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Test of molecular mode coupling theory for general rigid molecules

C. Theis,[1] F. Sciortino,[2] A. Latz,[3] R. Schilling,[3] and P. Tartaglia[2]
[1]*Fakultät für Physik, Albert-Ludwigs-Universität, Hermann-Herder-Straße 3, D-79104 Freiburg, Germany*
[2]*Dipartimento di Fisica and Istituto Nazionale per la Fisica della Materia, Università di Roma ''La Sapienza,''*
Piazzale Aldo Moro 2, I-00185 Roma, Italy
[3]*Institut für Physik, Johannes Gutenberg Universität, Staudinger Weg 7, D-55099 Mainz, Germany*

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We report recent progress on the test of mode coupling theory for molecular liquids (MMCT) for molecules of arbitrary shape. The MMCT equations in the long time limit are solved for supercooled water including all molecular degrees of freedom. In contrast to our earlier treatment of water as a linear molecule, we find that the glass-transition temperature T_c is overestimated by the theory as was found in the case of simple liquids. The nonergodicity parameters are calculated from the ''full'' set of MMCT equations truncated at $l_{co}=2$. These results are compared (i) with the nonergodicity parameters from MMCT with $l_{co}=2$ in the ''dipole'' approximation $n=n'=0$ and the diagonalization approximation $n=n'=0, l=l'$ and (ii) with the corresponding results from a MD simulation. This work supports the possibility that a reduction to the most prominent correlators may constitute a valid approximation for solving the MMCT equations for rigid molecules.

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I. INTRODUCTION

The mode coupling theory (MCT) for supercooled simple liquids proposed by Bengtzelius, Götze, and Sjölander [1] interprets the glass transition as a dynamical transition. This picture has been supported by many experiments on several glass formers (see, e.g., [2] and references therein) and more recently by detailed analysis of computer simulations for Lennard-Jones systems (see, e.g., [3] and references therein). The signature of the dynamical transition, i.e., the asymptotical power laws, has been discovered also via neutron scattering, light scattering, dielectric relaxation, and NMR in *molecular* glass formers (see [2] and references therein), stimulating the extension of MCT to molecular liquids.

Two approaches for such an extension have been proposed recently for rigid molecules. Chong and Hirata [4] introduced a theory based on a site-site description of the molecules. Their approach offers the advantage to be closely related to neutron scattering experiments. The structural information — which is a necessary input of the theory — can be readily obtained from theories of molecular liquids able to predict partial structure factors, like the reference interaction-site model (RISM) [5] approximation. The second approach is based on the expansion of the orientational density into a complete set of functions, in analogy to the Fourier expansion of the density related to the translational degrees of freedom. To distinguish the second approach from the site-site theory, it is called molecular mode coupling theory (MMCT). MMCT was derived for a single linear molecule in a simple liquid [6], for liquids of linear molecules [7] (for some application, see [8]), and for molecules of arbitrary shape [9,10]. MMCT allows us to calculate the glassy dynamics for all orientational degrees of freedom, and it is closely connected to dielectric and NMR experiments. As reorientational motion is also very important in light scattering experiments [11,12], it may also be helpful in their interpretation. The connection of MMCT to neutron scattering experiments has been discussed recently [13].

The fundamental MMCT quantities are the time-dependent correlation functions

$$S_{ln,l'n'}(q,m,t)=\langle\rho^*_{ln}(q,m,t)\rho_{l'n'}(q,m)\rangle \tag{1}$$

of the tensorial density modes

$$\rho_{ln}(q,m,t)=i^l(2l+1)^{1/2}\sum_{j=1}^{N} e^{i\vec{q}\cdot\vec{x}_j(t)}\mathcal{D}^{l*}_{mn}(\Omega_j(t)). \tag{2}$$

