A semi-log plot of the persistent bond parameter $B(\Delta t)$ versus lag time Δt for A particles.

The conjecture that the slower dynamics of attractive glass-forming liquids is due to the long-lived nearest-neighbor bonds, has been directly tested in the real-space colloidal experiments.[13] Here, we calculate the persistent bond parameter $B(\Delta t)$[38] which represents the fraction of nearest-neighbor bonds within Δt, identified by Delaunay triangulation, that remain unbroken across the interval $t \to t + \Delta t$ (Fig. 2(b)). We can observe that particles interacting via stronger attractive forces become progressively more trapped in potential wells, until quasi-permanent bonds are formed at the attraction-driven glass transition,[24,39,40] which impedes rearrangements and slows down particle dynamics, hence leading to smaller MSDs as shown in Fig. 2(a).

Particles involved in a cage rearrangement event move significant distances compared to when they are caged, and the distribution of displacements is unusually broad on the time scale of the rearrangement.[35,36] We quantify the distribution by calculating the non-Gaussian parameter $\alpha_2(\Delta t)$, which is defined as

$$\alpha_2(\Delta t) = \frac{\langle \Delta r^4 \rangle}{2\langle \Delta r^2 \rangle} - 1.$$

Here, $\Delta r = r(t + \Delta t) - r(t)$ denotes the displacement of particles for a time lag Δt, and the angle brackets indicate an average over particles and time. If the distribution of displacements Δr is Gaussian, then $\alpha_2 = 0$. However, when large displacements are more common than that would be expected from a Gaussian distribution, $\alpha_2 > 0$. Figure 3 shows the non-Gaussian parameter in systems with different potential cutoffs as examined in Figs. 1 and 2. Each curve in Fig. 3 has a peak which occurs at a time scale for cage rearrangement.[8,12] For the repulsive glass-forming liquid (black curve), the peak value of α_2 is very small and corresponds to a very short lag time Δt. With increasing attraction, the peak value of α_2 increases gradually. Similar to MSD and $B(\Delta t)$ shown in Fig. 2, the peak values of α_2 are nearly equal for $r_c > 1.7\sigma_{AA}$. These non-zero and larger values of α_2 in more attractive systems reflect that the motions of particles are more dynamically heterogeneous in the presence of the attractive forces, and the effect of attraction on dynamic heterogeneity will be discussed systematically in the following section.

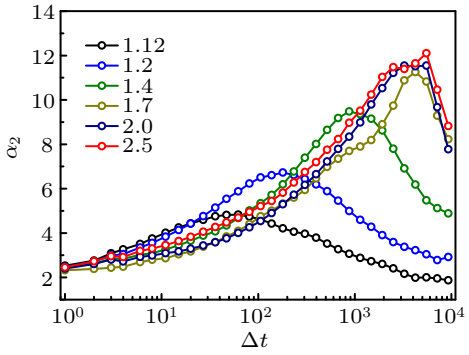

Fig. 3. A semi-log plot of the non-Gaussian parameter α_2 versus lag time Δt for A particles. With black indicating α_2 for $r_c = 1.12\sigma_{AA}$ (purely repusive); blue: $r_c = 1.2\sigma_{AA}$; olive: $r_c = 1.4\sigma_{AA}$; dark yellow: $r_c = 1.7\sigma_{AA}$; navy: $r_c = 2.0\sigma_{AA}$; red: $r_c = 2.5\sigma_{AA}$ (there are no visible differences for $r_c > 1.7\sigma_{AA}$).

