Molecular-dynamics study of binary soft-sphere glasses:
Quench-rate effects and aging effects

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Molecular-dynamics simulations (MD) have been carried out on a soft-sphere model for binary alloys quenched into glassy states at different quench rates. The main purpose of the present work is to investigate slow relaxation phenomena of the quenched glassy states close to the glass transition by calculating both the static and dynamical structural changes upon aging (annealing) of them. For this purpose the MD simulations were performed for a time interval as long as an order of 2000τ, where τ is a microscopic time scale, equivalent to an order of the Einstein period, which is the characteristic period of oscillation of atoms. It is shown that the static properties exhibit neither significant changes during annealing of samples, nor changes for different samples. On the other hand, the dynamic properties show remarkable aging effects as well as sample-dependent behavior, meaning that the quenched glassy state cannot attain to an equilibrium state for the time scale of our simulations, due to dynamical slowing-down phenomena. The quenched sample is, however, shown to tend to a seemingly quasiequilibrium state after a sufficient annealing. Detailed discussions are made on both aging effects and sample-dependent behaviors of quenched glassy states, by paying particular attention to the behavior of the single-atom motion, mean-square displacement, and self-diffusion constant of the quenched glassy state. A non-Gaussian parameter is also calculated, which we propose as a good candidate to represent an order parameter of the glass transition.

I. INTRODUCTION

The dynamic properties of glassy states are currently of great interest, since recent theoretical developments, based on the mode-coupling techniques of the theory of liquids, have pointed to the essentially dynamical origin of the glass transition.[1-9] These theories suggested that the glass transition is signalled by the divergence of the structural relaxation time and a resulting nonergodic behavior characteristic of the glassy state. Several recent inelastic neutron-scattering measurements of the density fluctuation spectrum in supercooled ionic[10,11] or polymetric[12,13] liquids show evidence of the critical behavior predicted by mode-coupling theories. The sharp transition from ergodic to nonergodic behavior of the density autocorrelation function predicted by the simplest version of mode-coupling theory has also been observed in molecular-dynamics (MD) simulations of a supercooled Lennard-Jones liquid.[14] However, the sharp glass transition is smeared by activated processes which lead to residual diffusion of atoms in the glassy state and restore ergodicity on a sufficiently long time scale. This behavior may be accounted for by an improved version of the original mode-coupling theory or the generalized hydrodynamic theory which allows for coupling to current fluctuations.[8,9,15] Evidence for jumping motions of atoms is shown in recent MD simulations of simple binary soft-sphere alloys.[16]

The main concern of the present work is further detailed in investigations of such slow relaxation phenomena characteristic of the glass transition, focusing our attention on aging (annealing under different processes) effects and quench-rate effects. For this purpose, we study a glassy state, close to the glass transition, of the soft-sphere model for binary alloys using molecular-dynamics simulations.

In our previous studies on the pure (one-component) soft-sphere model,[17] we have found that the supercooled liquid always exhibits a tendency towards nucleation over a sufficiently long time interval, typically around and over an order of 10 000 time steps, equivalent to an order of 10τ. Nucleation is much more easily bypassed in quenched samples of binary soft spheres with two different core diameters.[16,18-22] Indeed, in laboratory experiments, certain metallic alloys can be formed into stable glassy states, while monovalent metals are more difficult to quench into glassy states. Our simulations for the binary soft-sphere model have shown no sign of a tendency towards crystallization of the quenched samples, thus allowing a detailed study of slow relaxations of samples near the glass transition.

Although the soft-sphere potential is not a very realistic model, because it contains no attractive force, it can be regarded as the simplest model retaining the essential features of glass-forming binary alloys. Therefore, it is very useful to use such a model for the study of the nature of glass transition.

In the present work, we have carried out the molecular-dynamics simulations for the binary soft-sphere mixtures, quenched into a glassy state close to the glass transition by using different quench processes, i.e., different quench rates to obtain various glass samples

with which sample-dependent properties are studied in detail. We consider a particular thermodynamic state near the glass transition of binary soft-sphere glasses [$\Gamma_{\text{eff}} = 1.6$, see Eq. (5)]. The simulation (annealing) for each quenched sample was carried out over typically 2000τ, where τ is a microscopic time scale defined by $\sqrt{m_1 \sigma_1^2 / \epsilon}$ (see Sec. II for the definition), equivalent to an order of the Einstein period, the characteristic period of the oscillation of atoms.

The paper is organized as follows. The model and simulations used in this work are briefly presented in Sec. II. The results of the simulations are given in Sec. III on static and dynamic properties of the glass, focusing our attention to their relaxation characteristics as well as sample dependent behaviors. Finally, in Sec. IV, brief discussions are made.