Here it is $\mathbf{q}=(0,0,q)$ and l runs over all positive integers including zero, m and n take integer values between $-l$ and l, and $\mathcal{D}$ denotes the Wigner functions [14]. The reader should note that the correlators (1) are diagonal in m for $\mathbf{q}=(0,0,q)$. The MMCT equations of motion for the Laplace transform $\mathbf{S}(q,m,z)=i\int_0^\infty \mathbf{S}(q,m,t)e^{izt}$ (Im $z>0$) have been presented in a preceding paper. Here we focus on the *unnormalized* molecular nonergodicity parameters

$$\mathbf{F}(q,m)=\lim_{t\to\infty}\mathbf{S}(q,m,t)=-\lim_{z\to 0} z\mathbf{S}(q,m,z) \tag{3}$$

which we calculated using the following set of equations [10]:

$$\mathbf{F}(q,m)=[\mathbf{S}^{-1}(q,m)+\mathbf{S}^{-1}(q,m)\mathbf{K}(q,m)\mathbf{S}^{-1}(q,m)]^{-1}, \tag{4}$$

$$K_{ln,l'n'}(q,m)=\sum_{\alpha\alpha'}\sum_{\mu\mu'} q^{\alpha\mu}_{ln}(q)\,q^{\alpha'\mu'*}_{l'n'}(q)\,(\mathbf{m}^{-1}(q))^{\alpha\mu,\alpha'\mu'}_{lmn,l'm'n'}, \tag{5}$$

1063-651X/2000/62(2)/1856(6)/$15.00

$$m^{\alpha\mu,\alpha'\mu'}_{lmn,l'm'n'}(q)=\frac{\rho_0}{(8\pi^2)^3}\int_0^\infty dq_1\int_{|q-q_1|}^{q+q_1}dq_2\sum_{m_1m_2}\sum_{\substack{l_1l_1'\\ l_2l_2'}}\sum_{\substack{n_1n_1'\\ n_2n_2'}}$$

$$\times v^{\alpha\mu}_{ln,l_1n_1,l_2n_2}(qq_1q_2;mm_1m_2)$$

$$\times v^{\alpha'\mu'*}_{l'n',l_1'n_1',l_2'n_2'}(qq_1q_2;m'm_1m_2)$$

$$\times F_{l_1n_1,l_1'n_1'}(q_1,m_1)F_{l_2n_2,l_2'n_2'}(q_2,m_2). \qquad (6)$$

The memory function matrix **m** in Eq. (6) represent the mode coupling approximation for the correlation function of fluctuating forces. The index α labels the translational ($\alpha=T$) and rotational ($\alpha=R$) currents, each of them consisting of three (spherical) vector components $\mu\in\{-1,0,1\}$. The vertex functions v are determined only by the matrix of the static molecular structure factors $\mathbf{S}(q,m)$ and the number density ρ_0. Their explicit form has been given in Ref. [10]. The coefficients $q^{\alpha\mu}_{ln}(q)$ appearing in Eq. (5) are

$$q^{\alpha\mu}_{ln}(q)=\begin{cases}0, & \alpha=T,\mu=\pm1\\ q, & \alpha=T,\mu=0\\ \frac{1}{\sqrt{2}}\sqrt{l(l+1)-n(n+\mu)}, & \alpha=R,\mu\pm1\\ n, & \alpha=R,\mu=0.\end{cases} \qquad (7)$$

Since $q^{T\pm1}_{ln}=0$ [due to the choice of $\mathbf{q}=(0,0,q)$], the transversal translational components ($\alpha=T,\mu=\pm1$) of the memory functions enter only indirectly [via the inversion of $\mathbf{m}(q)$] and thus shall be neglected in the following.

The given set of equations (4)–(6) includes all interactions between translational and rotational degrees of freedom in molecular liquids and thus this set is rather involved. Obviously its numerical solution poses a formidable task.

As a model system for our analysis, we have chosen SPC/E water [15]. The water molecule possesses a twofold rotational symmetry (C_{2v}) around the axis given by its dipole moment, which has been chosen as the z axis of the body fixed frame. The z axis and the x axis define the plane which is spanned by the molecule. As discussed in detail in Ref. [10], the C_{2v} symmetry can be used to simplify the equations of motion by the restriction that n and n' are even. Preceeding publications on long time MD simulations [16] for this strong glass former showed that the center-of-mass dynamics is in good agreement with the predictions of the asymptotic laws of MCT. Recent work [17] demonstrated that the signature of the dynamic transition can also be observed for the orientational degrees of freedom of the molecule.

