

Testing mode-coupling theory for a supercooled binary Lennard-Jones mixture. II. Intermediate scattering function and dynamic susceptibility

Walter Kob*

Institut für Physik, Johannes Gutenberg-Universität, Staudinger Weg 7, D-55099 Mainz, Germany

Hans C. Andersen†

Department of Chemistry, Stanford University, Stanford, California 94305

(Received 10 April 1995)

We have performed a molecular dynamics computer simulation of a supercooled binary Lennard-Jones system in order to compare the dynamical behavior of this system with the predictions of the idealized version of mode-coupling theory (MCT). By scaling the time t by the temperature dependent α -relaxation time $\tau(T)$, we find that, in the α -relaxation regime, $F(q, t)$ and $F_s(q, t)$, the coherent and incoherent intermediate scattering functions, for different temperatures each follow a q -dependent master curve as a function of scaled time. We show that during the early part of the α relaxation, which is equivalent to the late part of the β relaxation, these master curves are well approximated by the master curve predicted by MCT for the β relaxation. This part is also fitted well by a power law, the so-called von Schweidler law. We show that the effective exponent b' of this power law depends on the wave vector q if q is varied over a large range. The early part of the β -relaxation regime does not show the critical decay predicted by MCT. The q dependence of the nonergodicity parameter for $F_s(q, t)$ and $F(q, t)$ is in qualitative agreement with MCT. On the time scale of the late α relaxation the correlation functions show a Kohlrausch-Williams-Watts behavior (KWW). The KWW exponent β is significantly different from the effective von Schweidler exponent b' . At low temperatures the α -relaxation time $\tau(T)$ shows a power-law behavior with a critical temperature that is the same as the one found previously for the diffusion constant [Kob and Andersen, Phys. Rev. Lett. **73**, 1376 (1994)]. The critical exponent of this power law and the von Schweidler exponent b' fulfill the connection proposed by MCT between these two quantities. We also show that the q -dependent relaxation times extracted from the correlation functions are in accordance with the α -scale universality proposed by MCT. The dynamic susceptibility $\chi''(\omega)$ data for different temperatures also fall on a master curve when frequency is scaled by the location of the minimum between the microscopic peak and the α peak and χ'' is scaled by its value at this minimum. The low frequency part of this master curve can be fitted well with a functional form predicted by MCT. However, the optimal value for the exponent parameter from this fit does not agree with the one determined from the corresponding fit in the time domain. The high frequency part of the master curve of $\chi''(\omega)$ cannot be fitted well by the functional forms predicted by MCT, in accordance with our findings from the time domain. We test various scaling laws predicted by the theory and find that they are qualitatively correct but that the exponents do not fulfill certain relations predicted by the theory if they involve the critical exponent a of MCT. This discrepancy can be rationalized by means of the strong influence of the microscopic dynamics on the β relaxation at early times. Those scaling laws that do not involve the critical exponent a are in qualitative and quantitative accordance with the theory.

PACS number(s): 61.20.Lc, 61.20.Ja, 64.70.Pf, 51.10.+y

I. INTRODUCTION

In a recent paper [1] we have reported some of the results we obtained from a large scale simulation of a supercooled binary Lennard-Jones mixture. The aim of this work was to test whether the mode-coupling theory

(MCT) is able to correctly describe the dynamical behavior of such a system. This theory was originally developed to describe the dynamics of simple liquids in the supercooled state [2,3]. However, in recent years the theory has also been successfully applied to rationalize the dynamics of more complex liquids. Despite these successes, there is still a great deal of controversy on whether the theory is really able to correctly describe the dynamics of liquids at low temperatures. The reader can find good introductions to the theory in some review articles [4,5] and a list of most relevant references on MCT in [1,4,5]. Recently also a useful collection of review articles on MCT has appeared [6].

Since MCT was originally developed for supercooled

*Electronic address: kob@moses.physik.uni-mainz.det
WWW: <http://www.cond-mat.physik.uni-mainz.de/~kob/home.kob.html>

†Electronic address: fb.hca@forsythe.stanford.edu

simple liquids various groups performed computer simulations of such systems in order to investigate whether MCT gives a correct description of the dynamical behavior of such systems [1,7–15]. The simulations showed that at low temperatures several features of the dynamical behavior of these systems could be rationalized by MCT. However, many of these simulations had the shortcoming that the systems were not well equilibrated at low temperatures, i.e., exactly in that range of temperature where MCT is supposed to apply. Since MCT is an *equilibrium* theory it is of utmost importance that at every temperature investigated the length of the run is sufficiently long to allow the system to equilibrate. Failure to do so will usually lead to a relaxation behavior that is quite different from the one observed if the system is in equilibrium and thus no firm conclusions can be drawn on whether MCT gives a correct description of the dynamics or not. Therefore one of the virtues of the work in Ref. [1] was to make sure that the system was thoroughly equilibrated at all temperatures investigated. A further important aspect of that work and the one reported here is the fact that many *different* aspects of the predictions of MCT are tested in order to investigate whether the theory is able to give a *self-consistent* picture of the dynamics of the system and is therefore more than a convenient way to analyze the relaxation data.

In Ref. [1] we mainly concentrated on the investigation of the mean squared displacement of a tagged particle, the diffusion constant, and the van Hove correlation function. We showed that at low temperatures the mean squared displacement showed a plateau in a time range that extended over several decades in time and could be identified with the β -relaxation regime (see Sec. II for a definition of this term) predicted by MCT. At low temperatures the diffusion constant showed a power-law behavior, in accordance with the theory. With the help of the van Hove correlation function we showed that the so-called cage effect is indeed present at these temperatures. These correlation functions allowed us also to show that the factorization property predicted by the theory holds for this system. Furthermore, we gave evidence that for this system the so-called hopping processes are not important in the temperature range we investigated and that therefore the dynamics of the system can be tested with the *idealized* version of the theory, in which such processes are neglected.

In this work we extend our analysis of the dynamical behavior of our system to the investigation of the intermediate scattering function and the dynamical susceptibility. Since several of the predictions of the theory can most conveniently be tested with these quantities, this investigation will allow us to perform more extensive tests of the theory and thus help to decide whether MCT is able to correctly describe the dynamical behavior of simple supercooled liquids. Some of these results have been reported already in a previous paper [7] where we presented the scaling behavior of the incoherent intermediate scattering function for a value of the wave vector q in the vicinity of the maximum of the structure factor. These results are here extended to a larger range of values of q and different types of correlation functions.

The rest of the paper is organized in the following way. In Sec. II we give a short review of some of the predictions of the theory in order to facilitate the understanding of the subsequent tests of the theory presented in this work. In Sec. III we introduce some of the details of our model and of our simulation. Section IV is then devoted to the presentation of our results, which are summarized in Sec. V.

II. MODE-COUPPLING THEORY

In order to facilitate the reading of this paper, we compile in this section some of the predictions of MCT. The derivation of these predictions can be found in the original papers or in review articles [4,5].

Mode-coupling theory attempts to describe the dynamics of strongly supercooled liquids at temperatures slightly above the glass transition temperature T_g . In its simplest version, the so-called idealized MCT, the theory predicts the existence of a temperature T_c at which the system undergoes a transition from ergodic behavior to nonergodic behavior. This means that certain types of correlation functions, such as, e.g., the intermediate scattering function for wave vector q , $F(q, t)$, do not decay to zero even for long times if $T < T_c$. This transition is predicted to be observable for all time correlation functions $\langle X(0)Y(t) \rangle$ between dynamical variables X and Y for which the overlap with the Fourier transform of the density fluctuations $\delta\rho(q) = \rho(q) - \langle \rho(q) \rangle$ is nonzero, i.e., for which $\langle \delta\rho(q)X \rangle \neq 0$ and $\langle \delta\rho(q)Y \rangle \neq 0$. Here $\langle \rangle$ stands for the canonical average. The following results are all of an asymptotic nature in the sense that they are valid only if $\epsilon \equiv (T - T_c)/T_c$, the small parameter of the theory, tends to zero.

The theory also predicts the existence of a parameter λ , the so-called exponent parameter, which is very important for the *quantitative* description of the relaxation behavior (see below for details). This exponent parameter can be computed if the structure factor of the system is known with sufficient precision, but since this is seldom the case it is usually treated as an adjustable parameter for fitting the data.

Consider a normalized time correlation function $\phi(t) = \langle X(0)Y(t) \rangle / \langle XY \rangle$ for two dynamical variables X and Y that have a nonvanishing overlap with $\delta\rho(q)$. MCT predicts that for temperatures just above T_c $\phi(t)$ shows a two step relaxation behavior. Starting with the value of unity at time zero $\phi(t)$ is supposed to decay to a value of $f_c > 0$, the so-called nonergodicity parameter, and then decay slowly to zero for long times. Thus if $\phi(t)$ is plotted versus the *logarithm* of time, the function quickly decays from unity on a microscopic time scale, then slowly decays to a plateau of height f_c , and finally slowly decays to zero. In the language of MCT the time range for which $\phi(t)$ is close to this plateau is called the β regime and the time range that starts with the correlators beginning to deviate from this plateau and that extends to infinite time is called the α regime. Two things should be noted in order to avoid getting confused. The first is that the β -relaxation regime of MCT should not be confused

with the β relaxation as it has been introduced by Johari and Goldstein [16], since the latter is a *peak* in the spectrum whereas the former corresponds to a *minimum* in the spectrum. The second thing to note is that the *late* part of the β regime overlaps with the *early* part of the α regime. Thus the two regimes should not be considered as *completely* unrelated relaxation regimes.

For times in the β -relaxation regime MCT predicts that the correlator $\phi(t)$ can be written in the following form:

$$\phi(t) = f_c + hG(t). \quad (1)$$

Here f_c is the above mentioned nonergodicity parameter and h is a positive amplitude factor. Both quantities will depend on the nature of ϕ , e.g., they will depend on the wave vector q if ϕ is the intermediate scattering function, but not on temperature T or time t . The whole temperature and time dependence of the right hand side of Eq. (1) is given by the function $G(t)$. This function is predicted by MCT to be of the form

$$G(t) = \sqrt{\epsilon} |g(t/t_\epsilon), \quad (2)$$

where ϵ is the small parameter of the theory introduced above and t_ϵ is the time scale of the β relaxation. This time scale is predicted to show a power-law dependence on T with a divergence at $T = T_c$:

$$t_\epsilon \sim (T - T_c)^{-1/2a}, \quad (3)$$

where the quantity a can be computed from the exponent parameter λ (see below). If λ is known, the function $g(t/t_\epsilon)$ in Eq. (2) can be computed explicitly. It has been shown analytically that, for times much larger than the microscopic times t_0 but less than t_ϵ , $g(t)$ is a power law, in this context often called the critical decay, i.e., it is of the form

$$g(t/t_\epsilon) = (t_\epsilon/t)^a, \quad t_0 \ll t \leq t_\epsilon. \quad (4)$$

Here the exponent a , often also called the critical exponent, is the same quantity that appeared in Eq. (3).

For times t that are larger than t_ϵ but much smaller than the α -relaxation time τ , $g(t/t_\epsilon)$ is predicted to be also a power law, which in this context is usually called the von Schweidler law, i.e.,

$$g(t/t_\epsilon) = -B(t/t_\epsilon)^b, \quad t_\epsilon \leq t \ll \tau. \quad (5)$$

Here the prefactor B and the exponent b , the so-called von Schweidler exponent, can also be computed when the exponent parameter λ is known. In particular, MCT predicts that the exponent a of the critical decay and the von Schweidler exponent b are related to the exponent parameter via

$$\lambda = \frac{[\Gamma(1-a)]^2}{\Gamma(1-2a)} = \frac{[\Gamma(1+b)]^2}{\Gamma(1+2b)}, \quad (6)$$

where $\Gamma(x)$ is the Γ function.

For times on the time scale of the α -relaxation regime MCT predicts that the correlator $\phi(t)$ obeys the so-called

time-temperature superposition principle. This means that correlators for different temperatures (all close to T_c , of course) will fall onto a master curve if time is scaled by the α -relaxation time τ , i.e.,

$$\phi(t) = F(t/\tau(T)). \quad (7)$$

For the late α -relaxation regime the master function $F(t/\tau)$ is predicted to be well approximated by a Kohlrausch-Williams-Watts (KWW) function, often also called stretched exponential, i.e.,

$$\phi(t) \approx A \exp[-(t/\tau)^\beta], \quad (8)$$

and recently it has been shown that if $\phi(t)$ is the intermediate scattering function for wave vector q of a simple liquid Eq. (8) becomes exact for large values of q [17,18]. The α -relaxation time τ will in general depend on the specific nature of ϕ , e.g., on the wave vector q if ϕ is the intermediate scattering function $F(q, t)$. However, MCT predicts that the relaxation times of all correlators should show a divergent behavior near T_c in the form of a power law with an exponent γ that is independent of the type of correlator studied. Thus the relaxation time of the intermediate scattering function $F(q, t)$ is predicted to be of the form

$$\tau(q) = C(q)(T - T_c)^{-\gamma}. \quad (9)$$

Here $C(q)$ is a smooth function of temperature. Thus in the vicinity of T_c the main dependence of τ on temperature is given by the power-law behavior in Eq. (9). This property is called the α -scale universality. The exponent γ in Eq. (9) can be computed once λ is known by means of

$$\gamma = \frac{1}{2a} + \frac{1}{2b}, \quad (10)$$

where a and b are the two exponents from Eq. (6).