II. THE MODEL AND SIMULATIONS

The binary-alloy model which has been studied in the present, work is identical to that used in some earlier MD works.[16,18-22] We consider binary mixtures of N_1 atoms of mass m_1 and diameter σ_1 and N_2 atoms of mass m_2 and diameter σ_2 in a volume V. Soft-sphere pair interaction is assumed between atoms, that is,

$$v_{\alpha\beta}(r) = \epsilon \left[\frac{\sigma_{\alpha\beta}}{r} \right]^{12} , \qquad (1)$$

where $1 \leq \alpha, \beta \leq 2$ are species indices, ϵ the energy unit, and the diameters are assumed to be additive, i.e.,

$$\sigma_{\alpha\beta} = \tfrac{1}{2}(\sigma_\alpha + \sigma_\beta) . \qquad (2)$$

The scaling property of the soft-sphere model reduces independent thermodynamic parameters into only one dimensionless coupling constant Γ, as defined in Eq. (3), with given the core-size ratio σ_2/σ_1 and the number concentration of species 1, $x_1 = N_1/N$, instead of two independent variables, say temperature T and number density N/V, where N is the total number equal to $N_1 + N_2$;

$$\Gamma = \frac{N\sigma_1^3}{V} \left[\frac{\epsilon}{k_B T} \right]^{1/4} . \qquad (3)$$

We have shown in previous papers that the conformal solution theory (effective one-fluid approximation) works very well for binary soft-sphere mixtures with the effective diameter of the atoms σ_x, and the corresponding effective coupling constant Γ_{eff} given by the following equations, respectively

$$\sigma_x^3 = \sum_\alpha \sum_\beta x_\alpha x_\beta \sigma_{\alpha\beta}^3 . \qquad (4)$$

$$\Gamma_{\text{eff}} = \Gamma(\sigma_x / \sigma_1)^3 . \qquad (5)$$

The equation of state (compressibility factor $PV/Nk_B T$, versus Γ_{eff}) for binary soft spheres has been found to yield the curve very close to that of the pure soft-sphere model,[17] and the self-diffusion constant for the pure and binary soft-sphere model becomes a vanishingly small value around $\Gamma_{\text{eff}} = 1.56$.[22] The freezing point

of the pure soft-sphere model ($\Gamma_{\text{eff}} = \Gamma$) is found to occur at $\Gamma = 1.15$.

In the present work we study a binary soft-sphere mixture for the core-size ratio $\sigma_2/\sigma_1 = 1.2$ and for equimolar system ($x_1 = 0.5$). Using the constant-temperature MD techniques by the Nosé and Verlet algorithm, we have carried out MD simulations with $N = 500$ particles, the pair potentials, Eq. (1), being cut off over distances $r/\sigma_{\alpha\beta} = 3.1$, and mass ratio $m_2/m_1 = 2.0$. First, we simulated an equilibrium liquid of $\Gamma_{\text{eff}} = 0.8$, then, using the configuration at the final step of the run, this liquid was quenched into $\Gamma_{\text{eff}} = 1.6$, close to the glass transition, through different routes (quench rates) by changing the parameter Q of Nosé's constant-temperature MD techniques, where Q is a parameter (mass) relevant to a freedom of an external "particle" introduced so as to keep the temperature of the system constant. The temperature and potential energy variations during the quench processes in our simulations are plotted in Fig. 1. The corresponding quench rates are ranged from $\Delta T^*/(\Delta t^*) = 0.03$ to 15.47, where ΔT^* is the decrement of the temperature in the reduced units ($T^* = k_B T/\epsilon$) and Δt^* the time taken for that quench in the unit of $\tau(= \sqrt{m_1 \sigma_1^2 / \epsilon})$. It is easily seen that these computer quench rates are much faster than those allowed by usual experimental conditions, say 10^6 K/sec ($\Delta T^*/\Delta t^* \sim 10^{-8}$ for argonlike liquids). The quenched state was further simulated (annealed) for a long time interval at a fixed temperature to study the aging effects of the sample. In the next section the results of the analyses are given in detail.

III. RESULTS

A. Static properties

Table I summarizes the $PV/Nk_B T$ data obtained from the present MD simulations. These indicate that our simulations with different quenched samples or different annealing processes yield nearly the same value of $PV/Nk_B T$ within a few percent; that is, no significant differences are observed among them.

