# Evidence of kinetic effects in liquid⇌glass transitions

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The Wendt-Abraham criterion [H. R. Wendt and F. F. Abraham, Phys. Rev. Lett. **41**, 1244 (1978)] is reexamined using computer simulations with an intention to study the kinetic effect in the liquid ⇌ glass transitions. This was carried out by heating an as-quenched glassy metal potassium via (i) a Monte Carlo simulation, which presumably is at an infinite heating rate, and (ii) a molecular-dynamics simulation, which was monitored to run from a finitely high to a lower heating rate. It was found that when the system is heated at a rapid heating rate (relative to cooling), structural relaxation was observed to delay, resulting in an almost linear change in the Wendt-Abraham parameter versus temperature relationship. This delay in structural relaxation was characterized also by virtually unchanged pair-correlation functions for both quenching and heating processes at various temperatures. On the other hand, at a lower heating rate, when the experimental time scale is approaching the average relaxation time, the system undergoes a structural relaxation displaying an anomalous change in the Wendt-Abraham parameter with temperature. In this case structural relaxation was accompanied also by a more orderly structure. These latter behaviors are quite similar to previously related experiments and their interrelation will be discussed in text.

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## I. INTRODUCTION

It is now well established from computer-simulation experiments that a liquid can be supercooled and transformed to a glassy state if a sufficiently high cooling rate is applied. A simple and yet useful empirical criterion that has been employed from both the Monte Carlo (MC) and molecular-dynamics (MD) simulations is to examine the Wendt-Abraham parameter $R$ [1,2]. This parameter, which can be derived from the simulated pair-correlation function $g(r)$, is defined to be $R = g_{\min}/g_{\max}$, where $g_{\min}$ ($g_{\max}$) is the magnitude of $g(r)$ at the first minimum (maximum). Already, $R$ has been utilized by various authors [3–6] to investigate the liquid→glass transition predicting structural results that are interesting and in reasonable accord with those features observed in laboratory experiments. In spite of this appealing success, the change in $R$ versus temperature $T$ has not been used to study the glass→liquid transition. It is the purpose of this work to study $R$ from an as-quenched glassy state.

We consider a monatomic liquid metal potassium as our system of investigation. This judicious choice of the liquid system is for two reasons. First, it will rule out at the outset the possibility of compositional and chemical short-range orderings and that any change that $g(r)$ may undergo is related almost entirely to the topological short-range ordering. Second, it aims at validating further the generality of several glassy features concluded in previous simulation works [2,4,7] for a few different monatomic systems.

The main motive of the present calculation, however, is threefold. In the first place, we propose to understand the $R$-$T$ relation when the as-quenched glassy metal is warmed up at an ideally high heating rate. Second, we intend to study the (kinetic) influences of the heating rate on the variation of $g(r)$ or $R$. This can be approached qualitatively by comparing the MC and MD simulations, because the former is known to have a much higher quenching or heating rate [1,4] than the latter, or quantitatively, by performing MD simulations at several different heating rates. Third, since for either the MC or the MD simulation one needs to supply the pair interaction potential with density evaluated at each $T$, the change in the density versus temperature relationship is a prerequisite. Inspired by the work of Abraham [2], we propose an alternative approach to the determination of such a physical quantity.

It will be shown here that when the as-quenched glassy metal potassium is warmed up by either the MC or the MD method, the $R$-$T$ curve undergoes the glass→liquid transition and displays first a structural relaxation and then returns to the liquid equilibrium state for both simulations. Of particular interest are MD simulations carried out at different heating rates. Here kinetic manifestation of the glass transition is amplified as the occurrence of the hysteresis. These interesting behaviors are quite analogous to some recent laboratory findings [8–12].

Our paper is organized as follows. In Sec. II, we briefly outline the MC and MD simulations giving enough details some of the parameters adopted in simulations. Section III is devoted to a presentation of numerical data. Here we compare our presently obtained data between MC and MD simulations and, wherever possible, with experiments. A possible interpretation of the results and a discussion of its relation to previous studies are given in Sec. IV. Finally, in Sec. V, we summarize our present work and give a conclusion.

## II. COMPUTER SIMULATION

In this section we first describe a reasonable approximation for the estimation of the atomic volume with tem-

 

perature. These data are used to prepare the interatomic potential at various $T$. For the construction of the latter, we employ the generalized nonlocal model pseudopotential theory of Li, Li, and Wang [13], the explicit expression of which can be found in several of our recent works [5,7,14,15]. Both the MC and MD methods will be introduced briefly. However, for technical details not mentioned below, the readers are referred to the works of Valleau and Whittington [16] (for the MC) and Tanaka [17] (for the MD) from which our computer codes were consulted.