By making use of Eqs. (1)–(3), (5), and (9) it is simple to show that on the time scale of the late β -relaxation regime, i.e., the time scale for which the von Schweidler law in Eq. (5) is predicted to hold, the von Schweidler law can also be written as follows:

$$\phi(t) = f_c - hB(t/\tau)^b, \quad (11)$$

where h and B are the temperature-independent constants of Eqs. (1) and (5). Note that MCT predicts that the exponent b in Eqs. (5), (6), (10), and (11) is in general not the same as the KWW exponent β in Eq. (8). Thus Eq. (11) is *not* the short time expansion of the KWW law in Eq. (8).

If the correlation function $\phi(t)$ is time Fourier transformed and multiplied by the frequency ω one obtains the dynamic susceptibility $\chi''(\omega)$. Since $\phi(t)$ is predicted to show, at temperatures just above T_c , a two step relaxation behavior, $\chi''(\omega)$ is predicted to show a double peak structure at these temperatures. MCT makes predictions about the following quantities: ω_ϵ , the frequency at the minimum between the two peaks; $\chi''_\epsilon \equiv \chi''(\omega_\epsilon)$, the value of the susceptibility at this minimum; and ω_{max} , the fre-

quency of the peak that occurs at lower frequency (the so-called α peak). In particular, MCT predicts the two following power-law dependencies for χ''_ϵ and ω_{max} on temperature:

$$\chi''_\epsilon \sim (T - T_c)^{1/2} \quad (12)$$

and

$$\omega_{max} \sim \tau^{-1} \sim (T - T_c)^\gamma, \quad (13)$$

where we made use of Eq. (9). Making use of Eqs. (3) and (12) it follows that

$$\chi''_\epsilon \sim \omega_\epsilon^a \quad (14)$$

and using Eqs. (3), (10), and (13) one can show that

$$\omega_\epsilon \sim \omega_{max}^{b/(a+b)}. \quad (15)$$

It should be recognized that Eq. (15) can be derived from Eqs. (3), (10), and (13) even if Eq. (6) does not hold. This point will be important later when we discuss our results.

Note that some of these predictions are consequences of the simplest version of MCT, the so-called idealized MCT. If thermally activated processes are present, in this context usually called hopping processes, some of the above predictions have to be modified. However, in a previous paper we have given evidence that for our system these hopping processes are not important in the temperature range we investigated [1]. Thus it is appropriate to compare the low temperature dynamics of our system with the idealized version of the theory.

III. MODEL AND SIMULATION

In this paper we give only some of the most important details of the model investigated and the essential features of the simulation. More details can be found in Ref. [1].

The system considered is a binary mixture of Lennard-Jones particles. Both types of particles (type A and type B) have the same mass m . The interaction potential $V_{\alpha\beta}(r)$ is given by $V_{\alpha\beta}(r) = 4\epsilon_{\alpha\beta}[(\sigma_{\alpha\beta}/r)^{12} - (\sigma_{\alpha\beta}/r)^6]$ ($\alpha, \beta \in \{A, B\}$) with the following set of parameters: $\epsilon_{AA} = 1.0$, $\epsilon_{AB} = 1.5$, $\epsilon_{BB} = 0.5$, $\sigma_{AA} = 1.0$, $\sigma_{AB} = 0.8$, and $\sigma_{BB} = 0.88$. For computational efficiency these potentials were truncated and shifted at a distance of $2.5\sigma_{\alpha\beta}$. In the following we report all quantities in reduced units, i.e., length in units of σ_{AA} , energy in units of ϵ_{AA} , and time in units of $(m\sigma_{AA}^2/48\epsilon_{AA})^{1/2}$. For argon these units correspond to a length of 3.4 Å, an energy of 120 K k_B , and a time of 3×10^{-13} s.

The number of A and B particles was 800 and 200, respectively. The equations of motions were solved with the velocity form of the Verlet algorithm with a time step of 0.01 and 0.02 at high ($T \geq 1.0$) and low ($T < 1.0$) temperatures, respectively. The length of the cubic box was $L = 9.4\sigma_{AA}$ and periodic boundary conditions were applied. The system was prepared at a high temperature

($T = 5.0$) and subsequently cooled to lower temperatures by coupling it to a stochastic heat bath. The temperatures investigated were $T = 5.0, 4.0, 3.0, 2.0, 1.0, 0.8, 0.6, 0.55, 0.50, 0.475$, and 0.466 . Multiple runs were performed at each temperature, and the correlation functions were averaged. Since MCT assumes the system to be in equilibrium, great care was taken in order to make sure that we gave the system enough time to equilibrate at *all* temperatures. This was done by allowing the system to equilibrate at each temperature for a time that was longer than the time of the α relaxation. In Ref. [1] we give strong evidence that the cooling schedule and the equilibration times are such that the dynamical behavior observed is really that of an equilibrium system.

IV. RESULTS

In this section we present the results of our simulation. In the first part we will report our findings about the intermediate scattering function. This quantity is very useful in order to test certain kind of predictions of MCT. However, other predictions of the theory are more easily tested with the help of the dynamic susceptibility and therefore we devote the second part of the section to the investigation of this quantity.

A. Intermediate scattering function

The intermediate scattering function is the space Fourier transform of the van Hove correlation function $G(r, t)$ [19]. The latter can be split into two parts, the self part $G_s(r, t)$, and the distinct part $G_d(r, t)$. Consequently, there exist also two different types of intermediate scattering functions: $F_s(q, t)$, the incoherent intermediate scattering function, which is the Fourier transform of $G_s(r, t)$, and $F(q, t)$, the coherent intermediate scattering function, which is the Fourier transform of the van Hove correlation function $G(r, t)$. We computed these two correlation functions by computing first the thermal average of the van Hove correlation function and then took the Fourier transform of this function [20]. Although this procedure is formally exact in the thermodynamic limit it turned out not to be the best way to compute the intermediate scattering function. The problem is that at low temperatures the distinct part of the van Hove correlation function does not attain its asymptotic value, i.e., unity, at the distance $L/2$, i.e., at the largest distance accessible in the simulation (L is the length of the box). This can be seen, e.g., in Fig. 9 of Ref. [1] where we plot $G(r, t)$ for $t = 0$. Therefore our computation of $F(q, t)$ involved taking the Fourier transform of a function that had a discontinuity at $r = L/2$ and, as can be easily shown, this leads to an oscillatory behavior of the Fourier transform at small values of q with a periodicity (in q) of $4\pi/L$ and an amplitude that decays like q^{-1} . A possible solution of this problem would have been to compute the intermediate scattering function from the positions of the particles and only afterwards do the average over different configurations; unfortunately, this problem

was recognized only after the simulations had been performed. However, we believe that the oscillations are only a minor flaw in the data and in particular do not obscure the relaxation behavior of the correlation functions.

In Fig. 1 we show the structure factors $S(q)$, given by $F(q, t)$ at $t = 0$, versus q for all temperatures investigated. For clarity the curves for the different tempera-

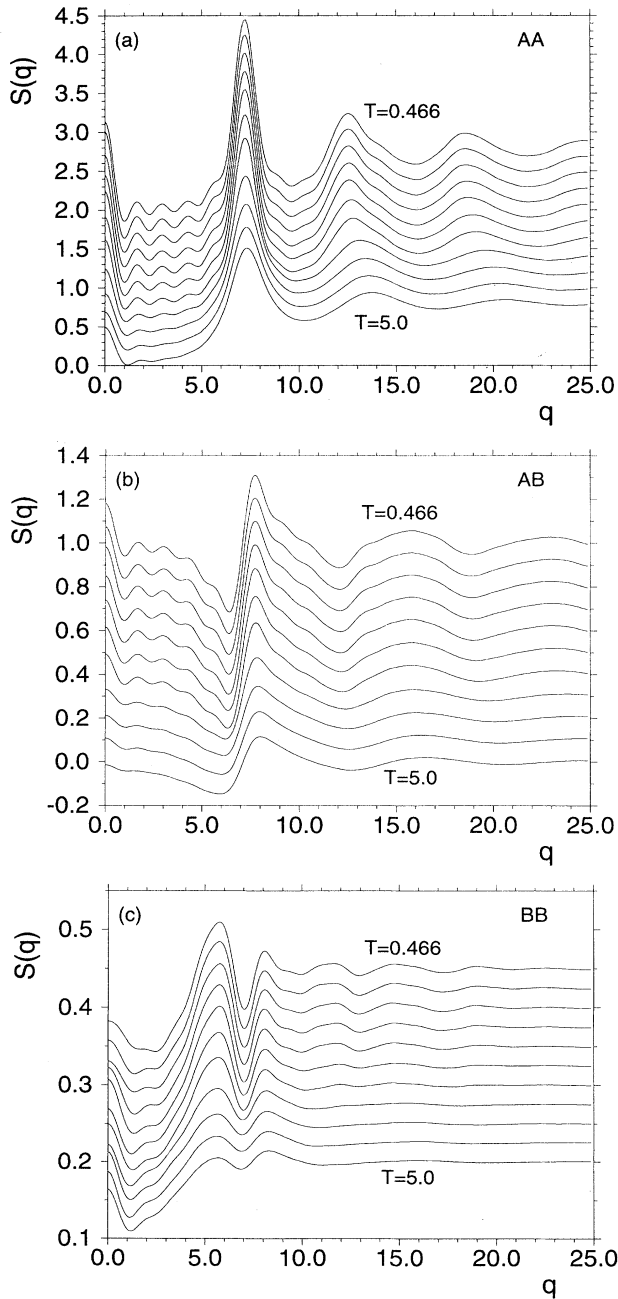


FIG. 1. Structure factor $S(q)$ for all temperatures investigated. For clarity the individual curves have been displaced vertically by $x \times n$ with $n = 0, 1, 2, \dots$ (a) AA correlation, $x = 0.2$; (b) AB correlation, $x = 0.1$; (c) BB correlation, $x = 0.025$.

tures have been displaced vertically (see figure caption for details). At low temperatures (top curves) we clearly see the oscillations mentioned in the previous paragraph. They are most pronounced for small values of q and hardly noticeable for large values of q . In the following we will mainly concentrate on the range of q that is between the first maximum and the first minimum in $S(q)$. As can be recognized from the figure, in this range the amplitude of the oscillations is small and thus this effect can probably be neglected. Note that the pronounced increase of $S(q)$ at $q \rightarrow 0$ should not be taken as an indication that fluctuations on a large distance scale are present but rather reflects the artifacts in the calculated Fourier transform introduced by the discontinuities in the calculated van Hove function as mentioned in the previous paragraph. Since in Ref. [1] we gave evidence that (1) the system is in equilibrium at all temperatures, (2) the thermodynamic quantities like pressure, potential energy, and total energy are a smooth function of temperature, and (3) these quantities did not show any drift *during* the runs it is very improbable that the rise of $S(q)$ for small values of q is an indication of a phase separation. Furthermore, also a visual inspection of the particle configurations excludes this possibility.

We recognize from Fig. 1 that $q_{max}(T)$, the location of the first maximum in $S(q)$, depends only very weakly on temperature. The same holds for $q_{min}(T)$, the first minimum in $S(q)$. In the following we will, among other things, study the temperature dependence of the correlation functions at $q = q_{max}$ and $q = q_{min}$. Since the temperature dependence of q_{max} and q_{min} is so weak we will neglect it altogether and fix the values of the two quantities to their corresponding values at the lowest temperature. From the same figure we can also see that at low temperatures the form of $S(q)$ depends only weakly on temperature. The only thing that changes at these temperatures is that the height of the peaks and the depth of the valleys become *slightly* more pronounced. In Refs. [1] and [7] we have found that at low temperatures the relaxation times show a strong temperature dependence. Since we recognize now that this slowing down is not accompanied by a significant change in the structure factor we can exclude the possibility that the slowing down is associated with some sort of divergence of the correlation length of the pair-correlation function. This observation is in accordance with the underlying idea of MCT that the slowing down of the dynamics is a purely dynamic phenomenon and has nothing to do with some sort of underlying phase transition. However, one has of course to keep in mind that the latter possibility is by no means excluded by our observation of the independence of $S(q)$ on temperature, since it may well be that a *different* quantity indeed shows a diverging correlation length.

In Fig. 2 we show the incoherent intermediate scattering function $F_s(q, t)$ for all temperatures investigated. Figures 2(a) and 2(b) are for the A particles with $q = q_{max} = 7.25$ and $q = q_{min} = 9.61$, respectively, and Figs. 2(c) and 2(d) are for the B particles for $q = q_{max} = 5.75$ and $q = q_{min} = 7.06$, respectively. We see that for *all* temperatures investigated the correlation functions decay

to zero in the long time limit. This means that the fluctuations that were present at time zero have disappeared in the time span of the simulation. Thus this is evidence that the length of the simulation is large enough that the system can come to equilibrium at all temperatures. Other evidence for this was presented in Ref. [1].

For short times the correlators show a quadratic dependence on time, which can be understood by remembering that for short times the motion of the particles is essentially ballistic. For intermediate and long times the correlators at high temperatures (curves to the left) show a relaxation behavior that is similar to a simple exponential decay. This behavior changes when we go to intermediate temperatures ($T \approx 1.0$). There we see that for intermediate times a small shoulder begins to form. This temperature is comparable to the one for which the diffusion constant D and the relaxation times τ started to show [1,7] the asymptotic behavior at low temperature predicted by MCT, i.e., a power law with critical temperature T_c and critical exponent γ [see Eq. (9)]. Thus the qualitative change in the relaxation behavior of the in-

termediate scattering function, e.g., the occurrence of a shoulder, is accompanied with the onset of the asymptotic behavior in D and τ .