The three partial pair distribution functions $g_{\alpha\beta}(r)$ are shown in Fig. 2, where α and β are species indices; the splitting of the second peak is clearly seen, which has been discussed so far by many authors as a characteristic of glass materials.[17,18,23-25] The origin of this splitting may be more reasonably understood in terms of Voronoi or Delaunay tessellation:[26,27] As approaching the glass transition, icosahedral structures increase significantly, and geometrical shapes (bond length, bond angle, etc.) of such characteristic clusters of atoms should be responsible to the origin of the splitting of the second peak of the pair distribution function. However, such a picture does not seem to be well succeeded to explain the origin, nor whether this splitting is related to the glass transition itself.[26] Only a qualitative discussion has been made so far. Indeed, the second peak splitting of the pair distribution function has been observed even well before the glass transition for binary soft-sphere mixtures,[18] suggesting that such a splitting will not necessarily be related to the
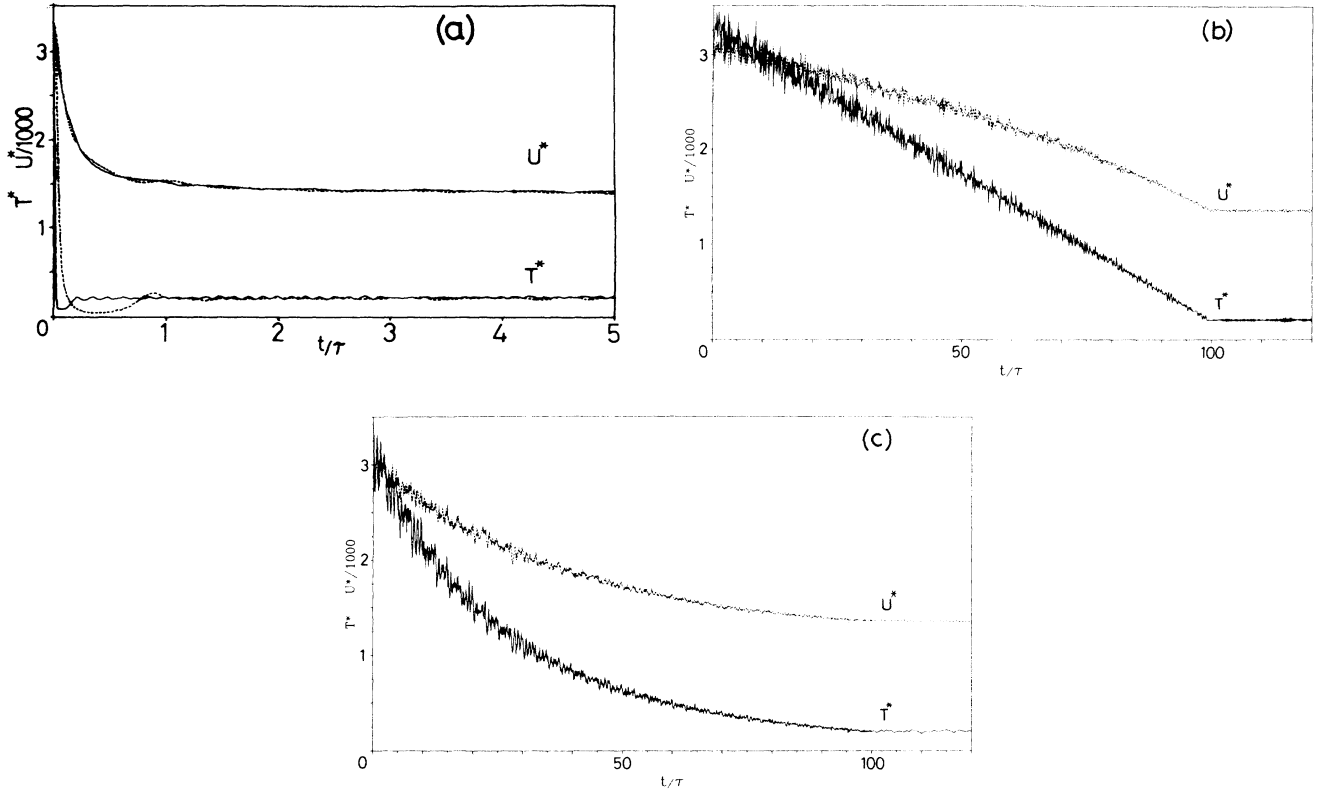
FIG. 1. Temperature and potential energy variations in molecular-dynamics (MD) simulations of quenching. The equilibrium liquid of $\Gamma_{eff}=0.8$ starts being quenched at $t=0$ towards $\Gamma_{eff}=1.6$ (flat portions) at different rates. (a) MD quenching for run 2 (dotted curve) and runs 3 and 4 (solid curve), where the upper curves indicate the total potential energy U^* in units of ϵ and the lower curves show the reduced temperature $T^*=k_B T/\epsilon$. (b) MD quenching for run 5. (c) MD quenching for run 6. Each solid curve is for the reduced temperature and the dotted curve for the total potential energy U^*. Each final quenched state ($\Gamma_{eff}=1.6$) was used as the initial state of further MD simulations for annealing in the respective runs. See also Table I.

characteristic property of the transition.

