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Reference-point-independent dynamics of molecular liquids and glasses in the tensorial formalism

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We apply the tensorial formalism to the dynamics of molecular liquids and glasses. This formalism separates the degrees of freedom into translational and orientational ones. Using the Mori-Zwanzig projection formalism, the equations of motion for the tensorial density correlators $S_{lmn,l'm'n'}(\vec{q},t)$ are derived. For this we show how to choose the slow variables such that the resulting Mori-Zwanzig equations are covariant under a change of the reference point of the body fixed frame. We also prove that the memory kernels obtained from mode-coupling theory (MCT) including all approximations preserve the covariance. This covariance makes, e.g., the glass transition point, the two universal scaling laws and particularly the corresponding exponents independent on the reference point and on the mass and moments of inertia, i.e., they only depend on the properties of the potential energy landscape. Finally, we show that the corresponding MCT questions for linear molecules can be obtained from those for arbitrary molecules and that they differ from earlier equations that are not covariant.

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I. INTRODUCTION

In this paper we will study the dynamics of a liquid of N rigid molecules. We will consider molecules of arbitrary shape, axially symmetric and linear molecules. To describe molecular liquids there exist two alternatives [1]: a *site-site description* or a *molecular representation*. The first one uses a partial density $\rho_\alpha(\vec{x},t)$, $\alpha=1,2,\ldots,M$ for the atomic sites of a molecule whereas the second one separates the $6N(5N)$ degrees of freedom into $3N$ translational and $3N(2N)$ rotational ones for arbitrary (linear) molecules, which leads to the molecular density $\rho(\vec{x},\Omega,t)$. $\Omega=(\phi,\theta,\chi)$, with ϕ, θ, and χ the Euler angles, characterizes the rotational dependence. In the following we will use the transformations

$$\rho_\alpha(\vec{q},t)=\int d^3x\,\rho_\alpha(\vec{x},t)e^{i\vec{q}\vec{x}} \tag{1}$$

and

$$\rho_{lmn}(\vec{q},t)=i^l(2l+1)^{1/2}\int d^3x\int d\Omega\,\rho(\vec{x},\Omega,t)e^{i\vec{q}\vec{x}}D^l_{mn}(\Omega)^*, \tag{2}$$

where $D^l_{mn}(\Omega)$ are Wigner's rotation matrices with $l=0,1,2,\ldots,-l\leqslant m\leqslant l$ and $-l\leqslant n\leqslant l$. This yields

$$\rho_\alpha(\vec{q},t)=\sum_j e^{i\vec{q}\vec{x}_j^{(\alpha)}(t)} \tag{3}$$

and the *tensorial* densities

$$\rho_{lmn}(\vec{q},t)=i^l(2l+1)^{1/2}\sum_j e^{i\vec{q}\vec{x}_j(t)}D^l_{mn}[\Omega_j(t)]^*. \tag{4}$$

$\vec{x}_j^{(\alpha)}(t)$ is the position of the atomic site α of molecule j at time t whereas $\vec{x}_j(t)$ and $\Omega_j(t)=[\phi_j(t),\theta_j(t),\chi_j(t)]$ are the chosen center and orientation of the jth molecule. The reader should note that this center need *not* coincide with the center-of-mass position. The corresponding density correlators

$$S_{\alpha\beta}(\vec{q},t)=\frac{1}{N}\langle\rho_\alpha(\vec{q},t)^*\rho_\beta(\vec{q},0)\rangle \tag{5}$$

and

$$S_{\kappa\kappa'}(\vec{q},t)=\frac{1}{N}\langle\rho_\kappa(\vec{q},t)^*\rho_{\kappa'}(\vec{q},0)\rangle \tag{6}$$

are of particular experimental and theoretical interest. Here we used the shorthand notation $\kappa=lmn$. For instance, neutron scattering yields direct information on the partial dynamical structure factors $S_{\alpha\beta}(\vec{q},t)$, but it does not allow us to determine the tensorial correlators $S_{\kappa\kappa'}(\vec{q},t)$, separately. In contrast, dielectric, light, and inelastic x-ray scattering experiments for *nonzero q* under some conditions allow us to determine the tensorial correlators for $l=l'=1$ and $l=l'=2$, but not the partial ones. From the knowledge of the *infinite* number of correlators $S_{\kappa\kappa'}(\vec{q},t)$ one can calculate the correlators $S_{\alpha\beta}(\vec{q},t)$ (which are of *finite* numbers) and also the neutron scattering cross section [2]. However, the opposite is not true. This makes the tensorial correlators superior to the partial ones. But on the other hand, the partial correlators have the advantage that their number is *finite* and that they are independent of any reference point. The separation of translational and rotational degrees of freedom in the tensorial formalism, however, requires the choice of a reference point within a molecule. Of course, choosing the center-of-mass position is the natural way. But any other choice is allowed as well. Consequently, equations of motion for the tensorial correlators must be *covariant*, i.e., they must keep their form under a shift of the reference point.

The Mori-Zwanzig projection formalism [1,3] allows us to derive equations of motion for $S_{\kappa\kappa'}(\vec{q},t)$. Although they are not closed, they are *exact*. In order that the physical quantities are independent on the reference point, it is of

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*Email address: rolf.schilling@uni-mainz.de primary importance to apply the projection formalism in such a way that the resulting Mori-Zwanzig equations are covariant. Their covariance is also crucial from a different point of view. For supercooled *simple* liquids it has been demonstrated that the Mori-Zwanzig equations can be closed by a so-called mode-coupling approximation and that the resulting set of mode-coupling equations yield an ideal glass transition [4]. For reviews the reader is referred to Refs. [5–8]. One of the essential features of this mode-coupling theory (MCT) is the independence of the long-time dynamics and of the glass transition singularity, of the particle's mass, i.e., there are no inertia effects except on a microscopic time scale [5]. This result is in full accordance with the ''potential energy landscape'' description of the glassy dynamics and the glass transition itself as has been stressed by Götze [5]. This type of approach, originally suggested by Goldstein [9] and further pioneered by Stillinger and Weber [10], considers properties such as minima and saddles of the potential energy, which of course do not depend on the mass. An interesting connection to the MCT glass transition temperature T_c has been discovered recently for simple liquids [11–13]. There it has been shown that the dynamical crossover at T_c relates to a geometrical transition at $T=T_c$, which is caused by a qualitative change in the topological properties of the potential energy landscape. In order to maintain this relationship for *molecular* liquids the corresponding Mori-Zwanzig equations and the resulting MCT equations must be covariant, which in turn will guarantee the absence of inertia effect on long time scales. The main motivation of the present contribution is to derive for molecular liquids such covariant equations in the tensorial formalism.