A first approach to solving Eqs. (4)–(6) has been given in Ref. [10]. Apart from the necessary truncation of the range of l by a cutoff l_{co} for which we have chosen $l_{co}=2$, we introduced in [10] two more approximations. In the *dipole approximation*, water was treated as a linear molecule oriented in the direction of its dipole moment. This approximation corresponds to setting in Eqs. (4)–(6) the angular indices n, n' and the corresponding summation indices n_i, n_i' to zero.

TABLE I. Comparison of the critical temperatures T_c for the MD simulation and the theoretical calculations in the diagonal-dipole approximation, the dipole approximation, and for the ''full'' theory.

	MD	MMCT diag	MMCT dipole	MMCT ''full''
T_c	200 K	206 K	208 K	279 K

In Ref. [10], we have also studied the stronger *diagonal-dipole approximation*, which makes the further assumption that structure factors, nonergodicity parameters, and memory functions are diagonal with respect to the angular indices l and l'. It is the main purpose of the present paper to present results obtained by solving the *''full''* set of MMCT equations up to $l_{co}=2$ without any further approximation. The main difference from the approach in Ref. [10] is that we take the nonlinear character of the water molecule seriously. In particular, this means that the rotational motion of both protons around the dipolar axis is taken into account. Therefore, we can study the influence of this degree of freedom on the ideal glass-transition temperature T_c and on the nonergodicity parameters $F_{l0,l'0}(q,m)$ obtained in Ref. [10]. In addition, we also obtain the new parameters $F_{ln,l'n'}(q,m)$ with n and/or n' different from zero.

II. RESULTS

We have solved Eqs. (4)–(6) iteratively on a grid of 100 equispaced wave vectors, up to 110.7 nm^{-1}. One complete iteration requires 4 days of cpu time on a 533 MHz alpha workstation. The entire calculation to locate the critical temperature and the corresponding nonergodicity parameter, with a tolerance of 2.5×10^{-4} per point, requested more than 250 iterations. On a dedicated four-node parallel machine it required about 250 days. The number of iteration at the critical temperature was 54. We found that at $T=282$ K, the MMCT equations predict a liquid phase, while at $T=272$ K unambiguously a glassy state is predicted. Within the chosen tolerance, we locate T_c^{MCT} at $T_c^{MCT}=279$ K.

Table I summarizes the critical temperatures at which a transition from ergodic to nonergodic behavior is found. While T_c is almost equal for both approximations used in Ref. [10] and quite close to the result of the MD simulation, we find that the critical temperature is overestimated by almost 50% using the ''full'' theory. Thus we confirm our supposition in [10] that the agreement for T_c between simulation and both approximation schemes was fortuitous. Such a finding could also be expected from the static structure factors used as input. Besides the dominating diagonal structure factors $S_{00,00}(q,m=0)$ and $S_{10,10}(q,m)$, the most prominent peaks are displayed by $S_{10,2\pm2}(q,m)$ and $S_{2\pm2,2\pm2}(q,m)$ (see the figures in Ref. [18]), which were neglected in [10]. The overestimation of the critical temperature is common to the MCT for simple liquids [3] and seems to be a general deficiency of the mode coupling approximation. Thus we see that –although the overestimation of the critical temperature is not desired — it is necessary to include all static structure factors with large amplitude to get a concise description in the MMCT framework.