When the temperature is lowered even further this small shoulder becomes more pronounced until we observe almost a plateau at the lowest temperature. Thus we find that at low temperatures the correlators exhibit the two step relaxation phenomenon predicted by MCT. We also note that at low temperatures the correlators for the A particles [Figs. 2(a) and 2(b)] show a small bump for times around 14 time units. A similar phenomenon was observed in a recent computer simulation of Lewis and Wahnström of orthoterphenyl [8]. In that work evidence was given that this bump is a finite size effect. A similar bump was also observed in a simulation of a different Lennard-Jones mixture [9], a simulation of a molten salt [10], and a simulation of a colloidal suspension [11]. However, no such feature was observed in simulations with soft spheres [12].

A comparison of the correlators plotted in Fig. 2(a) with those in Fig. 2(b) [or of Fig. 2(c) with Fig. 2(d)]

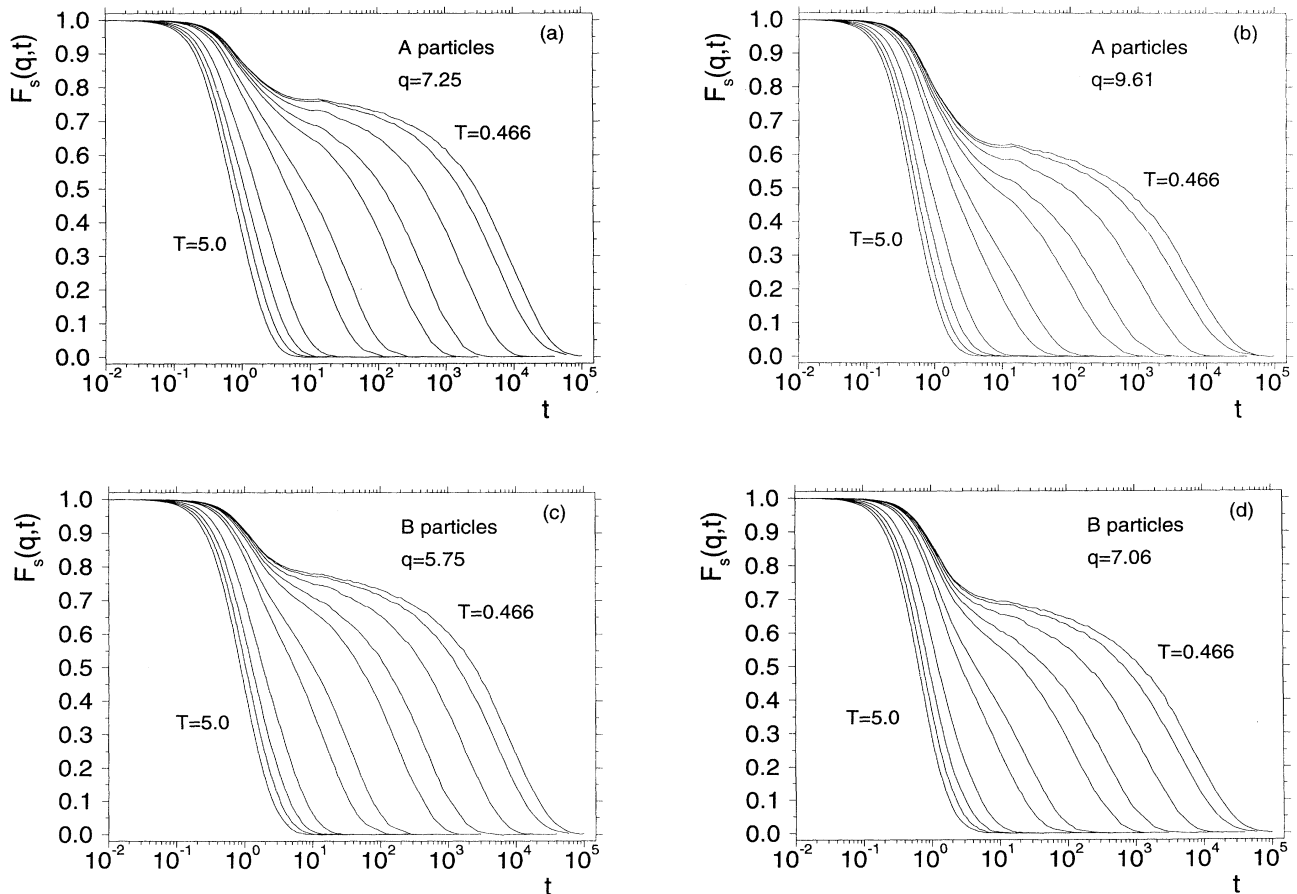


FIG. 2. Incoherent part of the intermediate scattering function $F_s(q, t)$ for all temperatures investigated. (a) and (b): A particles, $q = q_{max} = 7.25$ and $q = q_{min} = 9.61$, respectively. (c) and (d): B particles, $q = q_{max} = 5.75$ and $q = q_{min} = 7.06$, respectively.

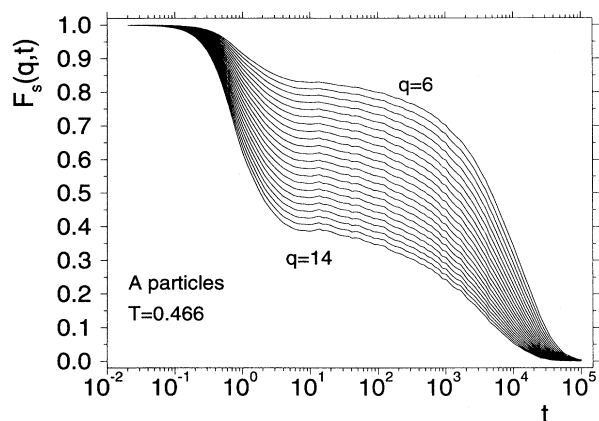


FIG. 3. Incoherent intermediate scattering function $F_s(q, t)$ for the *A* particles at $T = 0.466$. The values of q range from $q = 6.0$ (top) to $q = 14.0$ (bottom) and are given by $q = 6.0 + 0.36n$ with $n = 0, 1, 2, \dots$

shows that the height of the plateau as well as the time scale on which the correlator ultimately decays to zero depend on the value of q . In order to show these two effects more clearly we show in Fig. 3 $F_s(q, t)$ for the *A* particles at $T = 0.466$, the lowest temperature investigated, for values of q in the range $6.0 \leq q \leq 14.0$. From this figure it becomes evident that the height of the plateau depends strongly on the value of q and that also the relaxation time varies by about one order of magnitude in this range of q . MCT predicts the qualitative dependence of this height on q and later on we will present the result of our analyses of this dependence in more detail. Also the investigation of the dependence of the relaxation time on q will be postponed for the moment.

MCT predicts that for low temperatures the correlators should obey the time-temperature superposition principle in the α -relaxation regime, i.e., show a scaling behavior if they are plotted versus rescaled time t/τ , where $\tau(T)$ is the relaxation time for the α relaxation [see Eq. (7)]. To test this prediction of the theory we made such a scaling plot. We defined the α -relaxation

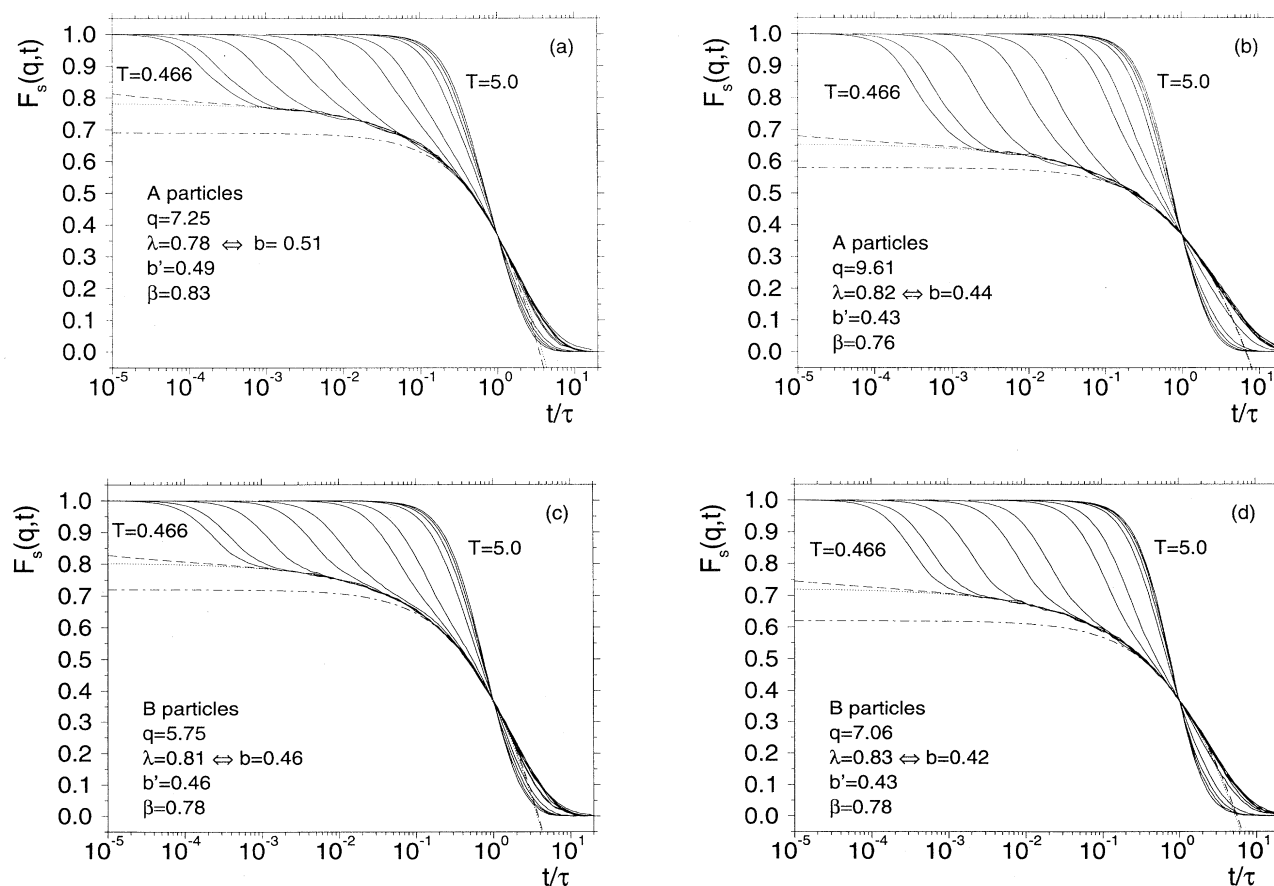


FIG. 4. Incoherent intermediate scattering function $F_s(q, t)$ for all temperatures investigated (solid lines) versus rescaled time. The dashed curve is a fit with a master curve in the β -relaxation regime proposed by MCT (see text for details). The dotted curve is a fit with a von Schweidler law and the chained curve is a fit with a KWW law. (a) and (b): *A* particles, $q = q_{max} = 7.25$ and $q = q_{min} = 9.61$, respectively. (c) and (d): *B* particles, $q = q_{max} = 5.75$ and $q = q_{min} = 7.06$, respectively.

time of a correlator as the time where the correlator has decayed to e^{-1} of its initial value. Note that this kind of arbitrary definition of the relaxation time makes, within the framework of MCT, perfect sense since, due to the time-temperature superposition principle [Eq. (7)], *any* definition of a relaxation time that measures the time scale of the α relaxation is predicted to show the same temperature dependence. In Fig. 4 we show the incoherent intermediate scattering function $F_s(q, t)$ versus this rescaled time for the *A* and *B* particles and for $q = q_{min}$ and $q = q_{max}$. From these figures we recognize that at low temperatures we find indeed a scaling behavior, thus confirming this prediction of the theory.

MCT predicts the functional form of the master function $G(t)$ in the β -relaxation regime [see Eq. (2)]. Thus we tried to fit our master curve with the one predicted by the theory and the best fit we obtained is included in the figures as well (dashed line) [21]. The value of the exponent parameter λ that gave the best fit is given in each figure. We recognize that this fit is very good for rescaled times in the interval $10^{-3} \leq t/\tau \leq 10^0$, thus over a time range spanning about three decades. This time range corresponds to the *late* β regime which is the same as the *early* α regime. From this we conclude that MCT is able to rationalize the master curve in the mentioned relaxation regime.

The theory predicts that the exponent parameter is independent of the type of correlator or the value of q . The values of λ that we obtained are not all equal, but instead have a variation of about 5% for the four correlators. Since the statistical uncertainty with which the fitting procedure can determine any value of λ is about 1% we thus find that for our system the exponent parameter is not constant. However, it should be remembered that the prediction of MCT, that λ is independent of the type of correlator or the value of q , is an *asymptotic* result of the solutions of equations that are only an approximation to the original MC equations, i.e., the equations in which the full q dependence is taken into account. Thus it can be expected that there will be corrections to these asymptotic results. Furthermore, the values of λ we determined depend to some extent on the time range where the fit to determine λ was done and thus an additional, systematic, error might be introduced which we estimate to be of the order of several percent. Thus a small dependence of λ on q or the type of correlator should not be viewed as a failure of the theory.