In order to apply the projection formalism one has to choose slow variables. For molecular liquids these are $\rho_\kappa(\vec{q},t)$ and the corresponding current densities

$$j_\kappa^{\alpha\mu}(\vec{q},t)=i^l(2l+1)^{1/2}\sum_j v_j^{\alpha\mu}(t)e^{i\vec{q}\vec{x}_j(t)}D^l_{mn}[\Omega_j(t)]^*, \tag{7}$$

where

$$v_j^{\alpha\mu}(t)=\begin{cases}\dot{x}_j^\mu(t), & \alpha=T,\\ \omega_j^\mu(t), & \alpha=R.\end{cases} \tag{8}$$

Energy density will not be taken into account. Note that the presence of translational and rotational degrees of freedom leads to translational ($\alpha=T$) and rotational ($\alpha=R$) current densities. The translational, $\dot{x}_j^\mu(t)$, and angular velocities $\omega_j^\mu(t)$ are given in spherical components, i.e., $\mu=0,+,-$. For the relation between Cartesian and spherical components of any vector $\vec{x}$ we adopt the convention of Ref. [14],

$$x^i=S^{i\mu}x^\mu,\quad x^\mu=\tilde{S}^{\mu i}x^i \tag{9}$$

with

$$(S^{i\mu})=\frac{1}{\sqrt{2}}\begin{pmatrix}1 & 0 & -1\\ i & 0 & i\\ 0 & \sqrt{2} & 0\end{pmatrix},\quad \tilde{\mathbf{S}}=\mathbf{S}^{-1}. \tag{10}$$

Here and in the following we will widely use the summation convention. ρ_κ and $j_\kappa^{\alpha\mu}$ are related by the continuity equation

$$\begin{aligned} iL\rho_\kappa(\vec{q},t)=\dot{\rho}_\kappa(\vec{q},t)&=iq^{\alpha\mu}_{\kappa\kappa'}(\vec{q})^*j^{\alpha\mu}_{\kappa'}(\vec{q},t)\\ &\equiv i[j^T_\kappa(\vec{q},t)+j^R_\kappa(\vec{q},t)]\end{aligned} \tag{11}$$

with L the Liouville operator,

$$q^{\alpha\mu}_{\kappa\kappa'}(\vec{q})=\begin{cases}q^\mu\delta_{\kappa\kappa'}, & \alpha=T,\\ (-1)^\mu L^{-\mu}_{l,mm'}\delta_{ll'}\delta_{nn'}, & \alpha=R,\end{cases} \tag{12}$$

and

$$L^\mu_{l,mm'}=\begin{cases}\mp\dfrac{1}{\sqrt{2}}[l(l+1)-m(m\pm1)]^{1/2}\delta_{m',m\pm1}, & \mu=\pm,\\ m\delta_{mm'}, & \mu=0.\end{cases} \tag{13}$$

There are several possibilities to choose slow variables among ρ_κ and $j_\kappa^{\alpha\mu}$. The simplest one is to choose ρ_κ and $\dot{\rho}_\kappa=i(j^T_\kappa+j^R_\kappa)$, i.e., besides ρ_κ we only take the sum of the *longitudinal* translational and the *''scalar''* rotational current densities. Since our main intention is to describe glassy dynamics and the glass transition for supercooled molecular liquids the long-time dynamics of the resulting equations of motion must not exhibit inertia effects, i.e., it must be independent of the mass M and the moments of inertia I_i, $i=1,2,3$. One can show that choosing ρ_κ and $\dot{\rho}_\kappa$ does not fulfill this condition. The simplest choice that is compatible with the inertia independence of the long-time dynamics is to use

(I) $\rho_\kappa, j^T_\kappa, j^R_\kappa,\quad \kappa=lm0$ (linear molecules),

(II) $\rho_\kappa, j^T_\kappa, j^{R\mu}_\kappa$ (arbitrary molecules).

In order to allow couplings to the *transversal*, translational current density, this can be extended to

(I′) $\rho_\kappa, j^{T\mu}_\kappa, j^R_\kappa, \kappa=lm0$ (linear molecules),

(II′) $\rho_\kappa, j^{T\mu}_\kappa, j^{R\mu}_\kappa$ (arbitrary molecules).

(I) has already been chosen for a linear molecule in an isotropic liquid [15] and for a liquid of linear molecules [16] and (II′) has been used for arbitrary molecules [17]. The account of transversal current density (I′) for linear molecules has recently been studied in Ref. [18]. It has already been stressed [19] that for linear molecules the choice (I) in Refs. [15,16] violates the covariance.

The covariance of the equations of motion will add a further condition to the choice of slow variables. The set of slow variables must be closed under a shift of the reference point. We will see that this condition is not fulfilled by (I) and (I′), but by (II) and (II′).

The outline of our paper is as follows. The following section contains the proof that the Mori-Zwanzig equations based on (II′) are covariant. In Sec. III we will show that the MCT equations obtained from the Mori-Zwanzig equations, by making a mode-coupling approximation for one of the memory kernels, preserves the covariance. The special case of axially symmetric molecules will be studied in Sec. IV and in Sec. V we will investigate linear molecules. Section VI contains a summary and some conclusions. Some technical proofs are given in the Appendixes.