As seen from $g_{\alpha\beta}(r)$ in Fig. 2 and $S_{N-N}(k^*)$, the number-number structure factor,[28] in Fig. 3, it turns out that these functions show no significant change during annealing of the sample. This implies that main local-structures in the glassy state are not changed significantly upon aging of the sample. However, this does not mean

such local structures in the glassy state are absolutely stable, such that each atom is located at its local equilibrium position with thermal vibrations around it. It is shown that there exists large amplitude motions, such as concerted jump motions, that is, several atoms move their positions by an order of σ_1 for a very short time interval.[16]

TABLE I. Conditions used in our molecular-dynamics simulations and the results for the compressibility factor $PV/Nk_B T$ and reduced self-diffusion constant D_α^*. $T^*=k_B T/\epsilon$, $D_\alpha^* =(T^*)^{-5/12}\sigma_1^{-2}D_\alpha\tau$, and $\Gamma_{eff}=\Gamma(\sigma_x/\sigma_1)^3$ [see Eqs. (3)–(5)]. The time mesh of the simulations are shown by the column h in units of τ. The column R_Q is the quench rate, being defined by $R_Q=(T_1^* - T_2^*)/(\Delta t^*)$, where $T_1^* =3.300$ (for liquid) and $T_2^* =0.206$(for glass). That is, the initial state of the glass sample at T_2^* for annealing was prepared by quenching the liquid of run 1 at T_1^* over a time interval $\Delta t^*(=\Delta t/\tau)$. For runs 3 and 4 or runs 5 and 6, different values of Q were used. $(N\sigma_1^3/V)=0.8$ for all runs. Runs 2–4 are presented in Fig. 1(a), run 5 in Fig. 1(b), and run 6 in Fig. 1(c).

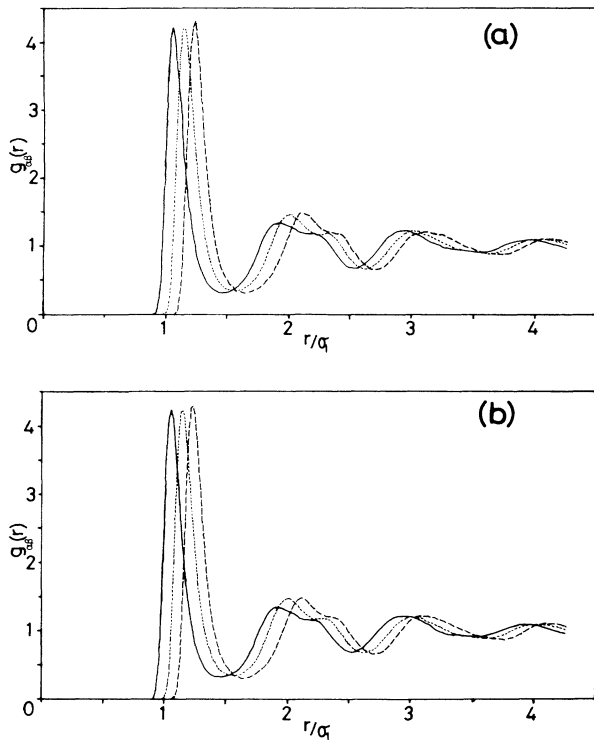| Run	T^*	Γ_{eff}	h	steps	$PV/Nk_B T$	D_1^*	D_2^*	R_Q
1	3.300	0.8	0.0025	4×10^4	8.477	1.11×10^{-1}	8.45×10^{-2}	
2	0.206	1.6	0.01	2×10^5	53.312	5.03×10^{-7}	4.78×10^{-7}	3.09
3	0.206	1.6	0.0025	6×10^5	53.034	1.10×10^{-5}	4.48×10^{-6}	15.47
4	0.206	1.6	0.01	1.6×10^5	53.183	2.68×10^{-5}	1.34×10^{-5}	15.47
5	0.206	1.6	0.0025	8×10^5	53.037	1.71×10^{-5}	9.53×10^{-6}	0.03
6	0.206	1.6	0.01	1.5×10^5	53.045	1.20×10^{-4}	4.09×10^{-5}	0.03