### A. Estimation of atomic volume

In order to construct a realistic interatomic potential, we need to input the atomic volume $\Omega_0$ at each quenched temperature. In this work, instead of proceeding with the isothermal-isobaric type of simulation [1,2,18], which is difficult to implement for a metallic system, or with the variational thermodynamic method [19], which is justified only in the supercooled liquid regime, we estimated the change in $\Omega_0$ with $T$ as follows.

First of all, we took the measured liquid and crystalline densities at $T=386$ and 26 K from Borgstedt and Mathews [20] and made a linear interpolation to obtain the atomic volume at each quenched temperature. Using these atomic volumes we constructed interatomic potentials and then carried out the MC and MD simulations. The Wendt-Abraham parameter $R$ was calculated from $g(r)$ and plotted against $T$. From the interception of linear extrapolations associated with the supercooled and glassy branches (see, for example, Ref. [7]), we determined the glass-transition temperature, say, with a value $T_g^i$. Next, we resorted again to the experimental density versus temperature data [20] for a crystalline solid and *assumed* that its thermal linear expansion coefficient (the gradient) is the same as that in a glassy region. Accordingly, the $\Omega_0$ for $T \leq T_g^i$ will be modified to assume this latter gradient. In this way, we arrived at a new $\Omega_0$-$T$ curve. With this new $\Omega_0$-$T$ relation, we performed our MC and MD simulations to obtain a new set of $R$-$T$ data and thereby a corresponding glass-transition temperature $T_g^{i+1}$. For liquid $K$ we found that this $T_g^{i+1}$ has a lower value than the preceding $T_g^i$. Next, we made an interpolation between the $\Omega_0$ at $T_g^{i+1}$ and that at $T=386$ K, while keeping the $\Omega_0$ in the whole of glassy branch below $T_g^{i+1}$ remains unchanged. The latter modification in the $\Omega_0$-$T$ relation is physically legitimate [21] and the whole procedure was repeated iteratively for $i=1,2,\ldots$ until $T_g^{i+1} \approx T_g^i$. We note at this point that this estimation for the variation of $\Omega_0$-$T$, though somewhat qualitative, is reasonable as implied from the works of Abraham ([2], Fig. 5) for the iterative procedure, and Owen ([21], Fig. 12.1) and Zallen ([22], Fig. 1.1) for the assumption that the thermal expansion coefficient of a glassy branch resembles that of its crystalline counterpart. Further support of the idea can be gleaned from some related works [23].

### B. Monte Carlo simulation

To simulate the bulk properties of an infinite system, first we followed the literature [24] by imposing the periodic boundary condition. Second, the minimum image convention was used to account for the interactions among particles which we modeled to be characterized by a pairwise interionic potential. Third, we have employed a total number of 1000 particles sufficiently large for the surface effects to be unimportant. Beginning at a liquid state 50 K above melting point, the system was quenched stepwisely. The starting temperature for our chosen liquid potassium system is therefore at $T=386$ K. The 1000 potassium particles were first placed randomly inside a periodic cubical cell and they were equilibrated through at least $3\times10^5$–$4\times10^5$ MC steps. The displacement vector was adjusted in each run to give an acceptance ratio of about 50%. When the liquid system was ensured to be in a well-equilibrated state (by checking the stability criterion derived from the potential-energy function) we made $7\times10^5$–$21\times10^5$ further MC moves to obtain the pair-correlation function. To examine each subsequent state, earlier configurations of the positions and velocities of particles were taken to be an initial configuration. This procedure was repeated at each quenched temperature with an interval of 40 K down to $T=26$ K. The MC $g(r)$ in the present work lies in the range of 1.6–21 Å.

The heating process merely reverses the above procedure with the as-quenched glassy state at $T=26$ K as the initial configuration. The system was heated up by raising the temperature also at a regular step of 40 K. The equilibration and the stability checking at each $T$ were monitored as in the quenching process. By this means, we warmed the system toward the highest liquid equilibrium state.

### C. Molecular-dynamics simulation

In this method the above-mentioned periodic boundary condition and the minimum image convention are equally applicable. Differing from the MC simulation, the MD approach describes the dynamical behavior of a liquid system. Here a total number of 686 particles were confined in a cube of volume $L^3=MN/\rho$, where $M$ is the mass of a potassium atom, $N$ is the number of particles used in the simulation, and $\rho$ is the mass density. Beeman's algorithm [25] has been adopted in the numerical integration of the Newtonian equations of motion. For a given accuracy, the time step $\Delta t$ depends on a number of factors such as the linear dimension, the square root of the mass, the square root of the interaction strength, etc. After many trials, we have chosen the time step $\Delta t=2\times10^{-15}$ s, which is appropriate for the liquid metal K. As with the MC simulation, we began our simulation at $T=386$ K and quenched the liquid stepwisely at an interval of 40 K. The quenching rate $Q$ in the present work was estimated to be $3\times10^{13}$ K/s. In the heating process, we have performed five heating rates ($2.3Q$, $1.7Q$, $0.8Q$, $0.3Q$, and $0.2Q$) in order to see the kinetic influence on the glass→liquid transition. Since we

were using a microcanonical ensemble, we required the total energy of the system to be a constant. This can be achieved by stabilizing the temperature following a method proposed by Tanaka [17] as follows.