II. COVARIANCE OF THE MORI-ZWANZIG EQUATIONS: ARBITRARY MOLECULES

Our main concern in this section is the change of the equations of motion in the tensorial representation under a shift of the reference point. Therefore, we will derive first in Sec. II A the transformation rules for the slow variables and their correlators. Then we will very briefly describe in Sec. II B the derivation of the Mori-Zwanzig equation and will finally prove their covariance. The reader should note that the present section will not involve any approximation.

A. Transformation under a shift of the reference point

Before applying the projection formalism we will just determine the transformation of ρ_κ and $j_\kappa^{\alpha\mu}$ under a shift,

$$\vec{x}_j(t) \rightarrow \tilde{\vec{x}}_j(t) = \vec{x}_j(t) + a\vec{u}_j(t), \quad |\vec{u}_j(t)| \equiv 1, \tag{14}$$

$$\Omega_j(t) \rightarrow \tilde{\Omega}_j(t) = \Omega_j(t), \tag{15}$$

of the reference point position $\vec{x}_j(t)$ by $a\vec{u}_j(t)$. From Eqs. (14) and (15) we get with Eq. (8),

$$\tilde{v}_j^{\alpha\mu}(t) = v_j^{\alpha\mu}(t) + \begin{cases} a\dot{u}_j^\mu(t), & \alpha = T, \\ 0, & \alpha = R. \end{cases} \tag{16}$$

The quantities $\tilde{\rho}_\kappa$ and $\tilde{j}_\kappa^{\alpha\mu}$ for the shifted reference point follow from ρ_κ and $j_\kappa^{\alpha\mu}$ by replacing in Eqs. (4) and (7) $\vec{x}_j, \Omega_j, v_j^{\alpha\mu}$ by $\tilde{\vec{x}}_j, \tilde{\Omega}_j, \tilde{v}_j^{\alpha\mu}$. $\tilde{\rho}_\kappa$ and $\tilde{j}_\kappa^{\alpha\mu}$ again are related by the continuity equation

$$\dot{\tilde{\rho}}_\kappa(\vec{q},t) = i\tilde{q}_{\kappa\kappa'}^{\alpha\mu}(\vec{q})^* \tilde{j}_{\kappa'}^{\alpha\mu}(\vec{q},t) \equiv i\tilde{L}\tilde{\rho}_\kappa(\vec{q},t); \tag{17}$$

$\tilde{q}_{\kappa\kappa'}^{\alpha\mu}(\vec{q})$ will be given below.

In the body fixed frame (BFF) the shift is given by

$$\vec{u}_j'(t) \equiv \vec{u}' \tag{18}$$

for *all* molecules. $\vec{u}_j(t)$ in the space fixed frame (SFF) and $\vec{u}'$ are related by

$$u_j^i(t) = R_{ii'}[\Omega_j(t)]u'^{i'}, \tag{19}$$

or in spherical components,

$$u_j^\mu(t) = D_{\mu\mu'}^1[\Omega_j(t)]^* u'^{\mu'}, \tag{20}$$

where the matrix $R[\Omega_j(t)]$ rotates the SFF into the BFF of the jth molecule. From Eq. (20) we get immediately

$$\dot{u}_j^\mu(t) = iq_{1\mu\mu',l'm'n'}^{R\mu''*} u'^{\mu'} \omega_j^{\mu''}(t) D_{m'n'}^{l'}[\Omega_j(t)]^*, \tag{21}$$

where we took advantage of the diagonality of $q_{\kappa\kappa'}^{R\mu}$, with respect to l and n [cf. Eq. (12)].

Replacement of $\vec{x}_j$ by $\tilde{\vec{x}}_j$ introduces in $\tilde{\rho}_\kappa$ and $\tilde{j}_\kappa^{\alpha\mu}$ an extra factor $\exp[ia\vec{q}\vec{u}_j(t)]$. Using the Rayleigh expansion [14], Eqs. (18) and (19) we find

$$\exp[ia\vec{q}\vec{u}_j(t)] = 4\pi \sum_{\kappa'} i^{l'} j_{l'}(aq) Y_{l'm'}(\Omega_q)^* \times Y_{l'n'}(\vec{u'}) D_{m'n'}^{l'}[\Omega_j(t)]^*, \tag{22}$$

where $j_l(x)$ are the spherical Bessel functions and Ω_q is the orientation of $\vec{q}$.

In the following we will also use the product rule [14],

$$[i^l(2l+1)^{1/2} D_{mn}^l(\Omega)^*][i^{l'}(2l'+1)^{1/2} D_{m'n'}^{l'}(\Omega)^*] = b_{\kappa\kappa'\kappa''}[i^{l''}(2l''+1)^{1/2} D_{m''n''}^{l''}(\Omega)^*], \tag{23}$$

where

$$b_{\kappa\kappa'\kappa''} = i^{l+l'-l''}[(2l+1)(2l'+1)(2l''+1)^{-1}]^{1/2} \times C(ll'l'';mm'm'')C(ll'l'';nn'n'') = b_{\kappa'\kappa\kappa''} \tag{24}$$

and $C(ll'l'';mm'm'')$ are the Clebsch-Gordon coefficients. With Eq. (16) and Eqs. (21)–(23) we find

$$\tilde{\rho}_\kappa(\vec{q},t) = T_{\kappa\kappa'}(\vec{q};a,\vec{u}')\rho_{\kappa'}(\vec{q},t), \tag{25}$$

$$\tilde{j}_\kappa^{\alpha\mu}(\vec{q},t) = Y_{\kappa\kappa'}^{\alpha\mu,\alpha'\mu'}(\vec{q};a,\vec{u}') j_{\kappa'}^{\alpha'\mu'}(\vec{q},t). \tag{26}$$