FIG. 2. Partial pair distribution functions $g_{\alpha\beta}(r)$ for run 2, (a) in the time window between 300τ and 400τ and (b) in the time window between 1800τ and 2000τ. Solid curve for $g_{11}(r)$, dotted curve for $g_{12}(r)$, and dashed curve for $g_{22}(r)$.
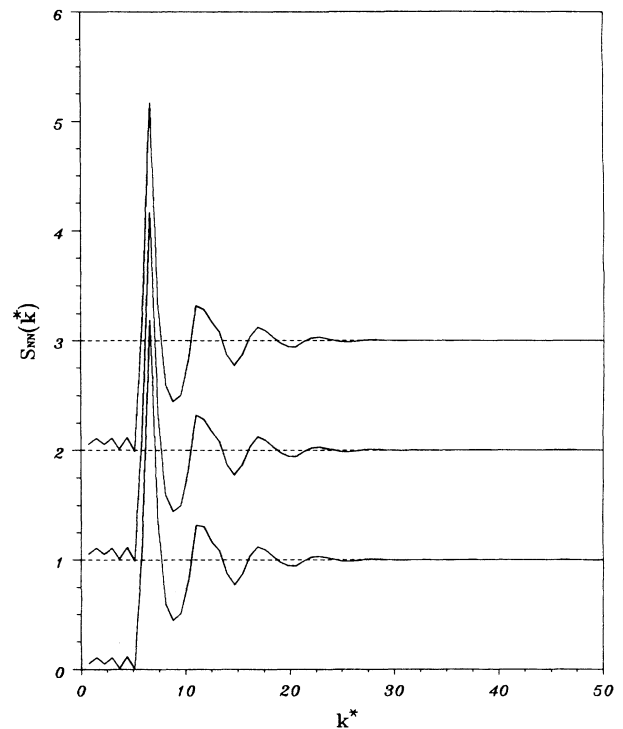

FIG. 3. The number-number structure factor $S_{N-N}(k^*)$, as calculated from the Fourier transform of $g_{\alpha\beta}(r)$ for run 2 in different time windows: $300 \le t/\tau \le 400$, $800 \le t/\tau \le 1000$, and $1800 \le t/\tau \le 2000$ from bottom to top. k^* is the scaled wave number, $k^* = \sigma_1 k$.

B. Dynamical properties

In Fig. 4 are shown mean-square displacements (MSD) averaged over atoms as a function of time, as measured from a subsequent series of the time origin ($t = 0$) indicated by the vertical lines. The time of abscissa begins with the final time step of the quenched glassy sample. It is clearly seen that the behavior of the MSD in an early time window such as $0 \le t/\tau \le 500$, which we call time region I, differs from that at the later time window of $500 \le t/\tau \le 2000$ (time region II). Dynamical fluctuations in the former time region are large and showing a clear tendency of decreasing for an increasing time, indicating the sample to be far from an equilibrium state, while in the latter time region they become small so as to be regarded as a quasi-equilibrium state. Separation of time into such regions for each run is rather properly made by monitoring the time variation of the MSD, as in Fig. 4, which may depend on the quench process used (the way of sample preparation). Figure 4 also shows that it takes a long time, say, an order of 500τ, for the quenched glassy state to reach the time region II. Figures 5 and 6 show MSD averaged over both atoms and initial times in two separate time regions. The MSD in the later time region ($500 \le t/\tau \le 1500$) is much smaller than that in the earlier time region ($100 \le t/\tau \le 500$), indicating significant aging effects of the sample.

The MSD shown in Figs. 5(b) and 6(b) for the time region II indicates the following three remarkable points (1) The slope of each curve at longer times, i.e., the (effective) self-diffusion constant (see the list of D_α^* in Table I), differs for each run. (2) The MSD at longer times is still curved, namely, the Einstein limit (t-linear
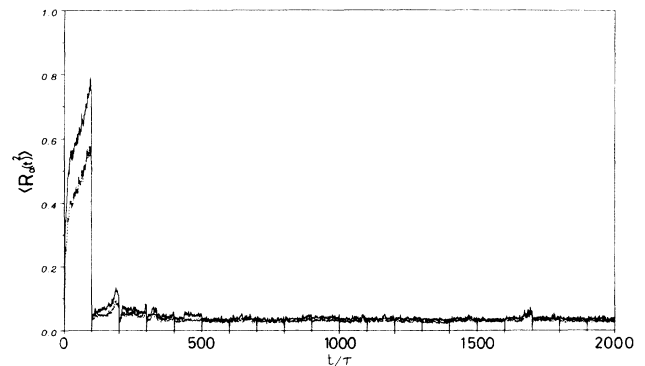