At each quenched or heated temperature, MD records for the velocities $v_i(t)$ of particles were scaled simultaneously for each particle at an arbitrary instant $t$ by a common factor $\xi$ as

$$\xi v_i(t)=\{1+\alpha[\sqrt{T_0/T(t)}-1]\}v_i(t)$$

where $0<\alpha<1$ is a numerical constant and $T(t)=\sum_{i=1}^{N} Mv_i^2(t)/3Nk_B$, $k_B$ being the Boltzmann constant, is the instantaneous temperature. Physically, this scaling in velocities is equivalent to allowing for the particles to have a contact with the heat bath which is maintained at the quenched or heated temperature $T_0$. Specifically, for the case of quenching when $T(t)>T_0$ [or for the case of heating when $T(t)<T_0$] the scaling of $v_i(t)$ will act to remove from (or add to) the particle the amount of kinetic energy required to reach the desired $T_0$. In general, a scaling of $500\Delta t$ was found to be sufficient for achieving the expected $T_0$, although in practice larger scaling steps were required at lower $T$.

## III. NUMERICAL RESULTS

### A. $\Omega_0$-$T$ relation

We depict in Fig. 1 the $\Omega_0$-$T$ relation estimated using the above-mentioned procedure. It can be seen readily from this figure that the MC glass-transition temperature yields a higher $T_g$ than the one determined from MD. This is not surprising since, as pointed out in the Introduction, the former has a much higher quenching rate than the latter. However, on the scale of this curve, the $\Omega_0$-$T$ curves overlap considerably and therefore we have

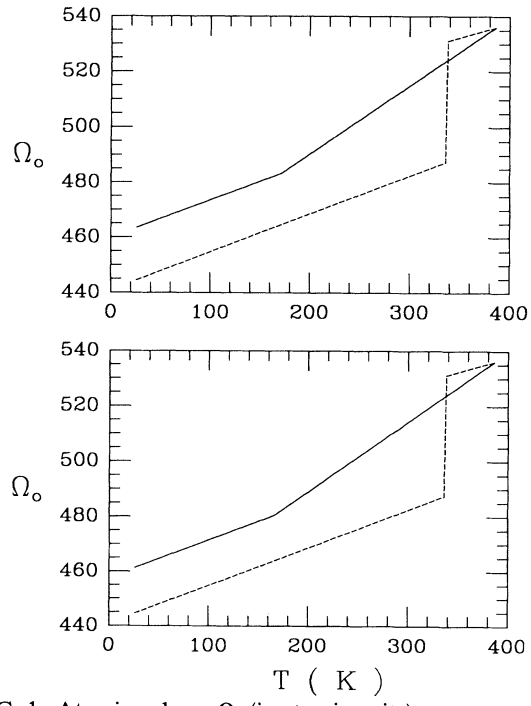


FIG. 1. Atomic volume $\Omega_0$ (in atomic units) vs temperature $T$ for liquid metal potassium. The Monte Carlo iterative result (full curve) is given by the top figure and the crystalline counterpart (dashed curve, calculated from Ref. [20]) is included here for reference; the bottom figure carries the same meaning, but for molecular-dynamics simulation at a quenching rate $Q\approx 3\times 10^{13}$ s.