3.3. Dynamical heterogeneity

To quantify dynamical heterogeneity in different attractive glass-forming liquids, the four-point susceptibility, χ_4, is

calculated as follows:[40–43]

$$\chi_4(a,\Delta t) = N(\langle Q_2(a,\Delta t)^2\rangle - \langle Q_2(a,\Delta t)\rangle^2).$$

The two-time self-correlation function Q_2 is defined as

$$Q_2(a,\Delta t) = \frac{1}{N}\sum_{i=1}^{N}\exp(-\Delta r_i^2/2a^2),$$

which is traditionally used for characterizing the dynamics of the glass-forming system. Here a is a preselected length scale to be probed, Δr_i^2 is the mean squared displacement of particle i in time lag Δt, and N is the total number of particles. The four-point susceptibility, χ_4, characterizes the temporal variance of particle dynamics and is directly related to the number of particles which participate in a correlated rearrangement.[38] Here we investigate the four-point susceptibility as a function of both the probing length scale a and time length Δt[41–43] for systems with different attractive interactions in Fig. 4. There is a maximum at ($\Delta t_{\max}$, $a_{\max}$) for all the systems examined. The maximum values indicate a characteristic timescale $\Delta t_{\max}$ and corresponding length scale $a_{\max}$, at which the dynamics is most heterogeneous. With the increase of the potential cutoff (or attraction), the maximum value of χ_4 gradually grows. The similar conclusion can also be drawn when plotting the maximum of χ_4 at each Δt, see Fig. 5. As shown in Fig. 5, for the repulsive system ($r_c = 1.12\sigma_{AA}$), the maximum value of χ_4 is ~ 5, whereas for the attractive system ($r_c = 2.5\sigma_{AA}$), the maximum is ~ 15. This observation implies that the number of particles participating in the primary cooperative rearrangements at the "maximum" length and time scale is about 3 times larger in the system composed of attractive particles than that composed of purely repulsive particles. We also find that for each system, the time scale where χ_4 shows the maximum is close to its α_2 counterpart shown in Fig. 3, which suggests that local cage rearrangements are the main contributor to the temporal fluctuations of dynamic heterogeneity.

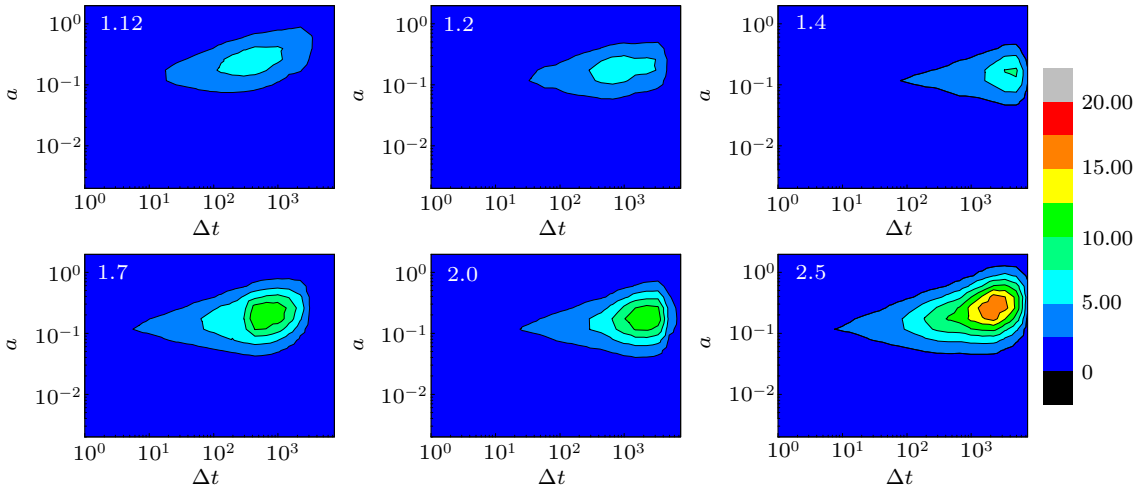

Fig. 4. Dependence of dynamical susceptibility, χ_4, in different attractive glass-forming systems on all probing length scales, a, and time scales, Δt.