FIG. 4. Mean-square displacement (in units of σ_1^2) averaged over atoms, as measured from a subsequent series of the time origin (shown as vertical lines) for run 2. Most of the upper curve is for species 1, and most of the lower curve is for species 2.

regime) is not yet attained. Thus, the self-diffusion constant in glassy states is very hard to calculate as precisely as in fluids because of such an extremely slow relaxation. Then it is easily overestimated due to the negative curvature of the MSD in intermediate times. A similar "subdiffusive" behavior is clearly apparent in our simulations of the supercooled one-component soft-sphere fluid[17] and in a recent MD study of supercooled fluids of atoms interacting through a repulsive Yukawa potential.[29] (3) There is no simple systematic quench-rate dependence of the MSD or the self-diffusion constant. This behavior may simply be apparent due to insufficient time ranges of our simulations for the glassy state which essentially involves slow dynamical relaxations.

Dynamical slowing down of glassy states may be understood by considering that a possible phase space ac-
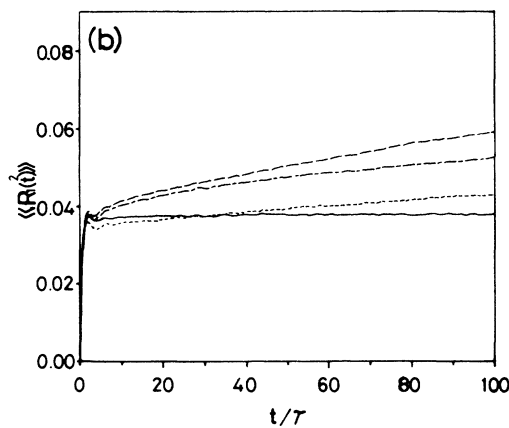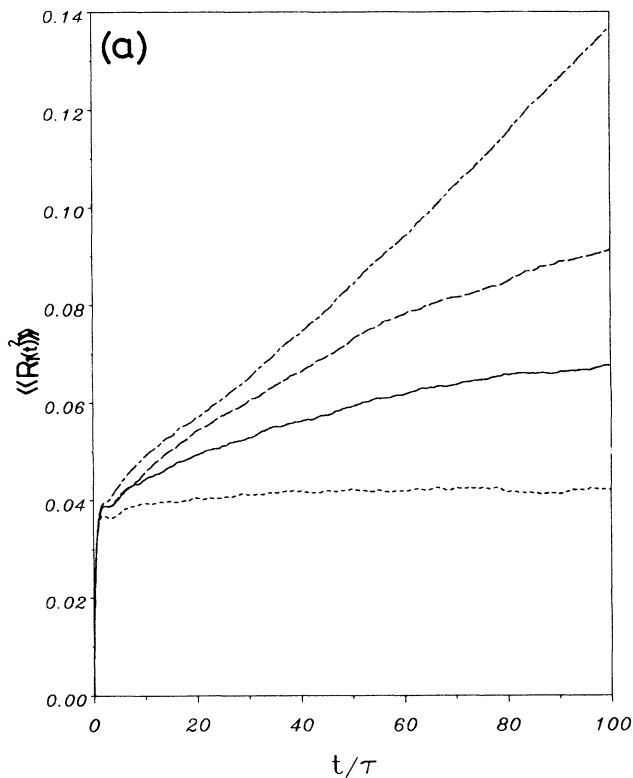