Figure 1 shows the comparison of the *unnormalized* criti-

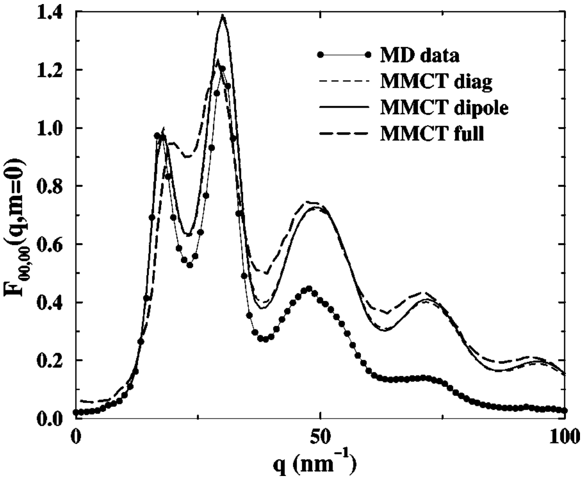

FIG. 1. Critical nonergodicity parameters $F_{00,00}(q,m=0)$ for the center of mass. The results from the MD simulation (symbols) are compared with the theoretical predictions of MMCT in the diagonal-dipole approximation (short dashed line), in the dipole approximation (solid line), and the results obtained using the ''full'' set of MMCT equations (long dashed line).

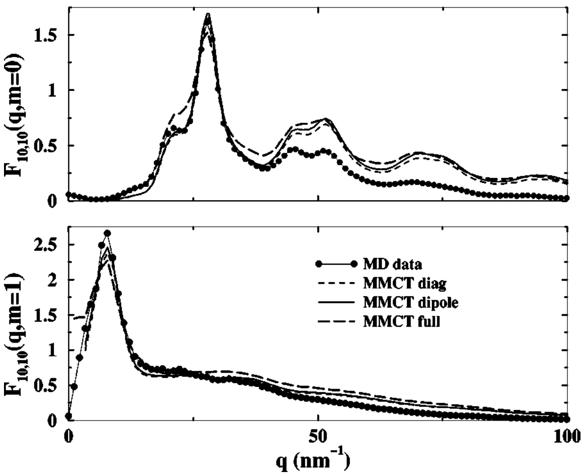

FIG. 2. Diagonal critical nonergodicity parameters $F_{10,10}(q,m)$ as calculated from the MD simulation (symbols) compared with the theoretical predictions of MMCT in the diagonal-dipole approximation (short dashed line), in the dipole approximation (solid line), and the results obtained using the ''full'' set of MMCT equations (long dashed line).

cal nonergodicity parameters $F_{00,00}(q,m=0)$ for the center-of-mass correlations. The oscillations of the MD result are captured well by all of our MMCT results. As for the critical temperature, we can also observe for the critical nonergodicity parameters that the dipole approximation and the diagonal-dipole approximation lead to nearly the same result. In the vicinity of the maximum of the structure factor, the agreement between theory and simulation is improved by removing the additional approximations, but for the prepeak as well as for large q both approximation schemes perform better than the ''full'' theory. In the region of the prepeak and especially for the minimum between prepeak and main peak, we observe that the oscillations are less pronounced and the peak positions are slightly shifted. Exactly the same behavior can be found by a comparison of the static structure factors at the different critical temperatures. Thus the worse performance of the ''full'' theory in comparison with the approximation schemes can be attributed at least partly to the overestimation of the critical temperature because of which the static input of the calculations does not reflect accurately the static structure at the ''true'' T_c of the simulation. Apart from that, it also has to be taken into account that in spite of the computational effort we have made, the fixed point of Eqs. (4)–(6) cannot be determined with very high precision. Therefore, the result for the critical nonergodicity parameters after 54 iterations may overestimate the exact one by a few percent. The latter reason may also be responsible for the worse performance of the ''full'' theory at the minima of $F_{00,00}(q,m=0)$ as Nauroth [19] found that the convergence of the iteration is extremely slow in these parts. Finally, we want to mention that the disagreement at large q between simulation and *all* theoretical calculations was also found for simple liquids [3] and was considered as a shortcoming of the mode coupling approximation.

As in the case of the center-of-mass correlators, the q dependence of the nonergodicity parameters $F_{10,10}(q,m)$ (see Fig. 2) is reproduced well by the theory. Here even in the vicinity of the maximum the approximations perform better than the ''full'' theory.