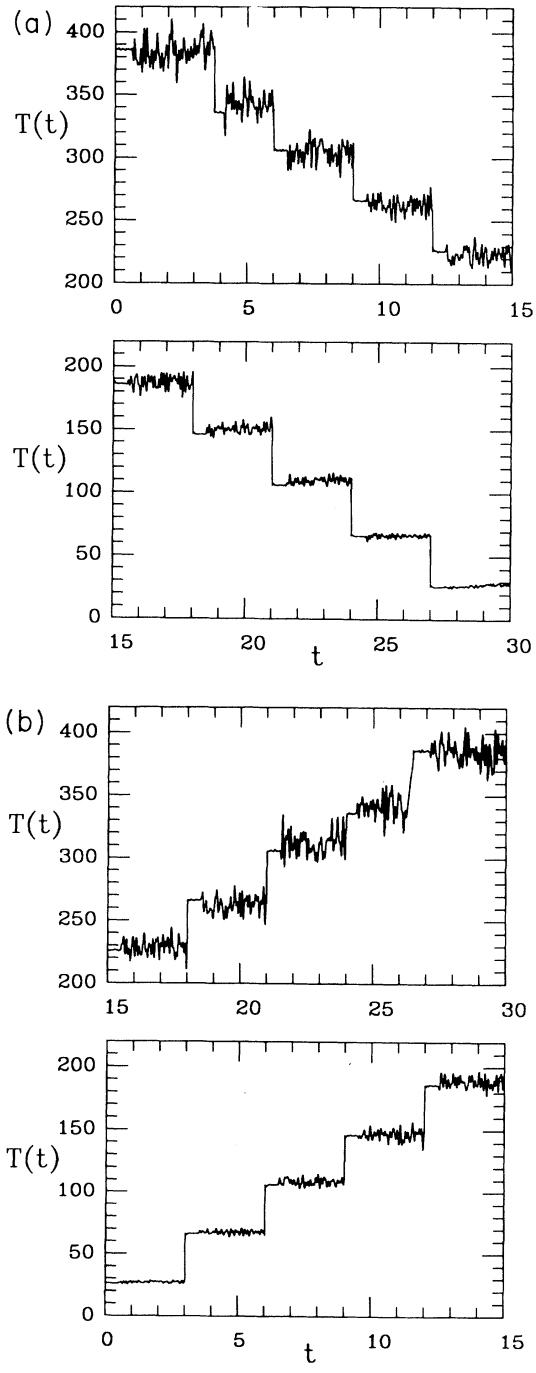


FIG. 2. (a) Instantaneous temperature $T(t)$ (in kelvins) vs time $t$ (in units of $10^3\Delta t$, $\Delta t=2\times 10^{-15}$ s) during cooling process from $T=386$ K down to $T=26$ K for liquid metal potassium. (b) Same as (a), but for the heating process at $\gamma=0.8$ (see text).

plotted them separately. Our simulation shows that the MC simulation yields a $T_g$ about 5 K higher.

### B. Stationary states

Before presenting our main results, it is important to examine our quenched and heated states for the MD simulation to ensure that the intermediate states are indeed stationary. To this end, we delineate in Figs. 2(a) and 2(b) the variation of $T(t)$ during cooling and heating processes for the sequence of heat-bath contacts and for configurations evolving freely at constant energy. Note that the temperature fluctuation at each quenched or heated temperature is approximately $\pm 6$ K or less in magnitude.

### C. Wendt-Abraham parameter

To demonstrate the high reliability of the present simulation, we give in Fig. 3 the Fourier-transformed liquid structure factor for K near melting. It is immediately evident from this figure that both the MC and MD simulations interpret the measured data very well.

Turning now to the quenching and heating results, we depict in Fig. 4 the MC simulation for the $R$-$T$ behaviors. A glance at this figure reveals two general features. First, the quenching and heating $R$-$T$ relations, which presumably are both at infinite cooling rates [1,4] can be divided distinctly into a linear supercooled liquid branch and a linear glassy solid branch. At the interception of these branches, the cooling $R$-$T$ relation predicts a glass transition $R_{LG}=0.11$ at the glass-transition temperature $T_g \approx 171$ K, which is to be compared with values $R_{LG}=0.12$ and $T_g=220$ K estimated from the heating $R$-$T$ curve. Second, the slope of the heated $R$-$T$ curve changes positively throughout the whole temperature range in the glass→liquid transition.

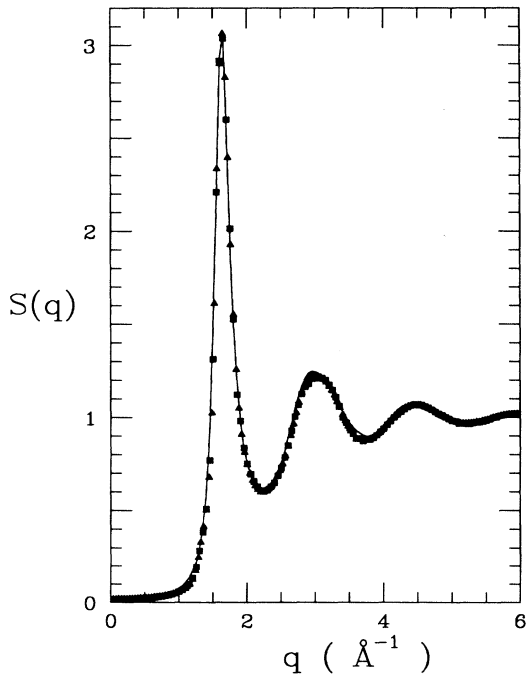


FIG. 3. Liquid structure factor $S(q)$ for liquid potassium calculated by Fourier transforming (1) MC $g(r)$ (solid triangles) and (2) MD $g(r)$ (solid squares) compared with experimental data from Ref. [29].