FIG. 5. Mean-square displacement (in units of σ_1^2) for species 1, averaged over atoms and initial times in the respective time windows: (a) $100 \leq t/\tau \leq 500$ and (b) $500 \leq t/\tau \leq 1500$. Solid curve for run 2, dotted curve for run 3, dashed curve for run 4, and dash-dotted curve for run 5.
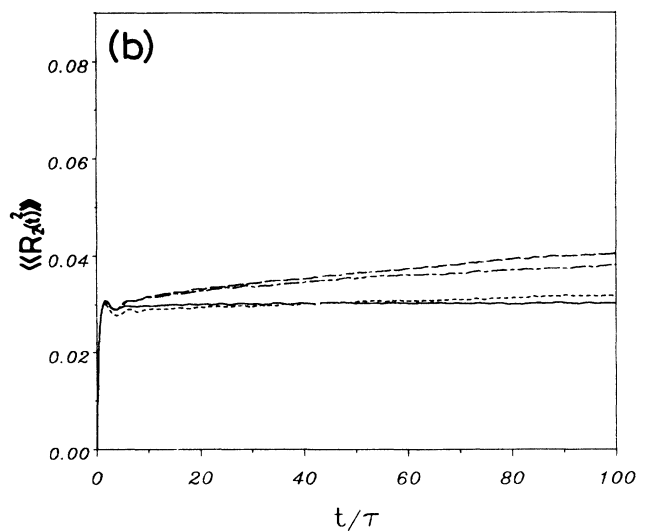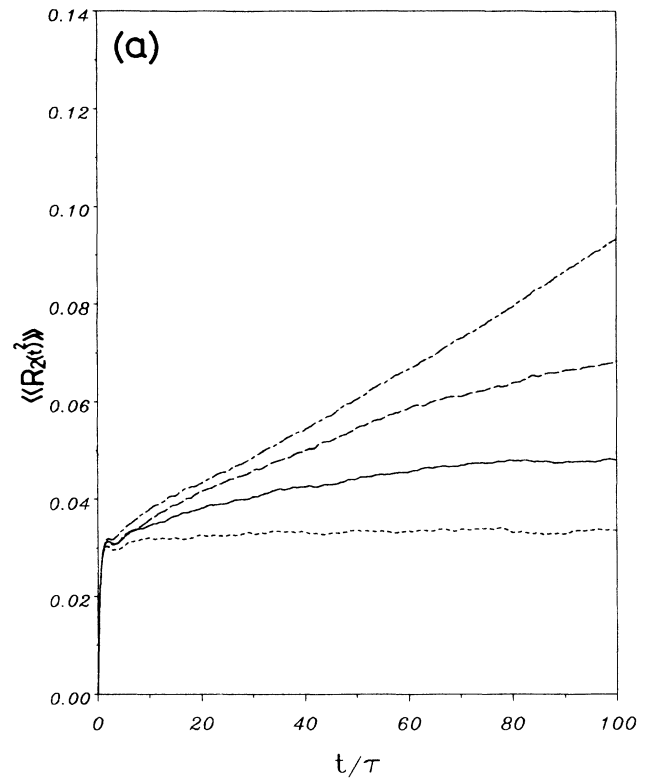
FIG. 6. The same as Fig. 5 but for species 2.

cessible to a quenched glass sample may be restricted to a certain domain of the whole phase space, like an island, depending on given initial conditions, which is possibly connected to other islands by a very narrow channel(s), depending upon detailed dynamical processes of the system under consideration. Thus, the glassy state is essentially a nonequilibrium state, so that every sample, prepared by different quench processes or different annealing processes, may exhibit different results, unless observations (MD simulations) were made for an infinitely long time. Instead, we can show quasi-equilibrium properties of each sample in mesoscopic time scales, shown above. Thirumalai *et al.* have recently discussed the nature of broken ergodicity in glassy states by an examination of an "energy metric" with molecular-dynamics simulations of soft-sphere mixture $(\sigma_2/\sigma_1 = 1.1)^{30}$.

Figure 7 shows a so-called non-Gaussian parameter defined by

$$a_\alpha(t) = \frac{3\langle |\mathbf{r}_{i\alpha}(t) - \mathbf{r}_{i\alpha}(0)|^4 \rangle}{5\langle |\mathbf{r}_{i\alpha}(t) - \mathbf{r}_{i\alpha}(0)|^2 \rangle^2} - 1 \ , \tag{6}$$

where $\mathbf{r}_{i\alpha}(t)$ is the position vector of the ith atom (species α) at time t,[31] and $\langle \ \cdots \ \rangle$ indicates the average over all atoms of the corresponding species (1 or 2) and initial times in the time window under consideration. This function tends towards zero after an order of τ for equilibrium liquids.[17,18,31] However, as shown in Fig. 7, this has even no sign of decaying towards zero for the glass. This highly non-Gaussian behavior is reminiscent of jump-diffusion processes occurring in superionic conductors[32] and underlines the importance of activated processes near the glass transition. From such a behavior, a non-Gaussian parameter at infinite time may be considered to be an order parameter for the glass transition, satisfying that $a_\alpha(t \to \infty) = 0$ above the glass transition temperature T_g $(T > T_g)$, while $a_\alpha(t \to \infty) \neq 0$ below it $(T < T_g)$. The fact that the parameter deviates from zero suggests that there should exist some intrinsic nonlinear motions unlike thermal harmonic vibrations of atoms around a local equilibrium position. Indeed, we have shown well-defined jump motions to appear for glassy states in our previous paper (Figs. 4 and 5 of Ref. 16).

FIG. 7. Non-Gaussian parameter a_α ($\alpha = 1$ for species 1 and $\alpha = 2$ for species 2) as a function of time for run 2 in the respective time windows: (a) $100 \le t/\tau \le 500$ and (b) $500 \le t/\tau \le 2000$. Upper curve for species 1 and lower curve for species 2.