Figure 3 shows the comparison for the critical nonergodicity parameters $F_{20,20}(q,m)$. Apart from the region of large wave vectors, the ''full'' theory shows better agreement with the simulation than the two approximations. Thus one observes that the approximations, which neglect terms with $n\neq 0$ and consequently $l\geqslant 2$, have a stronger effect on the $l=2$ correlators than on those for $l=1$ or $l=0$. Further, one observes that the agreement between theory and simulation is less good for the $l=2$ correlators than for those with $l=0$ or $l=1$. The reader should note that the good agreement at large q for the $l=2$ correlators must be considered as

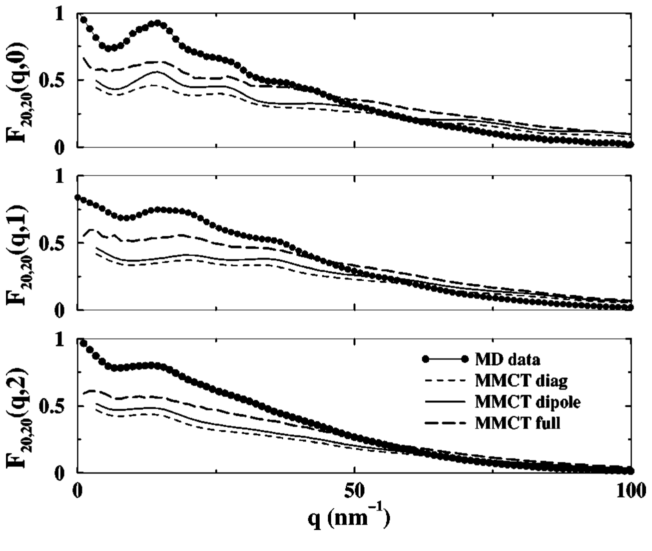

FIG. 3. Diagonal critical nonergodicity parameters $F_{20,20}(q,m)$ as calculated from the MD simulation (symbols) compared with the theoretical predictions of MMCT in the diagonal-dipole approximation (short dashed line), in the dipole approximation (solid line), and the results obtained using the ''full'' set of MMCT equations (long dashed line).

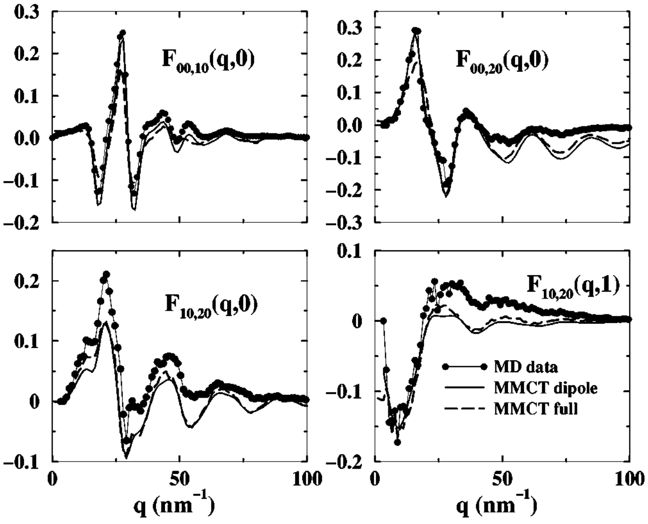

FIG. 4. Off-diagonal critical nonergodicity parameters $F_{l0,l'0}(q,m)$ with $l\neq l'$ as calculated from the MD simulation (symbols) compared with the theoretical predictions of MMCT in the dipole approximation (solid line) and the results obtained using the ‘‘full’’ set of MMCT equations (long dashed line).

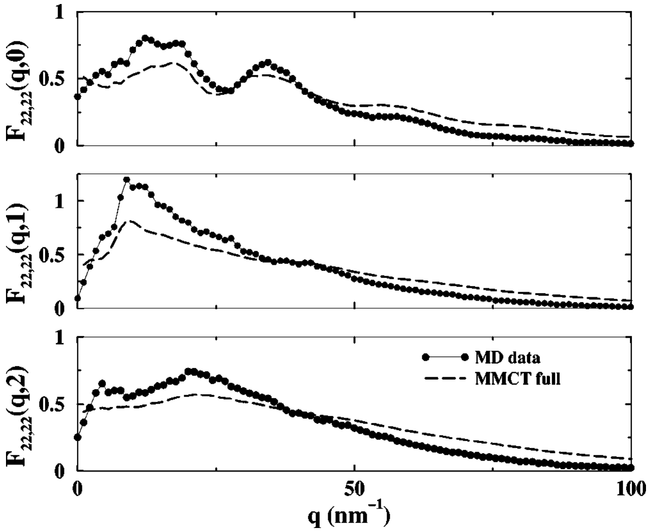

FIG. 5. Diagonal critical nonergodicity parameters $F_{22,22}(q,m)$ with $n\neq=0$ as calculated from the MD simulation (symbols) compared with the results obtained using the ‘‘full’’ set of MMCT-equations (long dashed line).