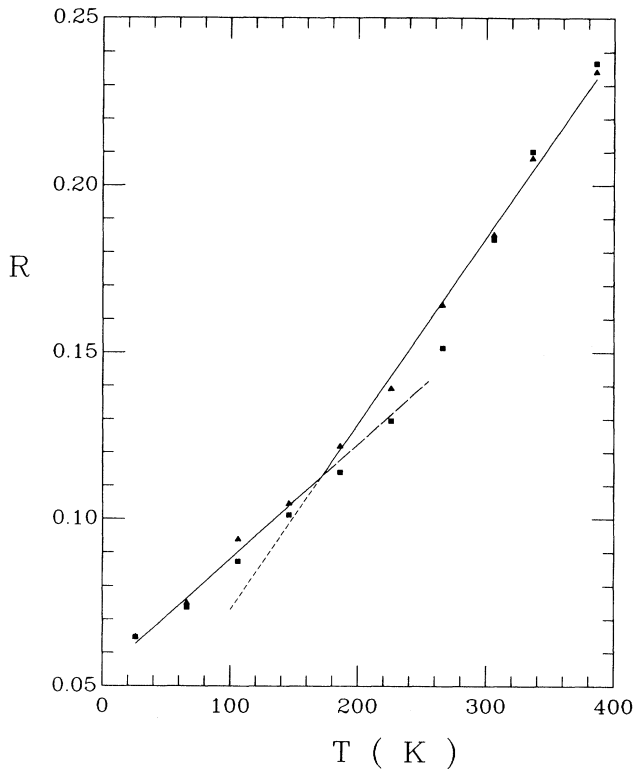


FIG. 4. Monte Carlo simulation data for the Wendt-Abraham parameter $R$ plotted against temperature $T$ for liquid metal potassium. The quenching process, denoted by solid triangles and joined by least-squares full curve, is compared with heating process (solid squares). The equilibrium lines are given by extrapolated dashed lines (see text).

For the MD simulation, quite disparate features are found. To see them, let us plot together the $R$-$T$ behavior for the cooling and individual heating rates. Denoting the ratio of the heating to cooling rates by $\gamma$, Figs. 5(a)–5(e) give values of $\gamma$ approximately 2.3, 1.7, 0.8, 0.3, and 0.2. There are two interesting points that deserve mentioning.

(1) We notice that throughout the temperature range the slope of the heated $R$-$T$ curve is found to vary positively and nearly linearly for $\gamma=2.3$, 1.7, and 0.8. Quantitatively, we find that the obtained $R_{GL}$ and $T_{g,GL}$ in each of the heating processes are generally smaller and higher than the quenched $R_{LG}$ and $T_{g,LG}$, respectively, although scatter of data for $\gamma=2.3$ and 1.7 are somewhat large.

(2) For $\gamma=0.3$ (0.2) the $R$-$T$ relation, given in Fig. 5(d) [Fig. 5(e)], first displays a linear behavior, varies drastically at $T\approx 148$ K (107 K), and then "bends" up from a minimum at 191 K (150 K) toward the same equilibrium