The transformation matrices **T** and **Y** are given by

$$T_{\kappa\kappa'}(\vec{q};a,\vec{u}') = 4\pi(2l''+1)^{-1/2} \times j_{l''}(aq) Y_{l''m''}(\Omega_q)^* Y_{l''n''}(\vec{u}') b_{\kappa''\kappa\kappa'} \tag{27}$$

and

$$Y_{\kappa\kappa'}^{\alpha\mu,\alpha'\mu'}(\vec{q};a,\vec{u}') = T_{\kappa\kappa''}(\vec{q};a,\vec{u}') U_{\kappa''\kappa'}^{\alpha\mu,\alpha'\mu'}(a,\vec{u}'), \tag{28a}$$

$$U^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(a,\vec{u}\,')=\begin{cases}\delta^{\mu\mu'}\delta_{\kappa\kappa'}, & \alpha=\alpha', \\ U^{\mu\mu'}_{\kappa\kappa'}(a,\vec{u}\,'), & \alpha=T, \quad \alpha'=R, \\ 0, & \alpha=R, \quad \alpha'=T,\end{cases} \tag{28b}$$

$$U^{\mu\mu'}_{\kappa\kappa'}(a,\vec{u}\,')=3^{-1/2}a q^{R\mu'*}_{1\mu\mu'',\kappa''}u'^{\mu''}b_{\kappa''\kappa\kappa'}. \tag{28c}$$

Note that $\mathbf{T}(\vec{q};a,\vec{u})$ and $\mathbf{U}(a,\vec{u}\,')$ commute with each other.

It is obvious that $\mathbf{T}(\vec{q};a,\vec{u})$ and $\mathbf{Y}(\vec{q};a,\vec{u})$ form a group. Furthermore, it is easy to prove that $\mathbf{T}$ is unitary,

$$\mathbf{T}(\vec{q};a,\vec{u}\,')^{\dagger}=\mathbf{T}(\vec{q};a,\vec{u})^{-1}, \tag{29}$$

and that

$$U^{\mu'\mu}_{\kappa'\kappa}(a,\vec{u}\,')^{*}=U^{\mu\mu'}_{\kappa\kappa'}(-a,\vec{u}\,'). \tag{30}$$

Equations (25) and (26) can be used in order to derive from Eqs. (11) and (17),

$$\tilde{q}^{\alpha\mu}_{\kappa\kappa'}(\vec{q})^{*}=T_{\kappa\kappa''}(\vec{q};a,\vec{u}\,')q^{\alpha'\mu'}_{\kappa''\kappa'''}(\vec{q})^{*}[\mathbf{Y}(\vec{q};a,\vec{u}\,')^{-1}]^{\alpha'\mu',\alpha\mu}_{\kappa''',\kappa'}, \tag{31}$$

which yields for $\alpha=T$,

$$\tilde{q}^{\mu}=q^{\mu}, \tag{32}$$

and for $\alpha=R$ the connection between $\tilde{L}^{\mu}_{l,mm'}$ and $L^{\mu}_{l,mm'}$ which involves q^{μ}.

The transformation law for the Hermitian matrices $\mathbf{S}(\vec{q},t)=[S_{\kappa\kappa'}(\vec{q},t)]$ and $\mathbf{J}(\vec{q},t)=[J^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)]$ with

$$J^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)=\frac{1}{N}\langle j^{\alpha\mu}_{\kappa}(\vec{q},t)^{*}j^{\alpha'\mu'}_{\kappa'}(\vec{q},0)\rangle \tag{33}$$

follows immediately from Eqs. (25) and (26),

$$\tilde{\mathbf{S}}(\vec{q},t)=\mathbf{T}(\vec{q};a,\vec{u}\,')^{*}\mathbf{S}(\vec{q},t)\mathbf{T}(\vec{q};a,\vec{u}\,')^{t}, \tag{34}$$

$$\tilde{\mathbf{J}}(\vec{q},t)=\mathbf{Y}(\vec{q};a,\vec{u}\,')^{*}\mathbf{J}(\vec{q},t)\mathbf{Y}(\vec{q};a,\vec{u}\,')^{t}, \tag{35}$$

where $\tilde{S}_{\kappa\kappa'}(\vec{q},t)$ and $\tilde{J}^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)$ is obtained from Eqs. (6) and (33) by replacing ρ_{κ}, $j^{\alpha\mu}_{\kappa}$ by $\tilde{\rho}_{\kappa},\tilde{j}^{\alpha\mu}_{\kappa}$.

B. Covariant Mori-Zwanzig equations

Equations (26) and (28)–(30) demonstrate that the shift of the reference point introduces a coupling between $\tilde{j}^{T\mu}_{\kappa}$ and $\tilde{j}^{R\mu}_{\kappa}$. For the longitudinal current density we get from Eqs. (26) and (28),

$$\tilde{j}^{T}_{\kappa}(\vec{q},t)=T_{\kappa\kappa''}(\vec{q};a,\vec{u}\,')[j^{T}_{\kappa''}(\vec{q},t)+q^{\mu*}U^{\mu\mu'}_{\kappa''\kappa'''}j^{R\mu'}_{\kappa'''}]. \tag{36}$$

Because the second term in the square bracket cannot be expressed by $j^{R}_{\kappa}(\vec{q},t)=q^{R\mu*}_{\kappa\kappa'}j^{R\mu}_{\kappa'}(\vec{q},t)$ but involves each component of $j^{R\mu}_{\kappa'}(\vec{q},t)$, the set (I) and (I') are not closed under a shift of the reference point. Therefore, we have to choose $j^{R\mu}_{\kappa},\mu=0,\pm$ as slow variables and not j^{R}_{κ}. Although we could still choose j^{T}_{κ}, since Eq. (36) shows that it is preserved, we also take $j^{T\mu}_{\kappa},\mu=0,\pm$ as additional variables. Therefore, we introduce the projectors

$$P_{\rho}=\sum_{\kappa_1\kappa_2}|\rho_{\kappa_1}(\vec{q})\rangle[\mathbf{S}(\vec{q})^{-1}]_{\kappa_1\kappa_2}\langle\rho_{\kappa_2}(\vec{q})^{*}|, \quad Q_{\rho}=1-P_{\rho}, \tag{37}$$