fortuitous because the overestimation of the nonergodicity parameters (seen for $l=0,1$) is compensated by a generally too small amplitude of the $l=2$ correlators. The reason for the worse agreement of the $l=2$ correlators is their higher sensibility to the cutoff at $l_{\text{co}}=2$, which can be understood on a mathematical level by a closer examination of the vertices which have been given in Ref. [10]. Let us pick out one special example to illustrate this point. The vertex factor $v^{\alpha\mu}_{00,20,20}(q,q_1,q_2;0,0,0)$ is responsible for the coupling of two correlators involving $l=0$ and $l=2$, respectively. It is of the form

$$v^{\alpha\mu}_{00,20,20}(q,q_1,q_2;0,0,0)$$

$$=\sum_{l,n} u^{\alpha\mu}_{00,ln,20}(q,q_1,q_2;0,0,0)c_{ln,20}(q_1,0)+(1\leftrightarrow 2), \quad (8)$$

where $u^{\alpha\mu}_{00,ln,20}(q,q_1,q_2;0,0,0)$ contains Clebsch-Gordan coefficients of the form $\mathcal{C}(l,2,0;m',-m',0)$, which enforces $l=2$. Therefore, the contribution to the memory function matrix caused by the vertex factor in Eq. (8) is exact, even for $l_{\text{co}}=2$. The vertex factor $v^{\alpha\mu}_{20,20,20}(q,q_1,q_2;0,0,0)$ instead, which describes the coupling of two correlators involving $l=2$ and $l=2$, respectively, has the form

$$v^{\alpha\mu}_{20,20,20}(q,q_1,q_2;0,0,0)$$

$$=\sum_{l,n} u^{\alpha\mu}_{20,ln,20}(q,q_1,q_2;0,0,0)c_{ln,20}(q_1,0)+(1\leftrightarrow 2). \quad (9)$$

The corresponding Clebsch-Gordan coefficient $\mathcal{C}(l,2,2;m',-m',0)$ allows $l\in\{0,1,2,3,4\}$. Thus for $l_{\text{co}}=2$ this vertex is not entirely taken into acount, although it is responsible for the coupling of $l=2$ correlations. From this example we can see that the $l=2$ quantities are more sensitive to the cutoff than those for $l=0$.

The *off-diagonal* critical nonergodicity parameters with $n=n'=0$ shown in Figure 4 are reproduced well by the theory. The difference between the dipole approximation and ‘‘full’’ theory is relatively small.

Figures 5 and 6 show the comparison between the new critical nonergodicity parameters with $n\neq 0$ and/or $n'\neq 0$ and the simulation results. Since those terms are neglected by the approximations, they can only be calculated using the ‘‘full’’ set of MMCT equations. The agreement between theory and simulation is satisfactory for almost all correlators.

In the discussion of the *unnormalized* critical nonergodicity parameters, we have seen that the worse performance of the ‘‘full’’ theory is partly due to the fact that the static input has to be taken at the ‘‘wrong’’ temperature. This influences the nonergodicity parameters in two ways. (i) $\mathbf{S}$ (q,m) is the initial value of the correlation function $\mathbf{S}(q,m,t)$ whose limit