In order to test this prediction of MCT more extensively we present in Fig. 5 the coherent part of the intermediate scattering function for the *AA*, the *AB*, and the *BB* correlations. The values of q were chosen to be at the maximum of the corresponding structure factors. From this figure we recognize that, in accordance with MCT, these correlation functions also show a scaling behavior in the α -relaxation regime. In the late β -relaxation regime the master curves can again be fitted very well by functional forms predicted by MCT (dashed lines). Thus we find that this prediction of the theory holds also for these types of correlation function. The values of the exponent parameter λ for the different correlators are similar to the ones we presented in Fig. 4. Thus we can conclude

from these two sets of figures that λ is indeed only a weak function of q for those values of q studied or the type of correlator investigated.

For times on the time scale of the late β -relaxation

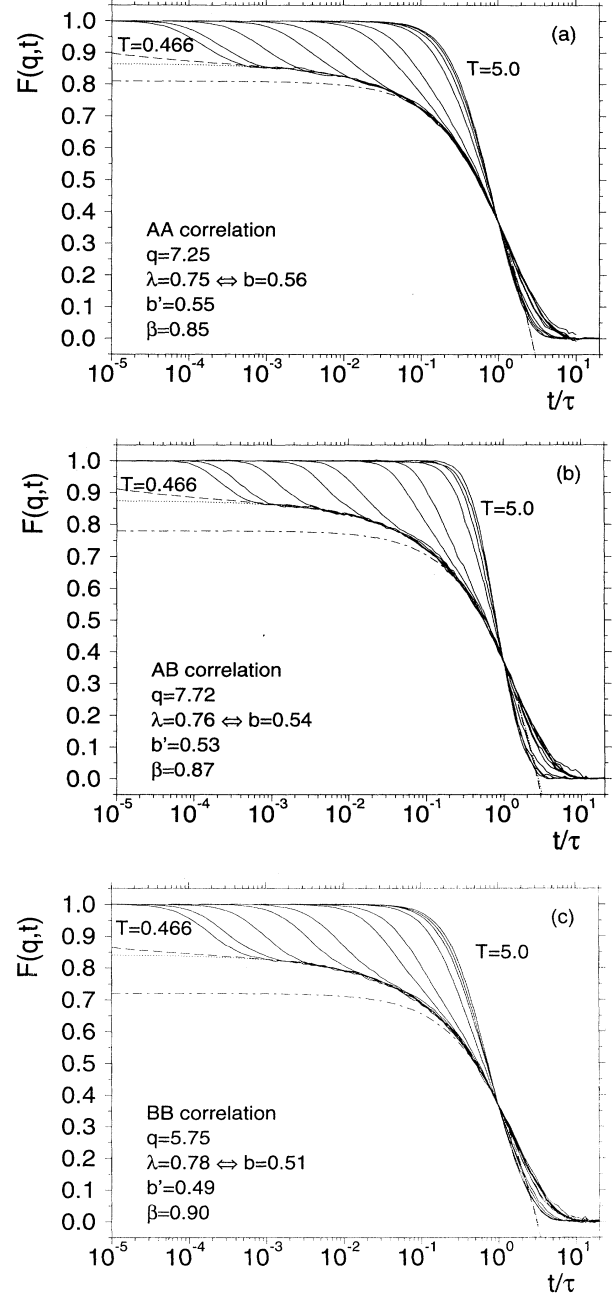


FIG. 5. Coherent intermediate scattering function $F(q, t)$ for all temperatures investigated (solid lines) versus rescaled time. The dashed curve is a fit with a master curve in the β -relaxation regime proposed by MCT (see text for details). The dotted curve is a fit with a von Schweidler law and the chained curve is a fit with a KWW law. (a) *AA* correlation for $q = q_{max} = 7.25$, (b) *AB* correlation for $q = q_{max} = 7.62$, and (c) *BB* correlation for $q = q_{max} = 5.75$.

regime MCT predicts that the theoretical master function is given by a power law, the so-called von Schweidler law [see Eq. (5)]. In previous work we have shown that the master curve of our data can be fitted very well with such a functional form [7,13]. In these papers the von Schweidler exponent b was treated as a fit parameter and it was found that b depended significantly on neither the type of correlator nor the value of q . Because of the one-to-one connection between the von Schweidler exponent b and the exponent parameter λ [see Eq. (6)], this can be seen as evidence that the exponent parameter is almost constant for our system. This observation is therefore in accordance with the findings presented here. The fits with the von Schweidler law are included in Figs. 4 and 5 as well, and we recognize that in the region where the theoretical master curve of MCT is fitting the data well the von Schweidler law does so too. Thus we can conclude that for our system the power law is a good approximation to the theoretical master curve for the whole time range of the late β relaxation and not only very close to the plateau as might be expected *a priori* from the asymptotic nature of the von Schweidler law. Since we have determined the exponent parameter for each of the correlation functions shown, we can compute the von Schweidler exponent b by means of Eq. (6) and compare it with the result from fitting only the power law. In order to distinguish these two quantities we will denote the latter by b' . The values of these quantities are given in Figs. 4 and 5. We find that b and b' are usually very close to each other and can be taken essentially to be equal. Therefore we will report in the following only the value of the von Schweidler exponent as determined from the power-law fit.

For times belonging to the late part of the α -relaxation regime, one can show that the master function predicted by MCT is well approximated by a Kohlrausch-Williams-Watts law [see Eq. (8)]. We have tried to fit this part of the master curve of our data with such a functional form and the result is included in Figs. 4 and 5 as well. Although it is not clearly seen in figures on this scale, these fits are very good at low temperatures and long times and therefore we can conclude that this prediction of the theory holds too. Note that the values of the KWW exponents β are significantly different from the ones of the von Schweidler exponent b' . Thus it is *not* the case that the von Schweidler law can be considered as the short time expansion of the KWW law. One might be tempted to try to fit the *whole* master curve with a KWW law, i.e., the whole α -relaxation regime. However, we found that such a fit is not convincing at all and can therefore be ruled out.

Besides the von Schweidler law, which MCT predicts to be present for the late β -relaxation regime, the theory also predicts that the correlation functions should show a power law, the so-called critical decay, also when *approaching* the plateau [see Eq. (4)]. We have thus tried to fit the data in the early β -relaxation regime with such a functional form but were not able to find any sign of the presence of such a relaxation behavior in this time regime. Inspection of Figs. 4 and 5 shows that this apparent absence of the critical decay is probably due to the

fact that on the time scale at which the critical decay is supposed to be present the correlators seem to be still strongly influenced by the relaxation behavior at short times, where the dynamics is essentially ballistic. This influence will thus make the detection of a critical decay very difficult. We will come back to this point in the next section when we discuss the dynamic susceptibility.

It is interesting to compare our results with the ones that Bengtzelius obtained from a numerical integration of the mode-coupling equations for a monatomic Lennard-Jones system [22]. These results were later improved by Smolej and Hahn [23]. By taking into account essentially the full q dependence of the structure factor and also making a reasonable modeling of the short time dynamics of this system, these authors obtained the full time dependence of the intermediate scattering function. Their results show that the correlation functions are qualitatively very similar to the ones presented in this work. In particular they also show that in the time domain the critical decay is hardly noticeable if the system is not very close to the critical point. In Refs. [1] and [7] we have shown that at the lowest temperature we investigate in this simulation $\epsilon = (T - T_c)/T_c$ is about 0.07. In order to compare this value of ϵ to the ones studied in Ref. [22] one has to remember that Bengtzelius varied the density of the system and not the temperature, as we have done in this work. Thus in the work of Bengtzelius we have $\epsilon = (n_c - n)/n_c$, where n is the particle density. Thus the meaning of the small parameter ϵ in his and our work is clearly not the same. However, we can compare the relaxation behavior of the incoherent part of the intermediate scattering function $F(q, t)$ for q close to the first peak in the structure factor, presented in Fig. 3 of Ref. [22], with the corresponding result of our work. [In Fig. 2(a) we show the relaxation behavior of $F_s(q, t)$ which is qualitatively very similar to that of $F(q, t)$.] From this comparison we conclude that the relaxation behavior at our lowest temperature ($\epsilon = 0.07$) corresponds to a value of ϵ of about 0.0023 in the work of Bengtzelius (curve C of Fig. 3 in Ref. [22]). Smolej and Hahn have shown that in the monatomic Lennard-Jones system the critical decay can be observed only for $|\epsilon| \approx 0.00042$; thus about a factor of 5 ($=0.0023/0.00042$) smaller than the ones accessible in our simulation. Thus if we also take into account that our correlators have a certain amount of noise it is very reasonable that the critical decay becomes completely obscured. Thus we conclude that, for this system and the temperature range investigated, the critical decay is either not present at all or not detectable with data of the accuracy we are able to obtain.

We turn now our attention to the nonergodicity parameter f_c . As we already noted in Fig. 3, f_c depends on q . MCT predicts that for the incoherent intermediate scattering function the nonergodicity parameter shows a Gaussian-like behavior as a function of q . For the coherent intermediate scattering function f_c is predicted to show an oscillatory behavior with oscillations that are in phase with the structure factor $S(q)$. In order to test these predictions we fitted the correlators $F_s(q, t)$ and $F(q, t)$ at the lowest temperature investigated with a von Schweidler law and used the offset as an approximation to

the nonergodicity parameter. The quality of this approximation was tested for the cases presented in Figs. 4 and 5 in that we compared the offset we obtained from the power-law fit with the nonergodicity parameter we obtained when we fitted the full theoretical master curve. In most cases investigated the difference between the two quantities was less than 1% and thus this approximation should be of sufficient accuracy.

In Fig. 6 we show the q dependence of the nonergodicity parameters f_c for the incoherent and coherent intermediate scattering function for the A particles. From this figure we recognize that the f_c for the incoherent part shows indeed a Gaussian-like behavior (upper dotted curve). The nonergodicity parameter of the coherent part (upper solid curve) shows an oscillatory behavior that is in phase with the structure factor $S(q)$ (dashed curve). Also the relative magnitude of the two curves is very similar to the one predicted by MCT [2]. Thus we conclude that the prediction of MCT concerning the q dependence of the nonergodicity parameter of the intermediate scattering function is in qualitative accordance with our data. A similar accordance with this prediction of the theory was reported also from scattering experiments [24] and another computer simulation [14].

Also included in the figure are the amplitudes A of the KWW law we fitted at long times for the incoherent intermediate scattering function (lower dotted curve) and the coherent one (lower solid curve). We recognize that these amplitudes show qualitatively the same behavior as the nonergodicity parameters and can thus serve as a *first* approximation to it. However, we also note that both KWW amplitudes are always smaller than the corresponding nonergodicity parameters. Thus this is evi-

dence that the KWW law which fits the data well on the time scale of the late α relaxation does not fit the data well on the time scale of the early α relaxation.

MCT predicts that the exponent parameter depends only on the structure factor at the transition. From this it follows that the von Schweidler exponent b is independent of the correlator studied, since the two quantities are related by means of Eq. (6). In Figs. 4 and 5 we gave evidence that the exponent parameter λ does not depend strongly on the type of correlator or the value of q . In order to make a more systematic test of this prediction of MCT we determined the von Schweidler exponent b' for various correlation functions and varied q over a large range. Similarly to the nonergodicity parameter, b' was determined by means of a power-law fit to the correlators at the lowest temperature. The range in time for which this fit was good was about three orders of magnitude for small values of q and about two orders of magnitudes for large values of q and we were able to determine b' with an absolute accuracy of about 0.02. Note that the von Schweidler exponent determined in this way is an *effective* von Schweidler exponent, since the fit was done by trying to fit the power-law to the data over a time interval which was as large as possible. Thus it might be that if one restricted the time range over which the fit was done to a smaller interval the value of the exponent would change. However, since the fits were usually quite good over several orders of magnitude in time, a restriction of the time interval would probably not lead to very different values for the exponent.

In Fig. 7 we plot the effective von Schweidler exponent b' , determined in the way explained above, versus q for the incoherent intermediate scattering function for the A and B particles as well as for the coherent one for the AA , AB , and BB correlations. From this figure we recognize that the effective von Schweidler exponent for the

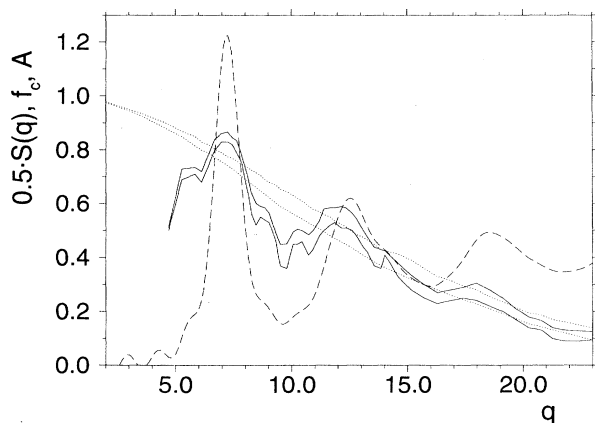


FIG. 6. Nonergodicity parameter f_c for the incoherent (A particles) and coherent (AA correlation) intermediate scattering function (upper dotted line and upper solid line, respectively). The lower solid line and lower dotted line are the amplitudes of a KWW fit at long times to the same incoherent and coherent intermediate scattering function. The dashed line is the structure factor $S(q)$ for the AA correlation divided by 2.0.