$$P_{j}=\sum_{\substack{\kappa_1\kappa_2\\ \alpha_1\mu_1,\alpha_2\mu_2}}|j^{\alpha_1\mu_1}_{\kappa_1}(\vec{q})\rangle[\mathbf{J}(\vec{q})^{-1}]^{\alpha_1\mu_1,\alpha_2\mu_2}_{\kappa_1\kappa_2}\langle j^{\alpha_2\mu_2}_{\kappa_2}(\vec{q})^{*}|, \tag{38}$$

$$P=P_{j}+P_{\rho}, \quad Q=1-P, \tag{39}$$

and similarly for $\tilde{P}_{\rho}=P_{\tilde{\rho}},\tilde{P}_{j}=P_{\tilde{j}}$, $\tilde{P}=\tilde{P}_{\rho}+\tilde{P}_{j}$, $\tilde{Q}_{\rho}=1-\tilde{P}_{\rho}$, and $\tilde{Q}=1-\tilde{P}$. The final result (cf. Refs. [15,16]) are the Mori-Zwanzig equations

$$\dot{\mathbf{S}}(\vec{q},t)+\int_0^t dt'\,\mathbf{K}(\vec{q},t-t')\mathbf{S}(\vec{q})^{-1}\mathbf{S}(\vec{q},t')=0, \tag{40a}$$

$$K_{\kappa\kappa'}(\vec{q},t)=q^{\alpha\mu}_{\kappa\kappa''}(\vec{q})k^{\alpha\mu,\alpha'\mu'}_{\kappa''\kappa'''}(\vec{q},t)q^{\alpha'\mu'}_{\kappa'\kappa'''}(\vec{q})^{*}, \tag{40b}$$

$$\dot{\mathbf{k}}(\vec{q},t)+\int_0^t dt'\,\mathbf{M}(\vec{q},t-t')\mathbf{J}(\vec{q})^{-1}\mathbf{k}(\vec{q},t')=0, \tag{40c}$$

and a similar set with $\mathbf{S},\mathbf{J},\mathbf{k},\mathbf{K},\mathbf{M},q^{\alpha\mu}_{\kappa\kappa'}$ replaced by $\tilde{\mathbf{S}},\tilde{\mathbf{J}},\tilde{\mathbf{k}},\tilde{\mathbf{K}},\tilde{\mathbf{M}},\tilde{q}^{\alpha\mu}_{\kappa\kappa'}$. The memory kernels are given by

$$k^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)=\frac{1}{N}\langle j^{\alpha\mu}_{\kappa}(\vec{q})^{*}|\exp(-iQ_{\rho}LQ_{\rho}t)|j^{\alpha'\mu'}_{\kappa'}(\vec{q})\rangle, \tag{41a}$$

$$M^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)=\frac{1}{N}\langle(Lj^{\alpha\mu}_{\kappa}(\vec{q}))^{*}|Q\exp(-iQLQt)Q|Lj^{\alpha'\mu'}_{\kappa'}(\vec{q})\rangle. \tag{41b}$$

$\tilde{\mathbf{k}}$ and $\tilde{\mathbf{M}}$ is obtained by replacing $j^{\alpha\mu}_{\kappa},Q_{\rho},Q$ and, L by $\tilde{j}^{\alpha\mu}_{\kappa},\tilde{Q}_{\rho},\tilde{Q}$, and $\tilde{L}$. Making use of Eqs. (25), (26), (34), and (35) it is easy to prove that the projectors are invariant,

$$\tilde{P}_{\rho}=P_{\rho}, \quad \tilde{P}_{j}=P_{j}, \quad \tilde{Q}_{\rho}=Q_{\rho}, \tag{42a}$$

and, therefore,

$$\tilde{P}=P, \quad \tilde{Q}=Q. \tag{42b}$$

The *invariance* of these projectors is of great importance. Taking Eqs. (42a) and (42b) into account we find with Eq. (26),

$$\tilde{\mathbf{k}}(\vec{q},t)=\mathbf{Y}(\vec{q};a,\vec{u}\,')^{*}\mathbf{k}(\vec{q},t)\mathbf{Y}(\vec{q};a,\vec{u}\,')^{t}, \tag{43a}$$

$$\tilde{\mathbf{M}}(\vec{q},t)=\mathbf{Y}(\vec{q};a,\vec{u}\,')^{*}\mathbf{M}(\vec{q},t)\mathbf{Y}(\vec{q};a,\vec{u}\,')^{t}. \qquad (43b)$$

Here we have used that for every phase space function $f(\underline{X})$ and $\tilde{f}(\tilde{\underline{X}})$,

$$\tilde{L}\tilde{f}(\tilde{\underline{X}})=L\tilde{f}[\tilde{\underline{X}}(\underline{X})], \qquad (44)$$

where the canonical transformation $\tilde{\underline{X}}(\underline{X})$ of a phase point $\underline{X}$ is generated by the coordinate transformation Eqs. (14) and (15). Now, making use of Eqs. (31), (34), (35), and (43a), (43b), it is easy to prove that the corresponding Eqs. (40) for the correlators $\tilde{\mathbf{S}}(\vec{q},t)$, $\tilde{\mathbf{K}}(\vec{q},t)$, etc., reduce to the Eqs. (40) for $\mathbf{S}(\vec{q},t)$, $\mathbf{K}(\vec{q},t)$, etc. This result means that the Mori-Zwanzig equations keep their form, or in other words, they are covariant under an arbitrary shift of the reference point.