Moreover, we find that $\Delta t_{\max}$ and $a_{\max}$ vary with r_c, and this dependence is shown in the inset of Fig. 5. $\Delta t_{\max}$ shows an increase with increasing r_c, which suggests that local rearrangements take longer time at higher r_c, in agreement with the extended MSD plateau observed at higher r_c in Fig. 2(a). In contrary to the r_c dependence of $\Delta t_{\max}$, we find that the length scale $a_{\max}$ decreases with increasing r_c, which suggests that the displacements for cage rearrangements are smaller as the attraction becomes stronger, in excellent agreement with prior work.[13] Therefore, for the purely repulsive glass-forming liquids, the local maxima of χ_4 occur within a relatively narrow range of a and Δt. Conversely, for the attractive glass-forming liquids, the local maxima of χ_4 span a wide range of length scales (a) and lag times (Δt) (Fig. 5). In other words, dynamics in the attractive glass-forming liquids is more heterogeneous over broader time and length scales than the purely repulsive ones.

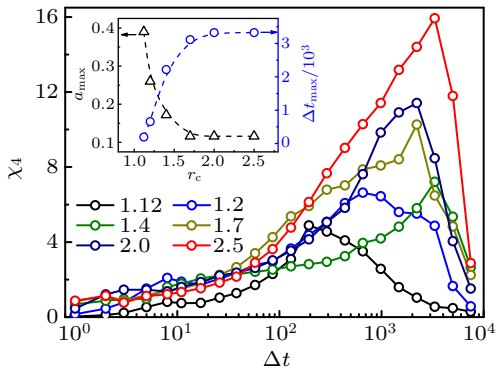

Fig. 5. Plot of the maximum of χ_4 at Δt in systems with different r_c. Inset: Plot of the length scale $a_{\max}$ (black) and the time scale $\Delta t_{\max}$ (blue) as a function of r_c.

To further quantify the size and shape of dynamic heterogeneities, we analyze the spatial distribution of particles participating in cooperative rearrangements. Following previous

study,[12] a CRR represents a group of highly mobile particles that are nearest neighbors. The nearest-neighbor pairings are identified using Delaunay triangulation. Particles with the 15% largest displacements over a given lag time Δt are defined as "mobile" particles. Δt is set to the same lag time that maximizes χ_4, i.e., the lag time over which dynamics is most heterogeneous.[38,41–43] Figure 6 shows six snapshots for repulsive and attractive glass-forming systems, wherein fast moving or mobile particles are depicted as large red spheres with arrows indicating their direction of motion; all other particles are shown as small black dots. In the purely repulsive system, the CRRs form string-like structures, consistent with results from previous work.[2,12,41,42,44–46] Interestingly, CRRs form more compact structures in glass-forming systems with longer attractions.