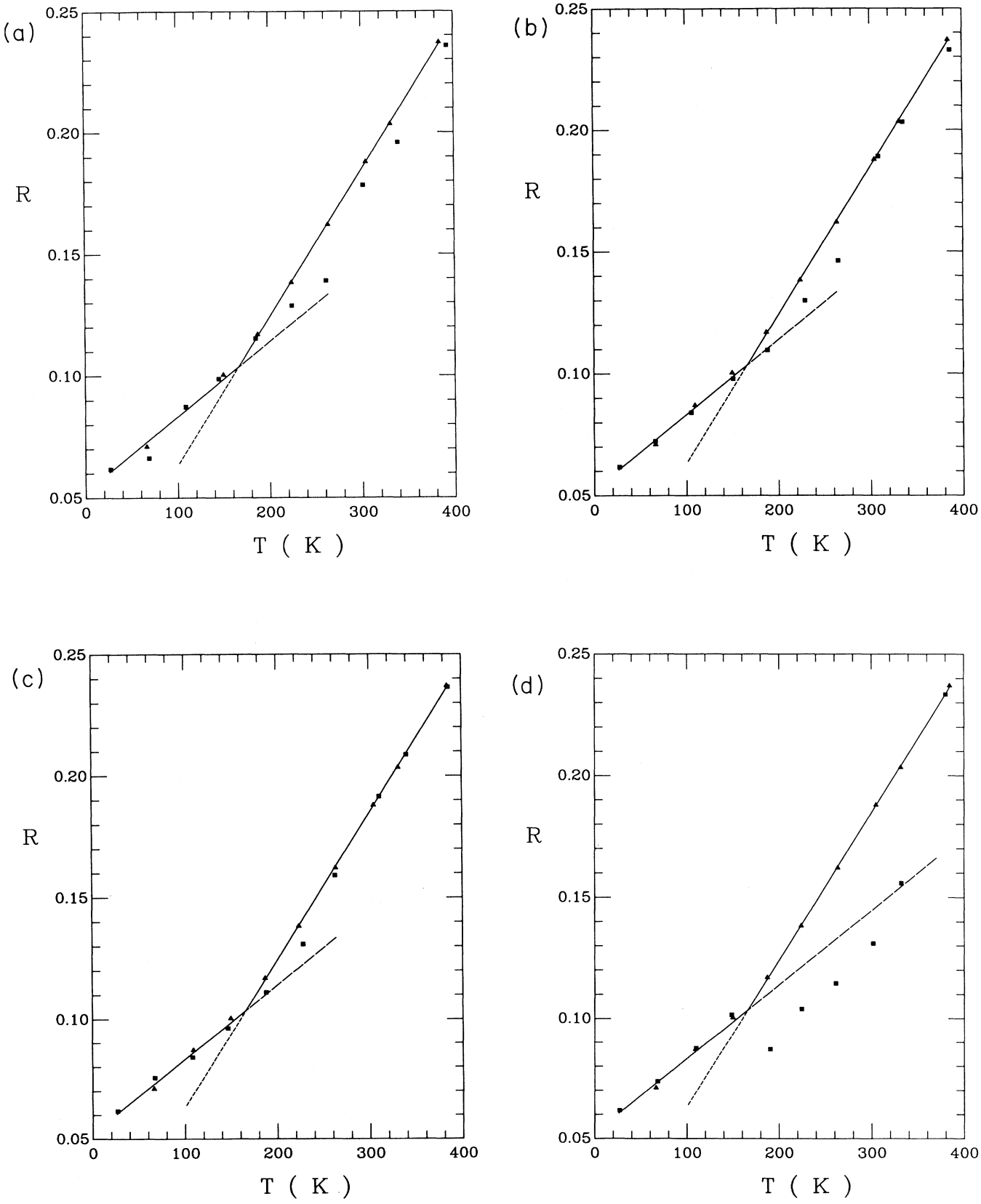


FIG. 5. Same as Fig. 4, but for the molecular-dynamics simulation at (a) $\gamma=2.3$, (b) $\gamma=1.7$, (c) $\gamma=0.8$, (d) $\gamma=0.3$, and (e) $\gamma=0.2$ (see text).

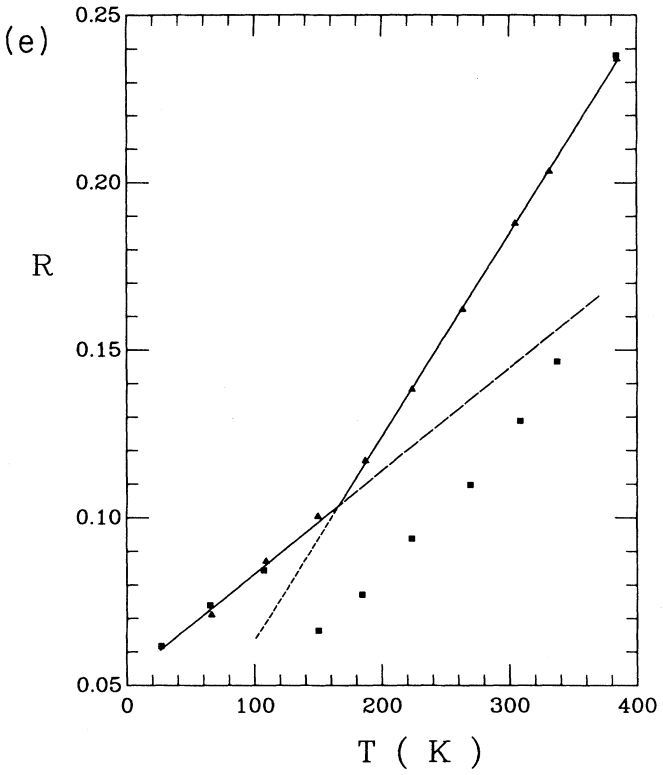


FIG. 5. (*Continued*).

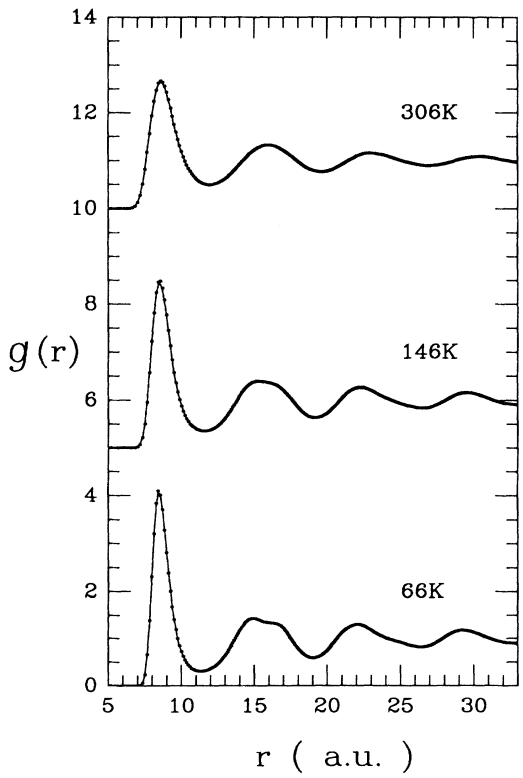


FIG. 6. Monte Carlo simulation results for pair-correlation functions $g(r)$ of liquid metal potassium. Full curve refers to quenching compared with heating denoted by solid circles.

state at $T=386$ K. It is interesting to note from Figs. 5(d) and 5(e) that there are negative slopes for 148 K $< T <$ 191 K and 107 K $< T <$ 150 K, respectively.