III. COVARIANCE OF THE MCT EQUATIONS: ARBITRARY MOLECULES

The MCT equations are given by Eqs. (40) where the memory matrix $\mathbf{M}$ and $\tilde{\mathbf{M}}$ are approximated. In Sec. II we have shown that the Eqs. (40) are covariant under a shift of the reference point. Accordingly the MCT equations are covariant if the MCT approximation and eventually further approximations for the vertices preserve the transformation law, Eq. (43b). In order to investigate this covariance we have to derive the memory matrix $\mathbf{M}(\vec{q},t)$ and $\tilde{\mathbf{M}}(\vec{q},t)$ in MCT approximation. This derivation is given in Ref. [16] in great detail for linear and is easily extended to arbitrary molecules [17]. Here we will repeat only those steps that are relevant for proving the covariance. The main idea of MCT is to project in Eq. (41b) the fluctuating force $QLj_{\kappa}^{\alpha\mu}(\vec{q})$ onto a pair $\rho_{\kappa_1}(\vec{q}_1)\rho_{\kappa_2}(\vec{q}_2)$ of density modes. Then the slow part of $\mathbf{M}(\vec{q},t)$ is approximated as follows:

$$\begin{aligned}
&[M_{\kappa\kappa'}^{\alpha\mu,\alpha'\mu'}(\vec{q},t)]_{\text{slow}}\\
&\quad\approx[\mathbf{M}^{MCT}(\vec{q},t)]_{\kappa\kappa'}^{\alpha\mu,\alpha'\mu'}\\
&\quad=\frac{1}{2N}\sum_{\substack{\vec{q}_1\vec{q}_2\\ \kappa_1\kappa_2,\kappa_2'\kappa_2'}} W^{\alpha\mu,'\mu'}(\vec{q}\kappa\kappa'|\vec{q}_1\kappa_1\kappa_1';\vec{q}_2\kappa_2\kappa_2')S_{\kappa_1\kappa_1'}\\
&\qquad\times(\vec{q}_1,t)S_{\kappa_2\kappa_2'}(\vec{q}_2,t).
\end{aligned} \qquad (45a)$$

The vertices are given by

$$\begin{aligned}
&W^{\alpha\mu,\alpha'\mu'}(\vec{q}\kappa\kappa')|\vec{q}_1\kappa_1\kappa_1';\vec{q}_2\kappa_2\kappa_2')\\
&\quad=\frac{1}{N^2}[\mathbf{S}^{-1}(q_1)]_{\kappa_1''\kappa_1}[\mathbf{S}^{-1}(\vec{q}_2)]_{\kappa_2''\kappa_2}[\mathbf{S}^{-1}(\vec{q}_1)]_{\kappa_1'\kappa_1'''}\\
&\qquad\times[\mathbf{S}^{-1}(\vec{q}_2)]_{\kappa_2'\kappa_2'''}\langle[Lj_{\kappa}^{\alpha\mu}(\vec{q})]^{*}Q\rho_{\kappa_1''}(\vec{q}_1)\rho_{\kappa_2''}(\vec{q}_2)\rangle\\
&\qquad\times\langle\rho_{\kappa_1'''}(\vec{q}_1)^{*}\rho_{\kappa_2'''}(\vec{q}_2)^{*}QLj_{\kappa'}^{\alpha'\mu'}(\vec{q})\rangle.
\end{aligned} \qquad (45b)$$

We remind the reader that summation convention with respect to κ_i, κ_i', κ_i'', and κ_i''' is used. Again, $\tilde{\mathbf{M}}^{MCT}(\vec{q},t)$ is obtained by replacement of the corresponding quantities in Eqs. (45) by the quantities with tilde. Doing this and taking Eqs. (25), (26), (34), (42b), and (44) into account it is straightforward to prove that $\tilde{\mathbf{M}}^{MCT}$ and $\mathbf{M}^{MCT}$ obey the transformation law Eq. (43b), i.e., $\mathbf{M}^{MCT}$ is covariant.

For practical applications the vertices Eqs. (45) are further approximated. Using $Q=1-P$, Eqs. (37)–(39) and the hermiticity of L we get

$$\begin{aligned}
&\langle[Lj_{\kappa}^{\alpha\mu}(\vec{q})]^{*}Q\rho_{\kappa_1}(\vec{q}_1)\rho_{\kappa_2}(\vec{q}_2)\rangle\\
&\quad=\langle j_{\kappa}^{\alpha\mu}(\vec{q})^{*}L[\rho_{\kappa_1}(\vec{q}_1)\rho_{\kappa_2}(\vec{q}_2)]\rangle\\
&\qquad-[\mathbf{S}^{-1}(\vec{q})]_{\kappa_1'\kappa_2'}\langle j_{\kappa}^{\alpha\mu}(\vec{q})^{*}L\rho_{\kappa_1'}(\vec{q})\rangle\\
&\qquad\times\langle\rho_{\kappa_2'}(\vec{q})^{*}\rho_{\kappa_1}(\vec{q}_1)\rho_{\kappa_2}(\vec{q}_2)\rangle.
\end{aligned} \qquad (46)$$

The vertices become simpler if we apply the following approximation for the static three-point correlator (cf. Ref. [16] for linear molecules):

$$\begin{aligned}
&\langle\rho_{\kappa_1}(\vec{q}_1)^{*}\rho_{\kappa_2}(\vec{q}_2)\rho_{\kappa_3}(\vec{q}_3)\rangle\\
&\quad\approx N\delta_{\vec{q}_1+\vec{q}_2,\vec{q}_3}b_{\kappa_2'\kappa_3'\kappa_1'}S_{\kappa_1\kappa_1'}(\vec{q}_1)S_{\kappa_2'\kappa_2}(\vec{q}_2)S_{\kappa_3'\kappa_3}(\vec{q}_3).
\end{aligned} \qquad (47)$$

For such additional approximations we must discuss the maintenance of the covariant structure. This means that we have to prove that the right-hand side (rhs) of Eq. (47) transforms like its left-hand side (lhs). For the lhs we find with Eq. (25),

$$\begin{aligned}
&\langle\tilde{\rho}_{\kappa_1}(\vec{q}_1)^{*}\tilde{\rho}_{\kappa_2}(\vec{q}_2)\tilde{\rho}_{\kappa_3}(\vec{q}_3)^{*}\rangle\\
&\quad=T_{\kappa_1\kappa_1'}(\vec{q}_1,a,\vec{u}\,')^{*}T_{\kappa_2\kappa_2'}(\vec{q}_2,a,\vec{u}\,')T_{\kappa_3\kappa_3'}(\vec{q}_3,a,\vec{u}\,')\\
&\qquad\times\langle\rho_{\kappa_1'}(\vec{q}_1)^{*}\rho_{\kappa_2'}(\vec{q}_2)\rho_{\kappa_3'}(\vec{q}_3)\rangle.
\end{aligned} \qquad (48)$$