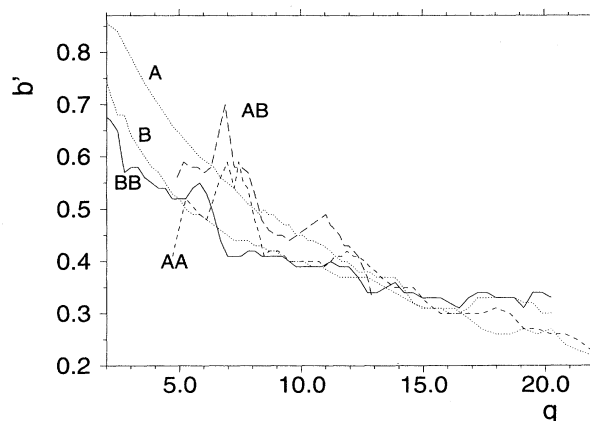


FIG. 7. Effective von Schweidler exponent b' , as determined from a power-law fit, versus q for the incoherent intermediate scattering function for the A and B particles (curves A and B , respectively) and for the coherent intermediates scattering function for the AA , AB , and BB correlations (curves AA , AB , and BB).

incoherent function decreases with increasing q . For the coherent function, AA and BB correlations, b' shows also a general trend to decrease but we see in addition some oscillations which are in phase with the structure factor (see Fig. 1). For the AB correlation b' also shows the trend to decrease but the peaks in the curve cannot be assigned to maxima in the corresponding structure factor. We see that for q in the range shown in the figure the variation of b' between the different curves is quite appreciable and also the systematic dependence on q is clearly visible. At first glance this seems to be in contradiction with our previous findings in which we reported only a weak dependency of b' on q (see, e.g., Fig. 4 in Ref. [7] or the results presented in Figs. 4 and 5). However, it should be noted that in Ref. [7] we focused on the dependency of b' for the incoherent part of the A particles only in the range of q between $6.5 \leq q \leq 9.6$, i.e., the range from about q_{max} to q_{min} . We see from Fig. 7 that in this relatively small range the value of b' indeed does not change a lot and can thus be considered to be almost constant. Only if the von Schweidler exponent is measured over a much larger interval of q is its dependence on q revealed. If the values of b' for $F_s(q, t)$ of the A and B particles are read off at $q = q_{max}$, where q_{max} is the location of the maximum in the *corresponding* structure factor, we find that these values are quite close together. This is in accordance with the finding presented in the context of Figs. 4 and 5. Thus we draw the conclusion from this figure that this *effective* von Schweidler exponent b' depends on q , but that its value at the maximum of the corresponding structure factor is almost independent of the type of correlator.

In Fig. 4 of Ref. [7] we presented a graph in which we plotted $F_s(q, t)$ for the A particles versus $t^{b'}$, where b' was the effective von Schweidler exponent determined for $q = q_{max}$, for values of q ranging from a bit less than q_{max} to values up to q_{min} . If $F_s(q, t)$ is a power law with exponent b' the curves will be straight lines. With this plot we gave evidence that the von Schweidler exponent was essentially independent of q in this range of q . Since we find now that, if q is varied over a larger range, the effective von Schweidler exponent is dependent on q we tried to test if it is possible to describe at least *part* of the late β -relaxation regime with a von Schweidler law with an exponent that is independent of q . MCT predicts that the time range in which the von Schweidler law, with a q -independent exponent, holds should depend on q . Thus it might be that the fitting procedure described above might make use of data over too large a range in time and thus extract an effective exponent b' that is significantly different from the real von Schweidler exponent b . Thus we tried to plot $F_s(q, t)$ for the A particles for q in the range $2.0 \leq q \leq 24.0$ versus $t^{0.49}$. Here the exponent $b' = 0.49$ stems from our fit for $q = q_{max}$ [see Fig. 4(a)]. Unfortunately the resulting plot was not very useful to decide whether a constant value of b' is compatible with the data or not, since noise in the data prevented us from determining reliably those parts of the individual curves where they are straight lines. Only in the range between q_{max} and q_{min} was the statistics good enough to identify clearly the time range where a straight line was present

and this result was already presented in Ref. [7]. Thus the only conclusion we can draw at the moment is that the exponent of the power law, if fitted over a time range as large as possible, is indeed dependent on q . If there does exist a q -dependent time range in which a power law with a constant value of b' fits the data well, this range is not observable within the accuracy of our data, except for q between q_{max} and q_{min} .

As already mentioned in the discussion of Figs. 4 and 5 the von Schweidler exponent b' and the KWW exponent β are significantly different for the correlators discussed in these figures. In order to investigate this effect in more detail we show in Fig. 8 the q dependence of b' and β for the A particles. We clearly recognize that b' is significantly smaller than β for all values of q . Thus this is further evidence that the functional form appropriate to describe the data on the time scale of the late α relaxation is not appropriate to describe the data on the time scale of the late β relaxation.

Also included in the figure is the KWW exponent β for the B particles. The general behavior of this curve is very similar to the one for the A particles except that for a given q the values of β for the B particles is a bit smaller than the one for the A particles. This is in accordance with the prediction of MCT for a binary mixture of soft spheres [18], namely, that the relaxation of $F_s(q, t)$ is more stretched for the small particles than that for the large particles.

We see in Fig. 8 that for large values of q β for the A particles is constant to within the noise. We have observed a similar effect in the case of the β determined from $F(q, t)$ for the AA correlation but *not* in the case of $F_s(q, t)$ (also included in Fig. 8) and $F(q, t)$ for the B particles and BB correlation, respectively. MCT predicts that for large values of q the KWW exponent should approach b as a limiting value [17,18]. From the figure one recognizes that unfortunately the quality of our data

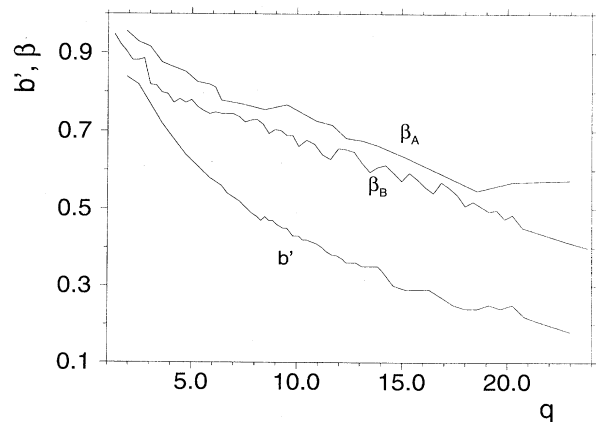


FIG. 8. KWW exponent β determined from the incoherent intermediate scattering function for the A and B particles and the effective von Schweidler exponent b' for the same correlator for the A particles.

is insufficient to test this prediction of the theory.

We now investigate the temperature dependence of τ , the relaxation time of the α relaxation [see Eq. (9)]. In Ref. [7] we showed that for low temperatures the relaxation time of $F_s(q, t)$ for the A and B particles for $q = q_{max}$ shows a power-law dependence on temperature, $(T - T_c)^{-\gamma}$, with a critical temperature $T_c = 0.432$, which is very close to the one we found for the diffusion constant, which was $T_c = 0.435$. In Fig. 9 we show the relaxation times versus $T - T_c$ for the following correlation functions: $F_s(q, t)$ for the A and B particles (squares and triangles pointing down, respectively) at $q = q_{max}$ and $q = q_{min}$ (of the corresponding structure factors), $F(q, t)$ for the AA and BB correlation (circles and diamonds, respectively) at $q = q_{max}$ and $q = q_{min}$, and $F(q, t)$ for the AB correlation (triangles pointing up and star) for $q = q_{min1}$, $q = q_{max}$, and $q = q_{min2}$. In each case the filled and open symbols correspond to $q = q_{max}$ and $q = q_{min}$, respectively and the star is for $q = q_{min2}$. The last three values of q are the location of the first minimum, the first maximum, and the second minimum in $S(q)$ for the AB correlation [see Fig. 1(b)] and have the values 6.40, 7.72, and 12.05. From Fig. 9 we recognize that at low temperatures the various relaxation times can all be fitted well by a power law with the same critical temperature $T_c = 0.430$. Thus the critical temperature for all correlators shown in this figure is very close to the one we found for the diffusion constant of the A and B particles, thus giving evidence that at this temperature the system becomes nonergodic. This transition from

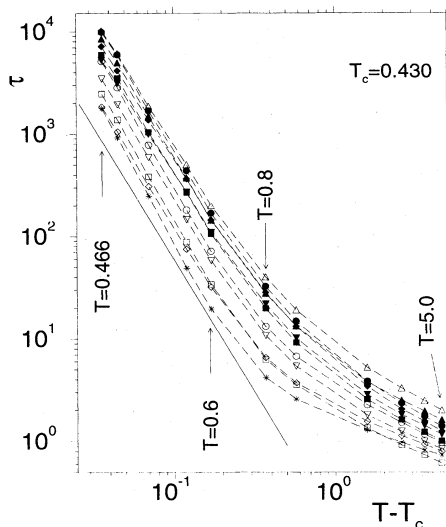


FIG. 9. Relaxation time τ versus temperature for various correlators. Squares and triangles pointing downwards: $F_s(q, t)$ for A and B particles, respectively. Circles and diamonds: $F(q, t)$ for AA and BB correlation, respectively. Triangles pointing upwards and stars: $F(q, t)$ for AB correlation. Filled and open symbols are for $q = q_{max}$ and $q = q_{min}$, respectively. Solid line: power law with exponent 2.6.

an ergodic to nonergodic behavior can of course only be expected if the hopping processes will not restore the ergodic behavior of the system. We have given evidence above that in the temperature interval investigated in this work these hopping processes are not present. However, it may very well be that for temperatures even closer to T_c the dynamics of the system is influenced by the hopping processes and thus no real transition to a nonergodic behavior will be observed.

The critical exponent γ with which the various relaxation times diverge at T_c is found to be essentially independent of the type of correlation function, in accordance with MCT. Its value is around 2.6, which is essentially the same as the exponent predicted by the theory if one uses the connection between γ and the von Schweidler exponent b and a value of b of 0.51 (leading to $\gamma = 2.7$) which was found for b from $F_s(q, t)$ for the A particles close to q_{max} (see Fig. 4).

We also note that in Ref. [1] we showed that at low temperatures the diffusion constant for the A and B particles were better fitted by a power law than by a Vogel-Fulcher law. We tried to fit the relaxation times presented in Fig. 9 also with a Vogel-Fulcher law and found that for these quantities the Vogel-Fulcher law was able to make a good fit which covered a slightly larger temperature range than the power law presented in Fig. 9 is able to do. Thus from the point of view of a mere fitting function the Vogel-Fulcher law has to be preferred for this data. However, this should not be taken as an argument for not using a power law to fit the data to extract an exponent for comparison with other exponents. The objective of the present work is to test whether MCT is able to give a consistent picture of the whole variety of data on the low temperature dynamics of the system. The fact that one subset of the data can be better described by a functional form not compatible with the theory should not be seen as evidence against the validity of the theory.

From Fig. 9 we also recognize that the relaxation times for the various correlation functions depends strongly on the correlator. In particular, we see that at the lowest temperature we find a variation of about a factor of 7 for the τ of the various correlation functions presented here. In this context it is interesting to test another prediction of MCT, the so-called α -scale universality [see Eq. (9)]. In order to do this we defined a new relaxation time τ' by requiring that at time τ' the correlator has decayed to 0.15. The reason for this definition is that with the old definition, for which the correlator is supposed to have decayed to $e^{-1} \approx 0.368$, the relaxation times τ do not measure the α -relaxation time if q is large, since for large values of q the height of the plateau becomes smaller than 0.368 (see Fig. 3).

In Fig. 10 we show the q dependence of τ' determined from $F(q, t)$ for the AA correlation for all temperatures investigated. From this figure we recognize that at low temperatures (top curves) the curves corresponding to different temperatures are just shifted vertically. This is exactly what is expected if the α -scale universality holds [see Eq. (9)]. Thus at low temperature the q dependence of each curve is given by $C(q)$ and the vertical shift comes from the strong temperature dependence of the factor

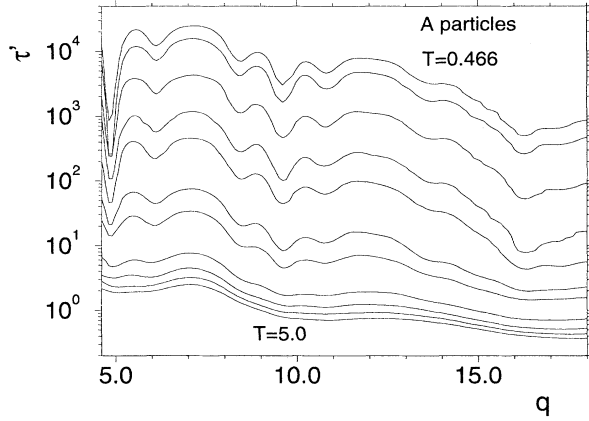


FIG. 10. Relaxation time τ' (see text for its definition) for the coherent intermediate scattering function for the AA correlation versus q for all temperatures investigated.

$$(T - T_c)^{-\gamma}.$$

In the figure we recognize that at low temperature the individual curves show quite a few minima and maxima. Some of them can be identified with the minima and maxima in the structure factor $S(q)$ [see Fig. 1(a)]. The other extrema can be traced back to the little oscillations in the structure factor, which we said was a finite size effect. Thus these other extrema in Fig. 10 should presumably also be considered to be a finite size effect. It is remarkable that the q dependence of the relaxation time found here is qualitatively similar to the one predicted by MCT [18] showing that also this aspect of the theory seems to be, at least qualitatively, correct. A similar agreement is observed for the relaxation times of the incoherent intermediate scattering function.