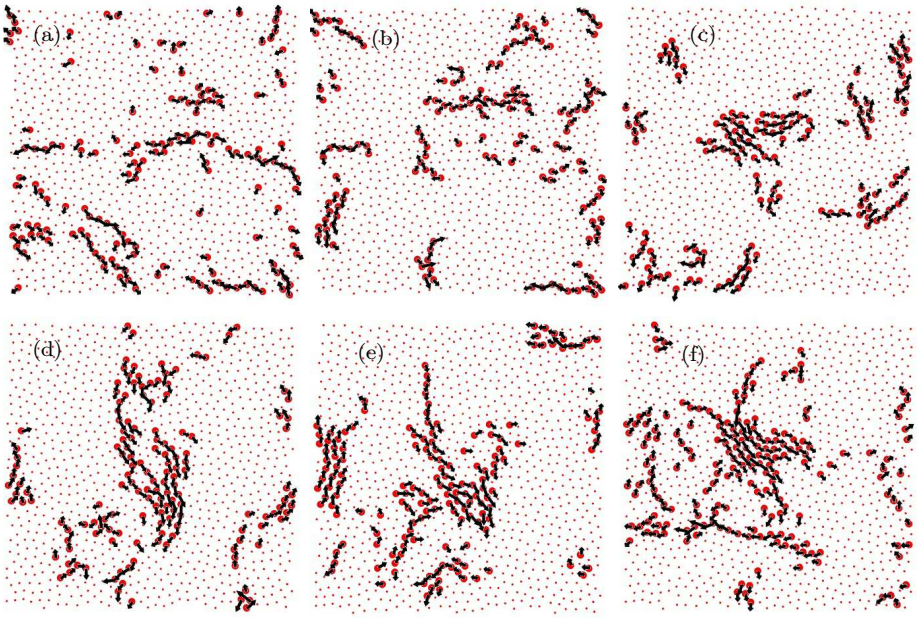

Fig. 6. Snapshots of cooperatively rearranging particles for (a) the purely repulsive WCA system; (b) $r_c = 1.2\sigma_{AA}$; (c) $r_c = 1.4\sigma_{AA}$; (d) $r_c = 1.7\sigma_{AA}$; (e) $r_c = 2.0\sigma_{AA}$, and (f) $r_c = 2.5\sigma_{AA}$. The large red spheres are drawn to scale and represent the 15% fastest particles. The rest of the particles are shown as small black dots, and reduced in size for clarity. Arrow indicates the direction of particle motion. The time interval between images used to generate the snapshots of CRRs was set to the value that maximized χ_4.

4. Conclusions

By examining the effect of the inter-particle attraction on the structure, dynamics, and dynamical heterogeneity in two-dimensional binary glass-forming liquids, we find that the attraction between particles has no influence on the structures but plays a nontrivial role in the dynamics and dynamical heterogeneity of the systems. After adding more attraction to a system, the dynamics shows a significant slowdown. We find that the slower dynamics in attractive systems results from the formation of more long-lived bonds between particles, which restricts further the particles to escape from their cages formed by the near neighbors. Moreover, a stronger dynamical heterogeneity is observed in systems with more attractive interactions, corresponding to the cooperative rearrangement regions with more particles involved. The topic on the role of attractions in controlling different properties of glass-forming liquids and glasses has attracted much attention[13,15,19–21,47–50] and has proven to be of importance to the understanding of glass transition. Our systematical study of attraction effects on the structure and dynamical properties in glass-forming liquids thus constitutes a necessary contribution to this topic.

References

[1] Wang Y J 2017 *Chin. Phys. B* **26** 014503
[2] Adam G and Gibbs J H 1965 *J. Chem. Phys.* **43** 139
[3] Qiao J C, Wang Q, Crespo D, Yang Y and Pelletier J M 2017 *Chin. Phys. B* **26** 016402
[4] Hempel E, Hempel G, Hensel A, Schick C and Donth E J 2000 *J. Phys. Chem. B* **104** 2460
[5] Richert R 2002 *J. Phys.: Condens. Matter* **14** R703
[6] Sillescu H 1999 *J. Non-Cryst. Solids* **243** 81
[7] Donth E 1996 *J. Polym. Sci. Part. B: Polym. Phys.* **34** 2881
[8] Donati C, Douglas J F, Kob W, Plimpton S J, Poole P H and Glotzer S C 1998 *Phys. Rev. Lett.* **80** 2338
[9] Poon W C K, Selfe J S, Robertson M B, Ilett S M, Pirie A D and Pusey P N 1993 *J. Phys. II* **3** 1075
[10] Marcus A H, Schofield J and Rice S A 1999 *Phys. Rev. E* **60** 5725
[11] Kegel W K and van B A 2000 *Science* **287** 290
[12] Weeks E R, Crocker J C, Levitt A C, Schofield A and Weitz D A 2000 *Science* **287** 627
[13] Zhang Z, Yunker P J, Habdas P and Yodh A G 2011 *Phys. Rev. Lett.* **107** 208303
[14] Berthier L and Tarjus G 2009 *Phys. Rev. Lett.* **103** 170601
[15] Weeks J D, Chandler D and Andersen H C 1971 *J. Chem. Phys.* **54** 5237
[16] Longuet-Higgins H C and Widom B 1964 *Mol. Phys.* **8** 549
[17] Widom B 1967 *Science* **157** 375
[18] Hansen J P and McDonald I R 1986 *Theory of Simple Liquids,* 2nd Edn. (London: Academic Press)
[19] Poon W C K, Selfe J S, Robertson M B, Ilett S M, Pirie A D and Pusey P N 1993 *J. Phys. II* **3** 1075
[20] Eckert T and Bartsch E 2003 *Faraday Discuss* **123** 51