IV. DISCUSSIONS

As the glass transition is approached, the self-diffusion constant becomes very small, so that the relaxation extremely slows down. Recnet theories based on mode-coupling approximations predict a critical slowing-down phenomenon at the transition. These theories predict that the self-diffusion constant vanishes following a power-law formula, and the intermediate scattering function (density autocorrelation function) obeys a stretched exponential decay formula. These predictions have also been supported by recent studies of molecular-dynamics simulations for pure and binary soft-sphere supercooled fluids down to not very close to the glass-transition temperature. Approaching the glass transition, such mode-coupling predicitions appear to be incorrect, since residual diffusion of atoms (activated processes) become important, which may result in smoothing of the transition.

In this paper we have investigated slow-relaxation phenomena of rapidly quenched glassy states of binary soft-sphere mixtures $(\sigma_2/\sigma_1 = 1.2$ and equimolar) prepared by using different quench rates. Quenched glassy states were annealed over a long period to see both static and dynamic relaxations and their sample-dependent behavior. Our main conclusions are the following. (1) Compressibility factor PV/Nk_BT and the partial pair distribution functions are stable without any significant change upon annealing and among different samples. (2) Rapidly quenched samples (glasses) are far from equilibrium states for relatively long initial times, typically for an order of
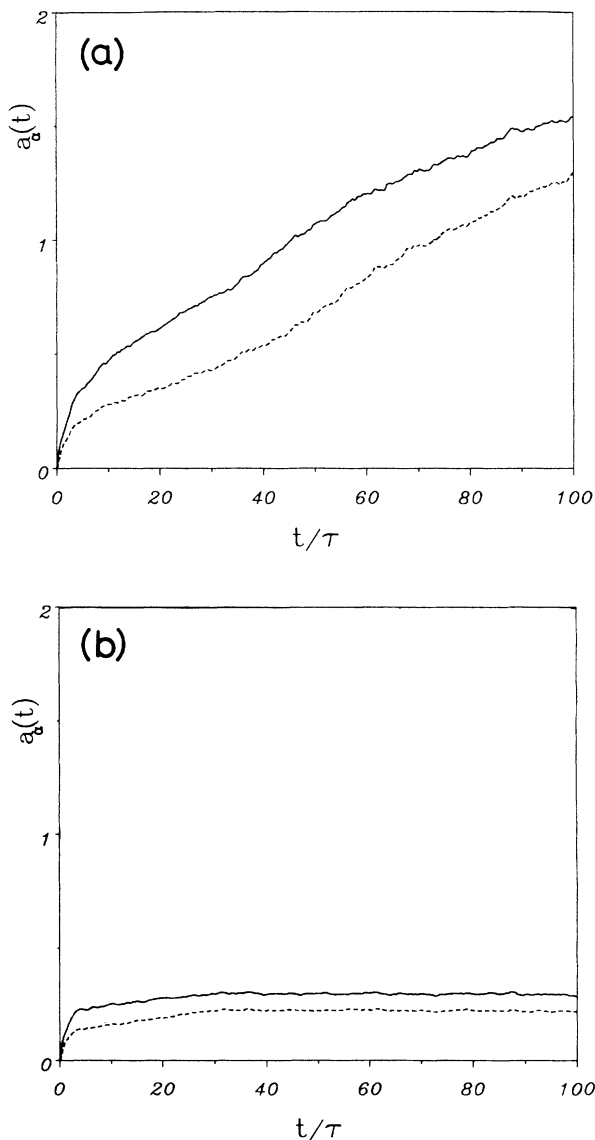
500τ upon annealing of them. For later times, the system appears to be in a quasiequilibrium state. However, the mean-square displacement even for the latter region of times still has a small negative curvature, namely, the t-linear formula (Einstein limit) is not yet sufficiently well attained. (3) No simple systematic quench-rate dependence of the self-diffusion constant is obtained.

The non-Gaussian parameter shows a characteristic property such that it tends toward a nonzero value at long times. Our recent theoretical work based on the trapping model (jump-diffusion model) predicts the non-vanishing nature of the non-Gaussian parameter in glassy states.[33] This property of the non-Gaussian parameter may enable us to consider that it can be used as an order parameter of the glass transition.

The incoherent scattering function, the self part of in-termediate scattering function, is also affected by dynamical slowing-down phenomena near the glass transition and found to yield a stretched exponential decay with an exponent dependent on wave numbers and temperatures. This will be considered in detail in a separate paper.[34]

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