Note that the ratio of the relaxation time at $q = q_{max} = 7.25$ and at $q \approx 16$, the second minimum in $S(q)$, is about 50. Thus this shows even more dramatically than it did in Fig. 9, that the relaxation times are dependent on q . A similar q dependence was also found in experiments by Mezei *et al.* on CKN [25].

B. Dynamic susceptibility

In this subsection we test the prediction of MCT for the imaginary part $\chi''(\omega)$ of the dynamic susceptibility $\chi(\omega)$. This quantity can be obtained by taking the time Fourier transform of the intermediate scattering function and multiplying it by the frequency ω . Since in this work the intermediate scattering function extends, at the lowest temperature, over almost seven decades in time the computation of the Fourier transform is not a trivial task and great care has to be taken in order to avoid the generation of spurious features in $\chi(\omega)$, which in turn might prevent the testing of certain predictions of the theory. In order to overcome this problem we parametrized the intermediate scattering function with a spline under tension [26] and computed the Fourier transform of the latter

by means of the Filon algorithm [27].

In Fig. 11 we show the dynamic susceptibility $\chi''_s(q, \omega)$ computed from $F_s(q, t)$ for the A particles at $q = q_{max}$ and $q = q_{min}$ and all temperatures investigated. We recognize that at high temperatures $\chi''_s(q, \omega)$ is just a single peak located at microscopic frequencies. The form of this peak is approximated very well by a Lorentzian. On lowering the temperature this peak starts to split into two peaks. The first one stays at microscopic frequencies and the second one moves quickly to small frequencies with decreasing temperature. Thus we observe nicely how the α peak separates from the microscopic peak. Note that this splitting into two peaks looks strikingly similar to the result of a theoretical calculation with a schematic model of MCT, as a comparison of Fig. 11 with Fig. 2 of Ref. [28] shows. Thus the theory is able to describe this effect at least qualitatively. By comparing Fig. 11(a) with Fig. 11(b) we recognize that at high temperatures the height of the microscopic peak does not depend strongly on q . This is not the case at low temperatures where this height for $q = q_{max}$ is about 30% smaller than the one for $q = q_{min}$. On the other hand, just the opposite

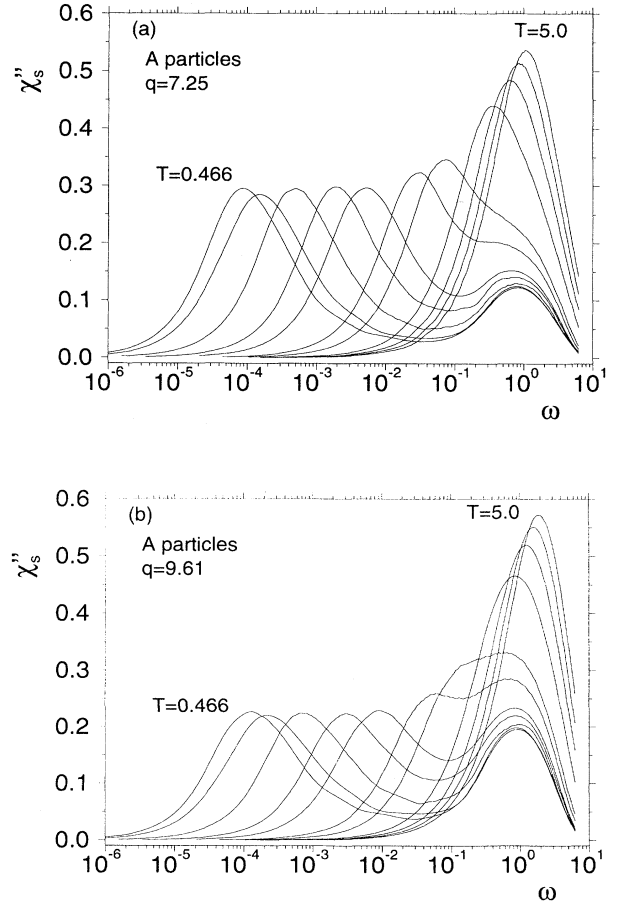


FIG. 11. Dynamic susceptibility $\chi''_s(q, \omega)$ for the A particles versus ω and all temperatures investigated. (a) $q = q_{max}$; (b) $q = q_{min}$.

trend is observed for the height of the α peak, which at low temperatures is significantly larger for $q = q_{max}$ than for $q = q_{min}$. All these observations can be easily understood by remembering that the height f_c of the plateau in the intermediate scattering function depends on q . Since the height of the α peak is proportional to this height and the height of the microscopic peak is proportional to $1 - f_c$, the above described dependence of the heights of the α and microscopic peak follows directly from the q dependence of the nonergodicity parameter f_c .

From Fig. 11 we also recognize that at low temperatures the shape of the α peak does not depend on temperature. This is the consequence of the time-temperature superposition principle predicted by MCT, which we showed to hold very well for this system (see Figs. 4 and 5). In order to investigate this property more closely we plot in Fig. 12 the dynamic susceptibility for the A particles for $q = q_{max}$ scaled by its value at the maximum versus the scaled frequency ω/ω_{max} , where ω_{max} is the location of the α peak. From this figure we recognize that the shape of the peak changes when we go from high to intermediate temperatures. This change seems to be most pronounced on the high frequency side of the peak whereas the low frequency side seems to be essentially independent of temperature in the whole range of temperatures investigated. For intermediate and low temperatures also the high frequency side of the peak does not change with temperature. The only change we observe in the curves when the temperature is lowered is that the high frequency wing of the peak extends to higher and higher rescaled frequencies before the curves turn up again to the microscopic peak. Thus this plot shows that the time-temperature superposition principle holds very well for this system.

It is interesting to compare this figure with results from a depolarized light-scattering experiment on orthoterphenyl [29]. Figure 15 of Ref. [29] shows how this type of plot looks when the time-temperature superposition

principle supposedly does not hold. In that figure the curves for different temperatures fall also onto a master curve on the low frequency side of the α peak. However, on the high frequency side of the peak a clear dependency of the curves on temperature is observed, in contrast to our findings presented in Fig. 12.

A more quantitative way to show that the time-temperature superposition holds for a given correlator is to measure the full width at half maximum (FWHM) of the α peak and to investigate the temperature dependence of this quantity. We define the FWHM as the ratio of the two frequencies at which a horizontal line at half the height of the peak intersects the low and high frequency wings of the peak. Note that the determination of the FWHM at intermediate temperatures is not always possible as can be seen, e.g., from Fig. 12 since at these temperatures the microscopic and the α peak are too close together and thus there is no well separated α peak. We determined the FWHM for those 11 correlators for which we also presented the relaxation times as a function of temperature in Fig. 9 and show this quantity as a function of temperature in Fig. 13. In order to spread the temperature scale at low temperatures we plot $T - 0.435$ on a logarithmic scale. Note that this representation has nothing to do with some sort of critical behavior but is only a convenient way to present the data. The meaning of the symbols is the same as in Fig. 9. We recognize that at high temperatures the FWHM is relatively small for all correlators. It is interesting to note that at these temperatures the FWHM's of the correlators for the AB correlation functions (triangles pointing up and star) are significantly smaller than the ones of the other correlators. Thus we conclude that at high temperatures the relaxation behavior for the former, being non-Debye, is different from the one of the latter, which are of Debye type.

On lowering the temperature the FWHM increases significantly up to $T = 2.0$. For temperatures in the range

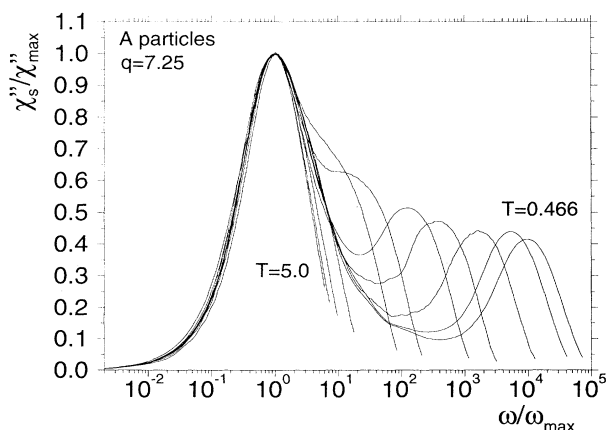


FIG. 12. Dynamic susceptibility $\chi''_s(q, \omega)$ scaled by χ''_{max} for $q = q_{max}$ for the A particles versus rescaled frequency ω/ω_{max} for all temperatures investigated.

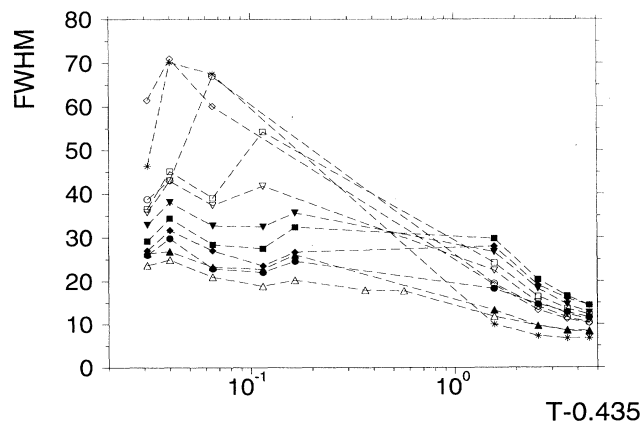


FIG. 13. Full width at half maximum of the α peak for various dynamic susceptibilities (see text). The meaning of the symbols is the same as in Fig. 9.

$2.0 > T > 0.6$ we are not able to measure the FWHM because of the above mentioned problem. (The only correlator for which we have data even in this temperature interval has a microscopic peak which is much smaller than the α peak and therefore does not interfere significantly with the latter.) For temperatures $T \leq 0.6$ the FWHM can be considered constant to within the noise of the data. Thus this is further evidence that the time-temperature superposition principle holds for all correlators investigated here.

MCT predicts that in the time dependence of the function $G(t)$ in Eq. (1) only the ratio t/t_ϵ enters [see Eq. (2)], where t_ϵ is the time scale of the β relaxation. Consequently, the time Fourier transform of $G(t/t_\epsilon)$ will depend only on the ratio ω/ω_ϵ . Thus this prediction can be tested by scaling the dynamic susceptibility by its value at the minimum between the α peak and the microscopic peak and plotting it versus ω/ω_ϵ , where ω_ϵ is the location of this minimum. We have done this for $\chi''_s(q, \omega)$ for $q = q_{max}$ for the A particles. The resulting scaling plot is shown in Fig. 14 for all those temperatures for which a minimum could be identified, i.e., $T \leq 0.8$. From this figure we recognize that in the vicinity of the minimum the curves indeed fall onto a master curve. Thus the scaling behavior predicted by MCT holds for this correlation function. We made the same type of scaling plot also for the other ten correlation functions mentioned above and found in all cases that at low temperatures they fell onto a master curve.

Also included in Fig. 14 (dashed line) is a fit with the time Fourier transform of the theoretical master curve predicted by MCT for the time Fourier transform of $G(t)$.

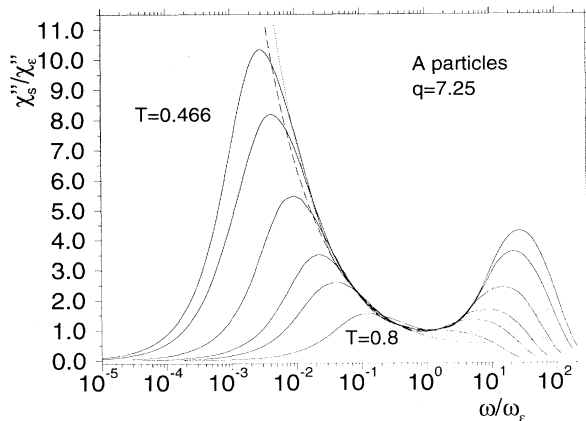


FIG. 14. Solid lines: dynamic susceptibility $\chi''_s(\omega)$ for $q = q_{max}$ for the A particles scaled by its value at the minimum versus frequency scaled by the location of this minimum. Dashed line: Fit with theoretical master curve from MCT with exponent parameter λ fixed to the value determined from the corresponding fit in the time domain [see Fig. 4(a)]. Dotted curve: Fit with the theoretical master curve from MCT with λ as free fit parameter.

This theoretical curve depends again on the exponent parameter λ which we fixed to the value we found for it from the fit of the theoretical master curve in the time domain, i.e., to $\lambda = 0.77$ (see the discussion in the context of Figs. 4 and 5). This theoretical master curve was computed by taking into account the first few correction terms to the so-called interpolation formula [30]. We recognize from Fig. 14 that for rescaled frequencies to the right of the minimum the fit with the theoretical master curve is not good at all. This is another manifestation of the fact stated earlier that for this system MCT does not describe well the early part of the β -relaxation regime (see the discussion in the context of Figs. 4 and 5). A similar discrepancy between the fit of the spectrum with master curves proposed by MCT and experimental data was discussed in Ref. [31] where it was argued that the reason for this discrepancy was the presence of the so-called boson peak.