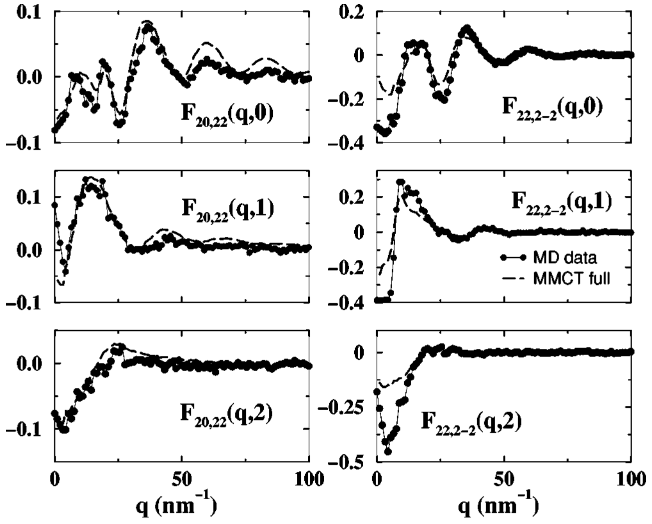

FIG. 6. Diagonal critical nonergodicity parameters $F_{2n,2n'}(q,m)$ with $n\neq 0$ and/or $n'\neq 0$ as calculated from the MD simulation (symbols) compared with the results obtained using the ‘‘full’’ set of MMCT equations (long dashed line).

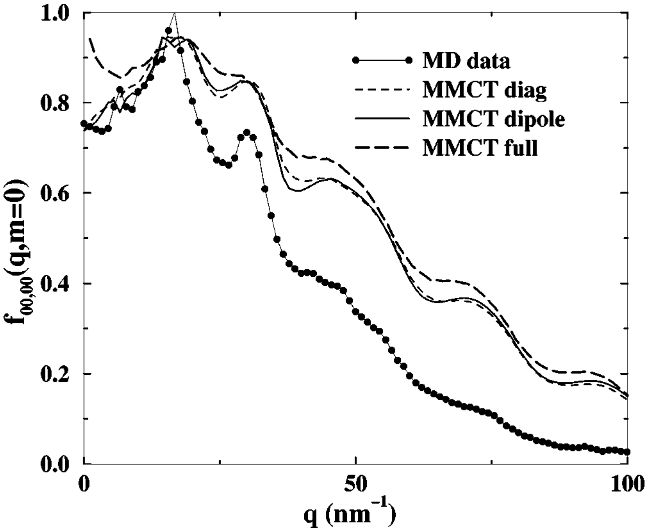

FIG. 7. Normalized critical nonergodicity parameters $f_{00,00}(q,0)$ for the center of mass. The results from the MD simulation (symbols) are compared with the theoretical predictions of MMCT in the diagonal-dipole approximation (short dashed line), in the dipole approximation (solid line), and the results obtained using the ''full'' set of MMCT equations (long dashed line).

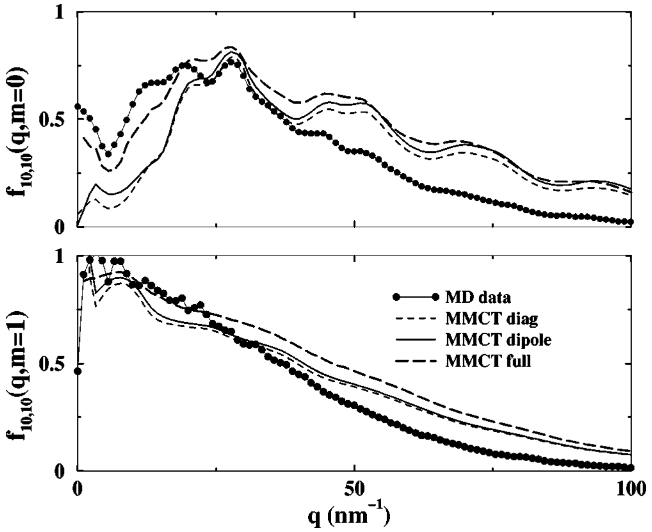

FIG. 8. Diagonal normalized critical nonergodicity parameters $f_{00,00}(q,m)$ as calculated from the MD simulation (symbols) compared with the theoretical predictions of MMCT in the diagonal-dipole approximation (short dashed line), in the dipole approximation (solid line), and the results obtained using the ''full'' set of MMCT equations (long dashed line).