In Appendix A we prove the following identity for $\vec{q}_1+\vec{q}_2=\vec{q}$ (!):

$$\begin{aligned}
b_{\kappa_1\kappa_2\kappa_3}&=T_{\kappa_1'\kappa_1}(\vec{q}_1,a,\vec{u}\,')^{*}T_{\kappa_2'\kappa_2}(\vec{q}_2,a,\vec{u}\,')^{*}T_{\kappa_3'\kappa_3}\\
&\quad\times(\vec{q}_3,a,\vec{u}\,')b_{\kappa_1'\kappa_2'\kappa_3'}.
\end{aligned} \qquad (49)$$

Using Eqs. (34) and (49) it is easy to prove that indeed the rhs of Eq. (47) transforms like its lhs. Thus we can conclude that the MCT equations including all approximations transform covariantly under a shift of the reference point.

In order to determine how far these MCT equations simplify for *axially symmetric* molecules and reduce to the corresponding MCT equations for linear molecules presented in

Ref. [16], we have to determine the explicit form of the memory kernel $\mathbf{M}^{MCT}(\vec{q},t)$. The MCT equations and the memory kernel given in Ref. [17] are not identical to Eqs. (40) with memory kernel Eqs. (45) since in Ref. [17] we used the rotational current density

$$j'^{R\mu}_{\kappa}(\vec{q},t)=i^{l}(2l+1)^{1/2}\sum_{j}\ \omega'^{\mu}_{j}(t)e^{i\vec{q}\vec{x}_j(t)}D^{l*}_{mn}[\Omega_j(t)] \tag{50}$$

in the BFF. The reason has been that those static correlators that involve rotational current densities are much easier to calculate in the BFF where the rotational kinetic energy does not depend on $\{\Omega_j\}$. In that case and where $\vec{x}_j$ are the center-of-mass positions, it is

$$\langle \omega'^{\mu *}_{j}\omega'^{\mu'}_{j'}\rangle = k_B T I_i^{-1}\tilde{S}^{\mu i *}\tilde{S}^{\mu' i}\delta_{jj'}. \tag{51}$$

To transform the MCT equations in BFF to those in SFF we must determine the relationship between $j'^{R\mu}_{\kappa}$ and $j^{R\mu}_{\kappa}$. This is easily done by use of

$$\omega^{\mu}_{j}(t)=D^{1}_{\mu\mu'}[\Omega_j(t)]^{*}\omega'^{\mu'}_{j}(t) \tag{52}$$

and the product rule Eq. (23). The result is as follows:

$$j^{R\mu}_{\kappa}(\vec{q},t)=\mathcal{R}^{\mu\mu'}_{\kappa\kappa'}j'^{R\mu'}_{\kappa'}(\vec{q},t) \tag{53}$$

with the *unitary* matrix $\mathcal{R}$,

$$\mathcal{R}^{\mu\mu'}_{\kappa\kappa'}=i^{l-l'}\left(\frac{2l+1}{2l'+1}\right)^{1/2}C(1ll';\mu m m')C(1ll';\mu' n n'). \tag{54}$$

Since $\rho'_{\kappa}(\vec{q},t)=\rho_{\kappa}(\vec{q},t)$, the continuity equation in BFF reads

$$\dot{\rho}_{\kappa}(\vec{q},t)\equiv\dot{\rho}'_{\kappa}(\vec{q},t)=iq'^{\alpha\mu}_{\kappa\kappa'}(\vec{q})^{*}j'^{\alpha\mu}_{\kappa'}(\vec{q},t).$$

Then we get from Eq. (11) with Eq. (53) and the unitarity of $\mathcal{R}$,

$$q^{T\mu}_{\kappa\kappa'}(\vec{q})=q'^{T\mu}_{\kappa\kappa'}(\vec{q}),$$

$$q^{R\mu}_{\kappa\kappa'}(\vec{q})=q'^{R\mu'}_{\kappa\kappa''}(\vec{q})\mathcal{R}^{\mu\mu'}_{\kappa'\kappa''}. \tag{55}$$

It follows from Eq. (40c) that the long-time dynamics of the MCT equations in SFF is governed by the kernel

$$\mathbf{m}(\vec{q},t)=\mathbf{J}^{-1}(\vec{q})\mathbf{M}^{MCT}(\vec{q},t)\mathbf{J}^{-1}(\vec{q}). \tag{56}$$

Substituting Eq. (56) into Eq. (40c) yields

$$\dot{\mathbf{k}}(\vec{q},t)+\mathbf{J}(\vec{q})\int_0^t dt'\,\mathbf{m}(\vec{q},t-t')\mathbf{k}(\vec{q},t')=0. \tag{57}$$

As already mentioned above $\mathbf{J}(\vec{q})$ and $\mathbf{M}^{MCT}(\vec{q},t)$ can easily be calculated in the BFF. Taking the result from Ref. [17] that involves $q'^{\alpha\mu}_{\kappa\kappa'}(\vec{q})$ and making use of Eq. (55) one obtains the explicit form for $\mathbf{m}(\vec{q},t)$,

$$m^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)=\frac{1}{2N}\sum_{\vec{q}_1\vec{q}_2}\sum_{\substack{\kappa_1\kappa'_1,\\ \kappa_2\kappa'_2}}V^{\alpha\mu,\alpha'\mu'}$$
$$\times(\vec{q}\kappa\kappa'|\vec{q}_1\kappa_1\kappa'_1;\vec{q}_2\kappa_2\kappa'_2)S_{\kappa_1\kappa'_1}(\vec{q}_1,t)$$
$$\times S_{\kappa_2\kappa'_2}(\vec{q}_2,t), \tag{58a}$$