We see from Fig. 14 that the theoretical master curve does not even give a very satisfactory fit to the master curve to the *left* of the minimum, i.e., where the von Schweidler law should be observed. The deviation of the theoretical curve from the master curve of the data should, however, *not* be considered as a flaw of the theory. We have shown in Fig. 4(a) that in the time domain the theoretical curve gives an excellent fit to the master curve over several decades in time. If we find now that the corresponding Fourier transforms do not look very similar to each other this has to be viewed as an unpleasant property of the Fourier transformation. The problem is that the Fourier transform of the theoretical master curve was obtained by Fourier transforming a series expansion (in powers of t/t_ϵ) of the theoretical master curve in the time domain. The dynamic susceptibility from the simulation, however, was obtained by taking the Fourier transform of the time correlation function. Therefore the dynamic susceptibility computed in this way will include in the vicinity of the minimum also contributions from times which are *not* in the β -relaxation regime. Therefore it cannot be expected that if a theoretical master curve fits the data well over a certain number of decades in time then also the corresponding Fourier transform will match over the same number of decades in frequency. This effect has also been observed in Ref. [30] where it was found that the theoretical master curve approximated the solution of a schematic model [4] well over 5.5 decades in time, but that the corresponding Fourier transforms matched only over four decades in frequency.

We also point out that the above mentioned effect might make the determination of the exponent parameter λ from measurements of the susceptibility somewhat problematic. The difficulty arises from the fact that one usually tries to obtain a good fit over a frequency interval that is as large as possible. In doing that, one might severely overestimate the range in frequency for which the fit with the theoretical master curve is supposed to hold, which in turn may lead to a wrong value of the exponent. In order to illustrate this we have tried to make a fit to the master curve with a theoretical master curve in which the exponent parameter was a free fit parameter. The result of this fit is included in Fig. 14 as well (dotted

line) and we recognize that now the high frequency wing of the α peak is fitted quite well by the theoretical master curve. The value of λ we obtained was 0.74. At first glance the difference of this value from the optimal one as determined from the time domain ($\lambda = 0.78$) does not seem to be large. However, when we used this new value of λ in order to make a fit in the time domain, the resulting fit was significantly inferior to the one presented in Fig. 4(a). Thus this value of λ is not compatible with the data from the time domain. Hence, if we had access only to the susceptibility data, we probably would have determined an incorrect value of λ . It has to be emphasized, however, that the determination of λ from data in the frequency domain might be much less problematic if the theoretical master curve gives a good fit on *both* sides of the minimum.

Apart from the correlator investigated in the context of Fig. 14 we tested whether the other ten correlation functions mentioned earlier also showed a master curve when scaled in the appropriate way, i.e., by χ''_e and ω_e , and found this to be indeed the case. MCT predicts that these various master curves should be identical, since they are all related to the time Fourier transform of the function $G(t)$ [see Eq. (1)], which is predicted to be independent of the correlator. In order to check this prediction of the theory we show in Fig. 15 the susceptibilities at $T = 0.466$, the lowest temperature investigated, scaled by its value at the minimum, versus ω/ω_e . We recognize from this figure that the overall form of the curves, e.g., the height of the α peak or that of the microscopic peak, for the various correlators is very different and thus this prediction of the theory is clearly not a trivial one. We also see that on the left hand side of the minimum most of the curves follow a master curve and thus the prediction of the theory is confirmed for these correlators. However, there are also a few curves which do not fall onto this

master curve. These curves are found to be the ones for which the nonergodicity parameter f_c is relatively small, and thus the ones with values of q which are close to the first minimum. This part of the master curve, i.e., left from the minimum, corresponds to the von Schweidler regime. Moreover, it is known [32] that corrections to the leading asymptotic results of the theory for the von Schweidler law are large if the nonergodicity parameter is small. Thus the deviation of small f_c curves from the master curve can be rationalized within the context of the theory.

For rescaled frequencies to the right of the minimum we recognize that the curves seem to cluster around *two* different “master” curves. A closer analysis of what type of correlator belongs to which bunch of curves showed that the lower bunch of curves all belong to correlators for the incoherent and coherent part of the intermediate scattering function of the B particles. Since we are not aware of any predictions of MCT on the size dependence of the corrections to this master curve we cannot offer any reason for this behavior and thus only report the observation.

Since MCT predicts that at a given temperature the function $G(t)$ is, for a given system, independent of the correlator, its time Fourier transform should also be independent of the correlator. In particular, this means that the location of the minimum in the susceptibility should be independent of the correlator as well. In order to test this prediction of the theory we present in Fig. 16 the susceptibilities for the 11 correlators mentioned above for the lowest temperature studied in this work, i.e., $T = 0.466$. From this figure we see that the form of the various curves depends strongly on the type of correlator. However, to within the noise of the data the location of the minimum is independent of the type of correlator, thus confirming this prediction of the theory.

The theory predicts that χ''_e , the value of the dynamic susceptibility at the minimum, should show a square-root dependence as a function of temperature [see Eq. (12)].

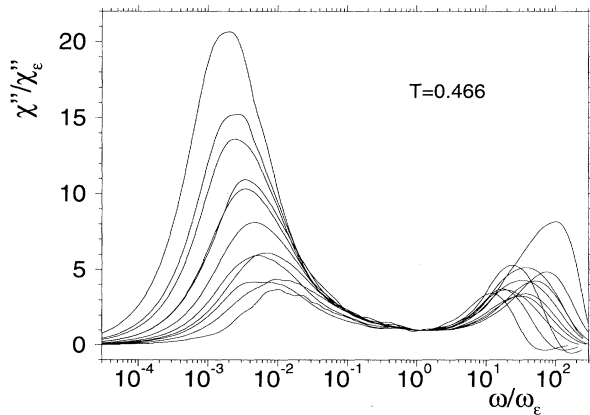


FIG. 15. Dynamic susceptibility $\chi''(q, \omega)$ for the 11 correlators investigated (see Fig. 9 for details) scaled by its value at the minimum versus frequency scaled by the location of this minimum.

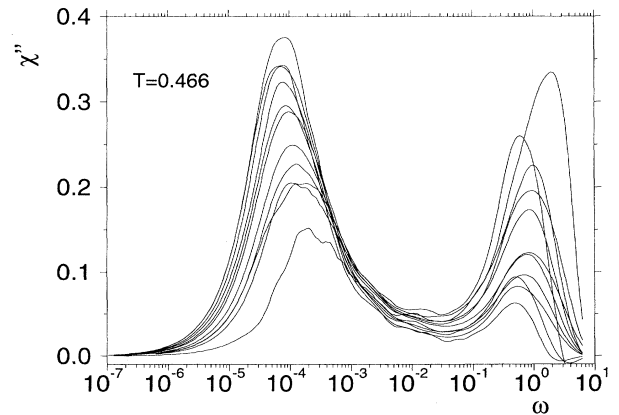


FIG. 16. Dynamic susceptibility $\chi''(q, \omega)$ for various correlators (see text for details) at the lowest temperature $T = 0.466$.

This prediction can easily be tested by plotting $(\chi''_{\epsilon})^2$ versus T . If the prediction of MCT is correct one should find straight lines which intersect the temperature axis at $T = T_c$. We made this plot for the 11 correlators mentioned earlier and found that the curves were not straight lines, thus contradicting the prediction of the theory. We also tested whether a different exponent than 2.0 would lead to straight lines in this type of plot and found that an exponent around 1.25 was indeed able to do so. Interestingly enough the extrapolations of the resulting straight lines to lower temperatures intersect the temperature axis all around the temperature $T = 0.438$, which is very close to the critical temperatures we found for the diffusion constant [1,7] or the relaxation times (see Fig. 9), which were 0.435 and 0.430, respectively. This result is presented in Fig. 17, where we plot the $\chi''_{\epsilon}(T)$ versus $T - T_c$ in a double logarithmic way for all the 11 correlators investigated. The solid line is a power law with exponent 0.8=1/1.25, showing that the slope of the various curves is close to this value. (If the prediction of MCT were correct this slope should be 0.5.)

The theory also predicts that ω_{ϵ} and χ''_{ϵ} are connected via a power law and that the exponent is the critical exponent a from MCT [see Eq. (14)]. In Fig. 18 we show that χ''_{ϵ} and ω_{ϵ} are indeed related by a power law but that the exponent is close to unity (solid line) and thus much larger than the one predicted by MCT for this system (which, with $b \approx 0.49$, would be around 0.28). Note that an exponent of 1.0 is expected if we assume that the power law seen to the right of the minimum is not the one predicted by MCT but just the low frequency wing of a microscopic peak that is the Fourier transform of a process whose integral in the time domain is finite. One possibility of such a process is thus a Debye-like process. Since we have already seen earlier (see the discussion in the context of Figs. 4 and 5) that it may well be that the critical decay predicted by MCT is, at the temperatures

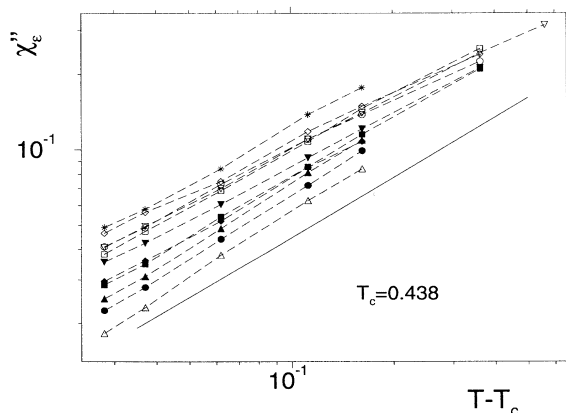


FIG. 17. χ''_{ϵ} , the value of the dynamic susceptibility at the minimum, versus $T - T_c$ for 11 correlators (see Fig. 9 for details) in a double logarithmic plot. Solid line: power law with exponent 0.8.

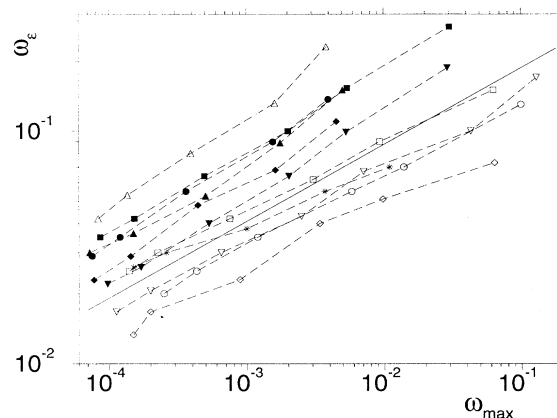


FIG. 18. χ''_{ϵ} , the value of the dynamic susceptibility at the minimum, versus ω_{ϵ} , the location of this minimum for 11 correlators investigated (see Fig. 9 for details). Solid line: power law with exponent 1.0.

investigated here, still severely disturbed by the Debye-like relaxation behavior occurring at short times, this explanation is in accordance with our previous finding.

A further test of the theory is to investigate the connection between the frequency ω_{ϵ} and ω_{max} , the frequency of the α peak. MCT predicts this connection to be also a power law but this time with an exponent $b/(a + b)$ which, assuming $b \approx 0.49$, is around 0.64 [see Eq. (15)]. Figure 19 shows that for our system the power law is indeed observable but that the exponent is around 0.33 (solid line). This value can again be understood by assuming that the value of a is 1.0, or in other words that the high frequency wing of the minimum is just the low frequency part of the Debye-like microscopic peak, thus giving an exponent of 0.33.

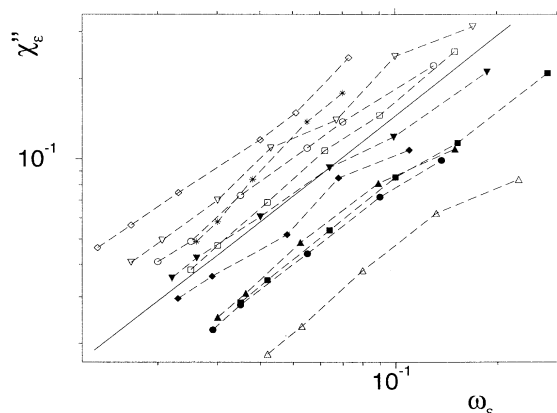


FIG. 19. ω_{ϵ} , the location of the minimum in the dynamic susceptibility, versus ω_{max} , the location of the α peak, for 11 correlators investigated (see Fig. 9 for details). Solid line: power law with exponent 0.33.

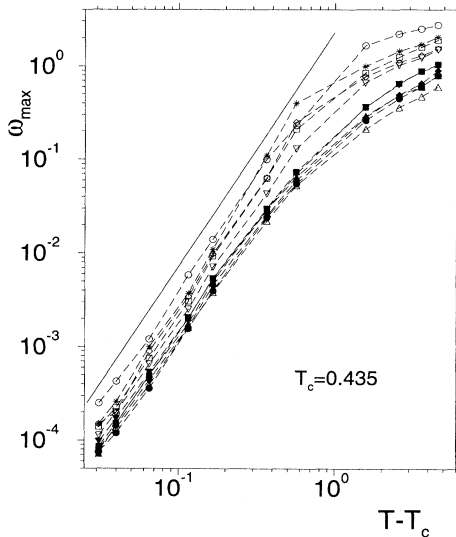


FIG. 20. Temperature dependence of ω_{max} , the location of the α peak, for 11 correlators investigated (see Fig. 9 for details). Solid line: power law with exponent 2.5.

To conclude we test whether the prediction of the theory that ω_{max} should show a power-law dependence on temperature with an exponent γ [see Eq. (13)] holds for this system. We tried to fit the low temperature behavior of ω_{max} with such a functional form and the result is presented in Fig. 20. From this figure we recognize that the power law predicted by the theory indeed holds and that also the exponent is in accordance with the one which follows from the prediction of MCT with the von Schweidler exponent $b \approx 0.49$, giving $\gamma = 2.7$. The critical temperature is the same as the one found for the constant of diffusion [1,7] and very close to the one found for the relaxation times and the one for χ''_e , which is in accordance with the theory.