In order to further highlight the distinction, we delineate the change in $g(r)$ at three selected $T$ for the MC cooling and heating simulations in Fig. 6 and similarly for the MD at $\gamma=2.3$, 0.8, and 0.2 in Figs. 7(a), 7(b), and 7(c), respectively. We emphasize two important points.

(a) The ideally infinite heating of the MC simulation shows a delay in structural relaxation which is indicated by virtually unchanged $g(r)$ (see Fig. 6) as the glass→liquid state.

(b) The relatively lower and finite heating of the MD simulation, on the other hand, exhibits a structural relaxation which becomes more apparent when $\gamma$ varies from 2.3→0.2 [compare Figs. 7(a)–7(c)].

## IV. DISCUSSION

The numerical results given in the preceding section clearly support the proposition that the liquid⇌glass transitions are a kinetic process. This phenomenon is manifested by the fact that the cooling curve differs from the heating curve, resulting in the appearance of hysteresis. The occurrence of hysteresis is entirely due to the relaxation mechanism for which the glass transition is intimately associated. Physically we may look at this phenomenon from the point of view of the distribution of relaxation times [26]. For a given cooling rate sufficiently rapid for a glassy state to be formed, the behavior of the glassy metal depends crucially on the heating rate. At a high heating rate it can be seen from Figs. 4 and 5(a)–5(c) that the magnitude of $R$, being slightly smaller than the cooling, deviates away from the equilibrium liquid line (dashed lines in these figures) at the lower temperature. As temperature increases at a rapid heating rate, structural relaxation is delayed, and the material remains in a glassy phase for a longer period of time. This explains why for $\gamma=2.3$, 1.7, and 0.8 the heated glassy branch is more extensive than the corresponding cooling glassy branch. On the other hand, at a lower heating rate [Fig. 5(d) or 5(e)] when the time scale of experiment (approximately equal to the inverse heating rate) is approaching the average structural relaxation time, we see that the glassy material relaxes toward equilibrium. The reason for this to occur is because at a lower heating rate, the system will behave more like a liquid which is in contrast to the solidlike glassy state during the (shorter) time scale characteristic of quenching. Accordingly the glassy system relaxes at a relatively lower temperature and approaches the equilibrium value (dashed curve). This will continue until it crosses the equilibrium line and the material relaxes toward it from below. This crossover behavior is reminiscent of the crossover experiment of Spinner and Napolitano [11] whose index-of-refraction measurement was explained by Moynihan *et al.* [27] using the two-relaxation-time approximation, and of Beukel and Sietsma [8], who discussed a similar hysteresis feature using the free-volume theory of Cohen and Turnbull [28]. The hysteresis behavior has been observed also in glass transition in the spectroscopic [10], enthalpy [30],