for $t\rightarrow\infty$ is the *unnormalized* nonergodicity parameters $F_{ln,l'n'}(q,m)$. (ii) In the mode coupling approximation, the static structure factors determine the vertices which describe the coupling between the tensorial density modes in the system. This influence can partly be eliminated by calculating the *normalized* critical nonergodicity parameters

$$f_{ln,l'n'}(q,m)=\frac{F_{ln,l'n'}(q,m)}{\sqrt{S_{ln,ln}(q,m)S_{l'n',l'n'}(q,m)}}. \tag{10}$$

A selection of the normalized diagonal nonergodicity parameters is shown in Figs. (7–9). Due to the normalization, the variation with q is less pronounced. We observe that the difference in amplitude between the ''full'' theory and the approximations is reduced as the initial value $\mathbf{S}(q,m,t=0)$ of the time-dependent correlation functions is set to 1 at all wave vectors and all temperatures. Nonetheless, the dipole and diagonal-dipole approximation still provide the better description of the normalized critical nonergodicity parameter. In part this may be explained by the better convergence of the iteration for the approximation schemes, but the wrong critical temperature also has its influence as can be seen from the fact that the peak positions for the ''full'' MMCT results are still slightly shifted.

III. SUMMARY AND CONCLUSIONS

From our analysis we can state the following conclusions: (i) The mode coupling theory for molecular liquids overestimates the critical temperature T_c in the same fashion as its counterpart for simple liquids. (ii) The q dependence of the critical nonergodicity parameters is well reproduced in the vicinity of the main peaks. Systematic differences exist for large wave vectors. (iii) Deviations between theory and simulation are partly due to the overestimation of the critical temperature. Therefore, the structural input of the theory does not reflect properly the structure of the liquid at the true glass-transition temperature. These deviations still exist for the normalized nonergodicity parameters. (iv) Approximation schemes taking into account only part of the correlators like the dipole and the diagonal-dipole approximation can already give a reasonable description. (v) The essential correlation functions can be selected on the basis of the static structure factors. That is, for supercooled water, the most important static correlators are those with $n\neq 0$ and/or $n'\neq 0$, and for $n=n'=0$ those which are diagonal in l and l'.

We think that in combination with suitable approxima-

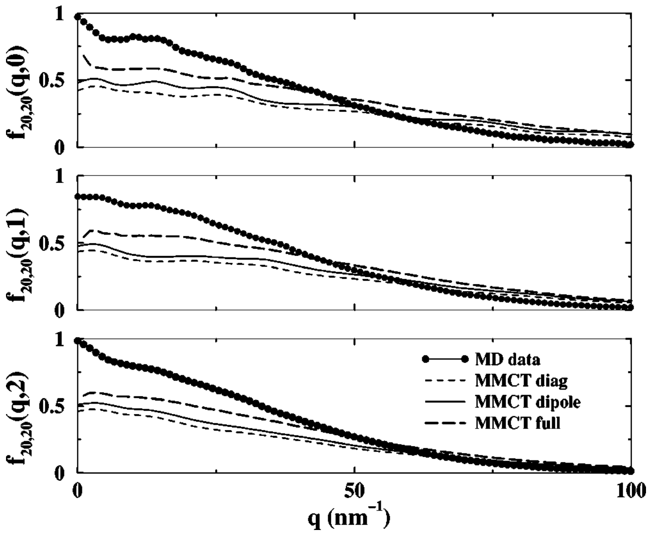

FIG. 9. Diagonal normalized critical nonergodicity parameters $f_{20,20}(q,m)$ as calculated from the MD simulation (symbols) compared with the theoretical predictions of MMCT in the diagonal-dipole approximation (short dashed line), in the dipole approximation (solid line), and the results obtained using the ''full'' set of MMCT equations (long dashed line).

tions for the static structure factors, MMCT will be a useful tool in the understanding of the glass transition in molecular liquids. The solution of the ‘‘full’’ set of MMCT equations is still a formidable task with present-day computers, but approximation schemes can be constructed on the basis of the importance of various static structure factors. A reduction to the most prominent correlators and q vectors offers the possibility to construct *microscopically based* schematic models for the description of molecular liquids.

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