V. SUMMARY AND CONCLUSIONS

We have presented the results of a large scale molecular dynamics computer simulation of a supercooled binary Lennard-Jones mixture. The goal of our investigation was to test whether MCT is able to give a correct description of the dynamics of this simple liquid at low temperatures. In contrast to our earlier paper [1] in which we mainly concentrated on the investigation of the diffusion constant and the van Hove correlation function, we focus in this work on the intermediate scattering function and the dynamic susceptibility. This allows us to perform additional tests in order to investigate whether MCT is able to describe the low temperature dynamics of our system. The main difference between the work reported here and earlier studies of the dynamics of supercooled liquids [8–12,14,15] is that (i) great care has

been taken to equilibrate the system at *all* temperatures investigated, since only in this case is a comparison with MCT, an *equilibrium* theory, justified, and (ii) *many different* predictions of the theory are investigated in order to see whether the theory actually gives a *self-consistent* description of the low temperature dynamics of simple liquids.

We find that at low temperatures $F_s(q, t)$ and $F(q, t)$, the incoherent and coherent intermediate scattering function, respectively, show the two step relaxation process predicted by MCT. By scaling the intermediate scattering function by the α -relaxation time $\tau(q, T)$ we find that the correlators for intermediate and low temperatures fall onto a master curve in the α -relaxation regime. Thus the time-temperature superposition principle predicted by MCT holds for this system. As predicted by the theory the early part of this master curve, which in the language of MCT corresponds also to the late β -relaxation regime, is fitted very well by a power law, the so-called von Schweidler law. Also the functional form predicted by MCT for the master curve in the β -relaxation regime, the so-called β correlator, which takes into account the corrections to the von Schweidler law, gives a very good fit to the master curve in the region of the late β relaxation. From the point of view of the quality of the fit, the two functional forms can be considered as equally good. Thus for those correlators investigated the corrections to the von Schweidler law do not seem to be very important for this system. Computing the von Schweidler exponent b from the exponent parameter λ and comparing it to the exponent b' as determined from the power-law fit we find that b and b' are very close together for those correlators and values of q for which we made both kinds of fits. Thus we can take b' as a substitute for b and, because of the connection between b and λ [see Eq. (6)], investigate the dependence of λ on q and the type of correlator by the investigation of the dependence of b' on these quantities. For values of q in the range of q_{max} and q_{min} , the location of the first maximum and the first minimum of the structure factor, respectively, the exponent b' shows only a weak dependence on q or the type of correlator [7]. If q is varied over a larger range, however, one finds that b' depends on q , which is in contradiction with the prediction of MCT. On the other hand, it has to be remembered that for q very small and q very large the theory predicts the existence of correction terms to the β correlator and thus these apparent deviations might just be the result of these correction terms.

For the early part of the β -relaxation regime the theory predicts that the correlators should show a different power law, the so-called critical decay [see Eq. (4)]. We do not find any hint that, in the temperature range studied, our system shows such a time dependence. Rather we find that the fast Debye-like relaxation behavior at short times goes directly over to the slow decay of the late β -relaxation regime. Thus it might be that for our system the critical decay is just not existent at all or that, in the temperature range we are able to investigate, the critical decay is not visible because of the interference with the dynamics at short times. It is interesting to note that the numerical solution of the mode-coupling

equations in which the full q dependence and a reasonable short time behavior were taken into account showed that the observation of the critical decay is very difficult if the thermodynamic state of the system is not very close to the critical point of MCT [22,23]. Thus the fact that we are not able to see the critical decay should not necessarily be seen as a failure of the theory.

It is interesting to note that the critical decay seems to be more readily observable if the dynamics of the particles is not Newtonian but stochastic. This has been observed, e.g., in a simulation by Löwen *et al.* of a colloidal suspension in which the two different types of dynamics were compared [11]. It was found that the stochastic dynamics led to a relaxation behavior for which the approach to the plateau is much slower than the one for the Newtonian dynamics. Also a recent simulation by Baschnagel of a polymer system with stochastic dynamics showed a very slow approach to the plateau and it was demonstrated that in this time region the correlators could be fitted well with a power law [33]. Very recently Götze and Sjögren were able to show within the framework of MCT that a stochastic dynamics will lead to a relaxation behavior of the correlators for which the critical decay is more easily observable than for a Newtonian dynamics, thus offering a theoretical explanation for these observations [34].

The late part of the α -relaxation regime can be fitted very well with a KWW law, in accordance with MCT. The exponent β of the KWW law is significantly different from b , the exponent of the von Schweidler law. Thus we conclude that, in accordance with the theory, the von Schweidler law is not the short time expansion of the KWW law.

The height of the plateau, i.e., the nonergodicity parameter f_c , is strongly dependent on the wave vector q . As predicted by the theory the nonergodicity parameter of $F_s(q, t)$, often also called the Lamb-Mössbauer factor, shows a Gaussian-like decay in q . Also the q dependence of the nonergodicity parameter for $F(q, t)$, the so-called Debye-Waller factor, is in qualitative accordance with the prediction of the theory, in that it shows an oscillatory behavior which is in phase with the structure factor.

The α -relaxation time $\tau(T)$ shows for all correlators investigated at low temperatures a power-law dependence on T , as predicted by the theory. The critical temperature, as well as the critical exponent γ , is independent of the correlator. The critical temperature is very close to the one we determined for the diffusion constant for both types of particles [1,7], which is also in accordance with the prediction of the theory. The critical exponent γ of the power law of $\tau(T)$ and the von Schweidler exponent b fulfill the connection put forward by MCT between the two quantities [see Eq. (10)], provided that one uses as a value of b the ones found in the vicinity of q_{max} and q_{min} . However, the exponent is not the same as the one we found for the critical behavior of the diffusion constant [1,7], which is in conflict with the prediction of the theory.

By investigating the q dependence of the relaxation time τ , we find that this quantity has a strong dependence on q . We also demonstrate that τ obeys the α -scale

universality [see Eq. (9)], as predicted by the theory.

We show that the time-temperature superposition principle can also be seen very well in the dynamic susceptibility $\chi''(q, \omega)$, in that we show that at low temperatures the width of the α peak does not depend on T to within the noise of the data.

By scaling frequency by ω_c , the location of the minimum in $\chi''(q, \omega)$, and scaling $\chi''(\omega)$ by $\chi''_c(q) = \chi''(q, \omega_c)$, we show that at low temperatures the curves for different temperatures fall onto a master curve. Thus we demonstrate that the scaling behavior predicted by MCT holds for our model. A fit with the functional form of MCT is not able to give a very good fit to this master curve when the exponent parameter λ is fixed to the value we determined from fits to the correlators in the time domain. This discrepancy is, however, not a flaw of the theory but traced back to an unpleasant property of the Fourier transformation of a time correlation function. If the parameter λ is allowed to float, we are able to generate a satisfactory fit in the frequency domain, but with the result that the λ determined in this way is not optimal anymore in the time domain. Thus we conclude that, if the theoretical curve does not give a good fit to the high frequency side of the minimum, but only gives a good fit on the low frequency side of the minimum, the determination of λ from fits to the master curve in the frequency domain is problematic in the sense that it might yield a wrong value of λ .

In contrast to the prediction of MCT χ''_c does not show a power-law dependence on temperature with an exponent 0.5 but rather with an exponent 0.8. However, the critical temperature is, as predicted by the theory, very close to the critical temperature for the diffusion constant or the relaxation time.

To summarize, we can say that MCT is able to describe the dynamics of our system at low temperatures in a surprisingly accurate way. There seem to be some differences between the behavior of our system and the predictions of the theory. However, all these discrepancies can be rationalized by taking into account that for values of q significantly different from q_{max} and q_{min} there are important corrections to the asymptotic results of the theory and that the theory is valid only very close to T_c . Understanding whether the observed discrepancies can really be understood within the framework of the theory or whether the theory has reached its limit of applicability is clearly of great interest, and we hope that this question can be answered in the future. One possible way to address this question is to solve numerically the mode coupling equations, in which the full q dependence is taken into account, and compare these solutions with the results of our simulation. This work is currently in progress [35].

ACKNOWLEDGMENTS

We thank Dr. J. Baschnagel, Dr. M. Fuchs, and Professor W. Götze for many useful discussions and a critical reading of the manuscript. Part of this work was supported by National Science Foundation Grant No. CHE89-18841. We made use of computer resources provided under NSF Grant No. CHE88-21737.

- [1] W. Kob and H. C. Andersen, Phys. Rev. E **51**, 4626 (1995).
- [2] U. Bengtzelius, W. Götze, and A. Sjölander, J. Phys. C **17**, 5915 (1984).
- [3] E. Leutheusser, Phys. Rev. A **29**, 2765 (1984).
- [4] W. Götze, in *Liquids, Freezing and the Glass Transition*, edited by J. P. Hansen, D. Levesque, and J. Zinn-Justin, Proceedings of the Les Houches Summer School of Theoretical Physics, Session LI, 1989 (North-Holland, Amsterdam, 1991), p. 287; W. Götze and L. Sjögren, Rep. Prog. Phys. **55**, 241 (1992).
- [5] R. Schilling, in *Disorder Effects on Relaxational Processes*, edited by R. Richert and A. Blumen (Springer, Berlin, 1994), p. 193.
- [6] *Relaxation Kinetics in Supercooled Liquids-Mode Coupling Theory and Its Experimental Tests*, edited by S. Yip, special issue of Trans. Theory Stat. Phys. **24** (6–8) 1995.
- [7] W. Kob and H. C. Andersen, Phys. Rev. Lett. **73**, 1376 (1994).
- [8] L. J. Lewis and G. Wahnström, Solid State Commun. **86**, 295 (1993); Phys. Rev. E, **50**, 3865 (1994).
- [9] G. Wahnström, Phys. Rev. A **44**, 3752 (1991).
- [10] G. F. Signorini, J.-L. Barrat, and M. L. Klein, J. Chem. Phys. **92**, 1294 (1990).
- [11] H. Löwen, J.-P. Hansen, and J.-N. Roux, Phys. Rev. A **44**, 1169 (1991).
- [12] J.-L. Barrat, J.-N. Roux, and J.-P. Hansen, Chem. Phys. **149**, 197 (1990); Y. Hiwatari, J. Matsui, K. Uehara, T. Muranaka, M. Miyagawa, M. Takasu, and T. Odagaki, Physica A **204**, 306 (1994).
- [13] W. Kob and H. C. Andersen, Nuovo Cimento D **16**, 1291 (1994).
- [14] J.-L. Barrat, J.-N. Roux, and J.-P. Hansen, Chem. Phys. **149**, 197 (1990).
- [15] G. Pastore, B. Bernu, J.-P. Hansen, and Y. Hiwatari, Phys. Rev. A **38**, 454 (1988); J. N. Roux, J. L. Barrat, and J.-P. Hansen, J. Phys. Condens. Matter **1**, 7171 (1989); S. P. Das, Phys. Rev. A **42**, 6116 (1990); L. J. Lewis, Phys. Rev. B **44**, 4245 (1991); J. Matsui, T. Odagaki, and Y. Hiwatari, Phys. Rev. Lett. **73**, 2452 (1994).
- [16] G. P. Johari and M. Goldstein, J. Chem. Phys. **53**, 2372 (1970).
- [17] M. Fuchs, J. Non-Cryst. Solids **172–174**, 241 (1994).
- [18] M. Fuchs and A. Latz, Physica A **201**, 1 (1993).
- [19] J.-P. Hansen and I. R. McDonald, *Theory of Simple Liquids* (Academic, London, 1986).
- [20] Note that our definition of the van Hove correlation function is slightly different from the one usually found in the literature (e.g., Ref. [19]). See Ref. [1] for details.
- [21] These theoretical curves were computed with a program written by M. Fuchs and provided to us by Fuchs *et al.* An alternative method would be to use the tables of Ref. [30].
- [22] U. Bengtzelius, Phys. Rev. A **34**, 5059 (1986).
- [23] F. Smolej and H. H. Hahn, Physica A **201**, 164 (1993).
- [24] W. van Meegen and S. M. Underwood, Phys. Rev. E **49**, 4206 (1994).
- [25] F. Mezei, W. Knaak, and B. Farago, Phys. Scr. **T19**, 363 (1987).
- [26] C. H. Reinsch, Num. Math. **10**, 177 (1967); C. H. Reinsch, *ibid.* **16**, 451 (1971).
- [27] *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (Dover, New York, 1964).
- [28] W. Götze and L. Sjögren, J. Phys. C **21**, 3407 (1988).
- [29] W. Steffen, A. Patkowski, H. Gläser, G. Meier, and E. W. Fischer, Phys. Rev. E **49**, 2992 (1994).
- [30] W. Götze, J. Phys. Condens. Matter **2**, 8485 (1990).
- [31] E. Rössler, A. P. Sokolov, A. Kisliuk, and D. Quitmann, Phys. Rev. B **49**, 14967 (1994).
- [32] G. Buchalla, U. Dersch, W. Götze, and L. Sjögren, J. Phys. C **21**, 4239 (1988).
- [33] J. Baschnagel, Phys. Rev. B **49**, 135 (1994).
- [34] W. Götze and L. Sjögren (private communications).
- [35] M. H. Nauroth, W. Kob, and K. Binder (unpublished).