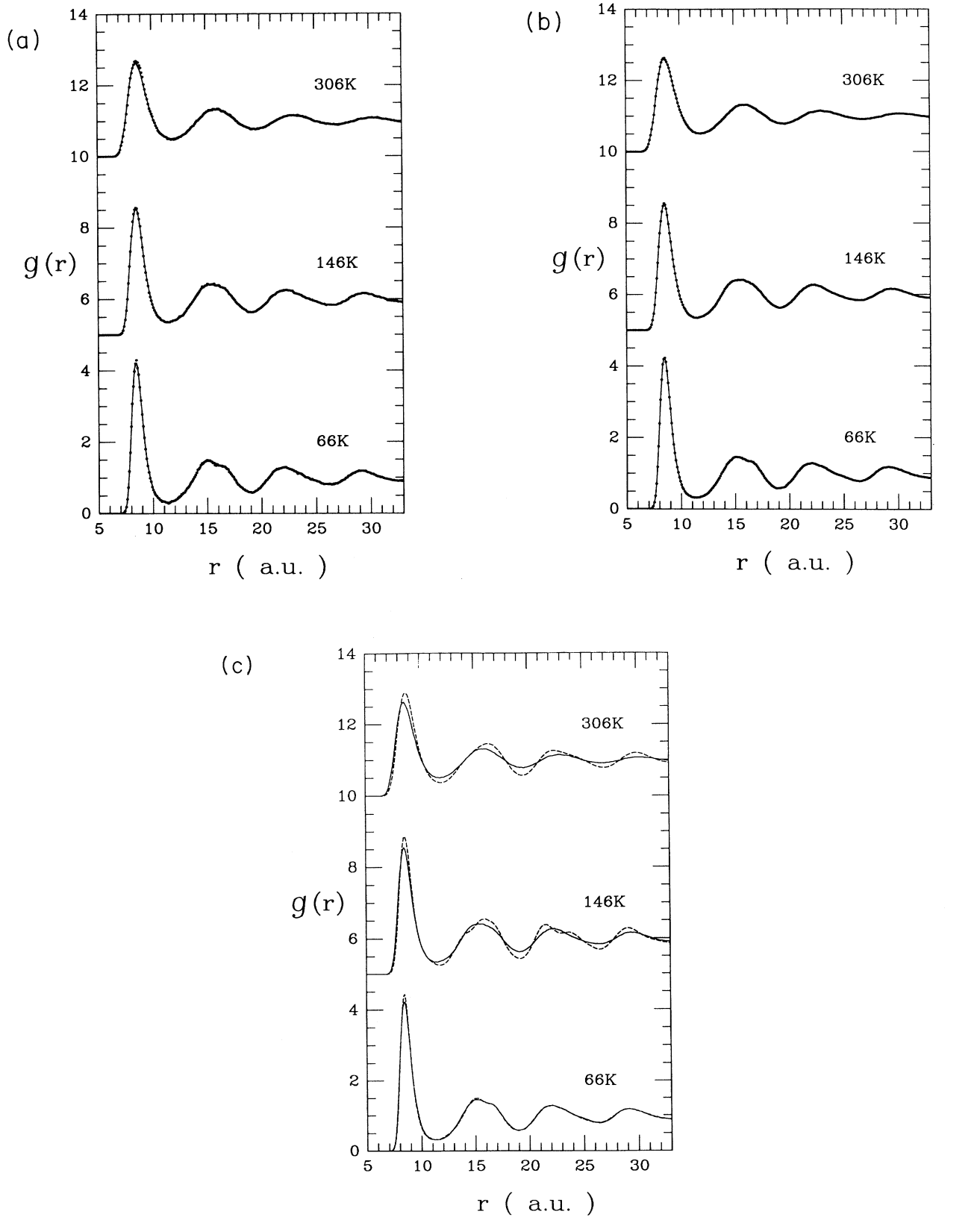


FIG. 7. (a) Molecular-dynamics simulation results for pair-correlation functions $g(r)$ of liquid metal potassium. Full curve refers to quenching compared with heating process (solid circles) at $\gamma=2.3$ (see text). (b) Same as (a), but for $\gamma=0.8$. (c) Molecular-dynamics simulation results for pair-correlation functions $g(r)$ of liquid potassium. Full curve refers to quenching compared with heating process (dashed curve) at $\gamma=0.2$ (see text).

and viscosity [12] experiments. We note that since the Wendt-Abraham criterion as used here is estimated directly from $g(r)$, the magnitude of the change in $R$ at each $T$ may reflect, though qualitatively, the feasibility of the dynamics of flow. There is, however, one point to be noted. In the present computer-simulation heating process, because of the lack of a reliable way to obtain $\Omega_0$-$T$ behavior, we have *assumed* the same quenched volume versus temperature relation and hence do not allow the volume of the system to be relaxed. This explains why structural relaxation at much lower heating rates ($\gamma=0.3$ and 0.2) does not contain more orderly structure, although tendencies toward solidlike behavior can still be gleaned from the anomalous behavior of $R$ [see Figs. 5(a)–5(e)] and from Figs. 7(a)–7(c).

## V. CONCLUSION

The electron theory of metals has been applied to calculate the total energy and hence the interatomic pair potential for liquid $K$ at various densities. This is based on a highly accurate generalized nonlocal model pseudopotential. The atomic volume which is needed at each quenched temperature for the present computer simulation is obtained via an iterative scheme inspired from the Monte Carlo simulation of Abraham for a Lennard-Jones system. The calculated liquid structure factor at the melting point has been examined against x-ray experiments and found to agree very well with each other. With confidence in the constructed pair potential, we proceed to determine the $R$-$T$ behavior both for quenching and for various heating rates. It is found that, generally, the cooling curve differs from the heating curves, implying that the glass transition is a kinetic phenomenon. Quantitatively, for a given cooling rate we observe the following.

(a) A rapid heating rate has a more extensive glassy branch indicating that the material undergoes a delay in structural relaxation. This can be seen from the MC simulation (infinite heating rate) and from MD simulations at $\gamma=2.3$, 1.7, and 0.8.

(b) At a lower heating rate, structural relaxation begins at a lower temperature, approaches an equilibrium fluid value, and relaxes toward the equilibrium line from below resulting in a crossover behavior—hysteresis.

These features are similar to previously reported experiments and is due to the existence of a relaxation-time distribution.

## ACKNOWLEDGMENTS

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