[21] Pham K N, Egelhaaf S U, Pusey P N and Poon W C K 2004 *Phys. Rev. E* **69** 011503
[22] Sciortino F, Tartaglia P and Zaccarelli E 2003 *Phys. Rev. Lett.* **91** 268301
[23] Puertas A M, Fuchs M and Cates M E 2002 *Phys. Rev. Lett.* **88** 098301
[24] Zaccarelli E, Lowen H, Wessels P P F, Sciortino F, Tartaglia P and Likos C N 2004 *Phys. Rev. Lett.* **92** 225703
[25] Lekkerkerker H N W, Poon W C K, Pusey P N, Stroobants A and Warren P B 1992 *Europhys. Lett.* **20** 6
[26] Lekkerkerker H N W, Tejero C F and Daanoun A 1994 *Phys. Rev. Lett.* **73** 752
[27] Tong H, Tanaka H 1969 *Phys. Rev. Lett.* **22** 124
[28] Bruning R, St-Onge D A, Patterson S and Kob W 2009 *J. Phys.: Condens. Matter* **21** 035117
[29] Kob W and Andersen H C 1994 *Phys. Rev. Lett.* **73** 1376
[30] Chen K 2017 *Acta Phys. Sin.* **66** 178201 (in Chinese)
[31] Rabani E, Gezelter D J and Berne B J 1997 *J. Chem. Phys.* **107** 6867
[32] Doliwa B and Heuer A 1998 *Phys. Rev. Lett.* **80** 4915
[33] Kasper A, Bartsch E and Sillescu H 1998 *Langmuir* **14** 5004
[34] Weeks E R and Weitz D A 2002 *Chem. Phys.* **284** 361
[35] Weeks E R and Weitz D A 2002 *Phys. Rev. Lett.* **89** 095704
[36] Reis P M, Ingale R A and Shattuck M D 2007 *Phys. Rev. Lett.* **98** 188301
[37] Toxvaerd S and Dyre J C 2011 *J. Chem. Phys.* **135** 134501
[38] Abate A R and Durian D J 2007 *Phys. Rev. E* **76** 021306
[39] T V, P V, Cipelletti L, Segre P N and Weitz D A 2001 *Nature* **411** 772
[40] Foffi G, De M C, Sciortino F and Tartaglia P 2005 *Phys. Rev. Lett.* **94** 078301
[41] Berthier L, Biroli G, Bouchaud J P, Cipelletti L, Masri D E, Lhote D, Ladieu F and Pierno M 2005 *Science* **310** 1797
[42] Dauchot O, Marty G and Biroli G 2005 *Phys. Rev. Lett.* **95** 265701
[43] Lacevic N, Starr F W, Schorder T B and Glotzer S C 2003 *J. Chem. Phys.* **119** 7372
[44] Stevenson J D, Schmalian J and Wolynes P G 2006 *Nat. Phys.* **2** 268
[45] Ediger M D 2000 *Annu. Rev. Phys. Chem.* **51** 99
[46] Wang Y, Wang H G and Zhang Z X 2016 *Acta Phys. Sin.* **65** 178705 (in Chinese)
[47] Wang L and Xu N 2014 *Phys. Rev. Lett.* **112** 055701
[48] Sun Y L, Wang H G and Zhang Z X 2018 *Acta Phys. Sin.* **67** 106401 (in Chinese)
[49] Chattoraj J and Massimo P C M P 2020 *Phys. Rev. Lett.* **124** 028001
[50] Wang Y, Fang S, Xu N and Deng Y 2020 *Phys. Rev. Lett.* **124** 255501