with the new vertices

$$V^{\alpha\mu,\alpha'\mu'}(\vec{q}\kappa\kappa'|\vec{q}_1\kappa_1\kappa'_1;\vec{q}_2\kappa_2\kappa'_2)$$
$$=\left(\frac{\rho_0}{8\pi^2}\right)^2\sum_{\kappa''\kappa''_1}[q^{\alpha\mu}_{\kappa''\kappa''_1}(\vec{q}_1)b_{\kappa''_1\kappa_2\kappa}c_{\kappa''\kappa_1}(\vec{q}_1)$$
$$+(1\leftrightarrow 2)]\sum_{\kappa'''\kappa'''_1}[q^{\alpha'\mu'}_{\kappa'''\kappa'''_1}(\vec{q}_1)b_{\kappa'''_1\kappa'_2\kappa'}c_{\kappa'''\kappa'_1}(\vec{q}_1)$$
$$+(1\leftrightarrow 2)]^{*}. \tag{58b}$$

They involve the direct correlation functions $c_{\kappa\kappa'}(\vec{q})$ that are related to $S_{\kappa\kappa'}(\vec{q})$ by the Ornstein-Zernike equation

$$\mathbf{S}(\vec{q})=\left(\mathbf{1}-\frac{\rho_0}{8\pi^2}\mathbf{c}(\vec{q})\right)^{-1}. \tag{59}$$

This equation is covariant due to the unitarity of **T**.

Note that (i) the vertices $V^{\alpha\mu,\alpha'\mu'}$ and, therefore, $m^{\alpha\mu,\alpha'\mu'}(\vec{q},t)$ do not depend anymore on M and $\{I_i\}$ explicitly, and (ii) the MCT equations in the SFF and BFF have the same form and differ only with respect to $q^{\alpha\mu}_{\kappa\kappa'}(\vec{q})$ and $q'^{\alpha\mu}_{\kappa\kappa'}(\vec{q})$, respectively.

This means that they are covariant under transformation from the SFF to BFF.

The static current density correlators become most *simple* if $\vec{x}_j$ are the center-of-mass positions. In that case we get

$$J^{T\mu,T\mu'}_{\kappa\kappa'}(\vec{q})=\frac{k_B T}{M}\delta^{\mu\mu'}\delta_{\kappa\kappa'}, \tag{60a}$$

$$J^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q})=0,\quad \alpha\neq\alpha', \tag{60b}$$

and for $\alpha=\alpha'=R$ with Eq. (51) in the BFF,

$$J'^{R\mu,R\mu'}_{\kappa\kappa'}(\vec{q})=k_B T I_i^{-1}\tilde{S}^{\mu i *}\tilde{S}^{\mu' i}\delta_{\kappa\kappa'}, \tag{60c}$$

which with the help of Eq. (53) leads to

$$J^{R\mu,R\mu'}_{\kappa\kappa'}(\vec{q})=\mathcal{R}^{\mu,\mu''}_{\kappa\kappa''}\mathcal{R}^{\mu',\mu'''}_{\kappa'\kappa''}k_B T I_i^{-1}\tilde{S}^{\mu'' i *}\tilde{S}^{\mu''' i}. \tag{60d}$$

IV. MCT EQUATIONS FOR AXIALLY SYMMETRIC MOLECULES

In this section we will choose $\vec{x}_j$ as the center of mass positions, because then the equations become most simple due to the axial symmetry. The equations for arbitrary $\vec{x}_j$ follow from Eqs. (40) by the use of the transformation laws (34), (35), and (43a), (43b). In that case axially symmetric molecules are characterized by two properties. First, two of the moment of inertia are equal,

$$I_1 = I_2 = I, \quad I_3 = I' \neq I, \tag{61}$$

and second, the potential energy does not depend on $\{\chi_j\}$,

$$V(\{\vec{x}_j\},\{\phi_j,\theta_j,\chi_j + \triangle\chi_j\}) \equiv V(\{\vec{x}_j\},\{\phi_j,\theta_j,\chi_j\}). \tag{62}$$

Equation (61) makes the rotational kinetic energy independent of $\{\chi_j\}$. This together with Eq. (62) implies that the component of angular momentum $L_j'^{i=3} \equiv L_j'^{\mu=0}$ (in the BFF) is a conserved quantity for each molecule. A second consequence of axial symmetry is the diagonality of the correlators with respect to n,

$$S_{\kappa\kappa'}(\vec{q},t) = S_{lmn,l'm'n}(\vec{q},t)\,\delta_{nn'},$$

$$J_{\kappa\kappa'}(\vec{q},t) = J_{lmn,l'm'n}(\vec{q},t)\,\delta_{nn'}. \tag{63}$$

For the static density correlators one can even prove that

$$S_{\kappa\kappa'}(\vec{q}) = \begin{cases} S_{\lambda\lambda'}(\vec{q}), & (n,n') = (0,0), \\ \delta_{\kappa\kappa'}, & (n,n') \neq (0,0), \end{cases} \tag{64}$$

with

$$S_{\lambda\lambda'}(\vec{q}) \equiv S_{lm0,l'm'0}(\vec{q}), \tag{65}$$

and $\lambda = (lm)$. Equation (64) together with the Ornstein-Zernicke equation (59) yields for the direct correlation function

$$c_{\kappa\kappa'}(\vec{q}) = 2\pi c_{\lambda\lambda'}(\vec{q})\,\delta_{n0}\delta_{nn'}, \tag{66}$$

$c_{\lambda\lambda'}(\vec{q}) = (1/2\pi)c_{lm0,l'm'0}(\vec{q})$ is the direct correlation function determined by $V(\{\vec{x}_j\},\{\ \phi_j,\theta_j\})$. The missing third angles χ_j introduce the factor 2π in Eq. (66). The fact that $c_{\kappa\kappa'}(\vec{q})$ vanishes for $(n,n') \neq (0,0)$ will strongly simplify the mode-coupling matrix $\mathbf{m}(\vec{q},t)$.

The static current density correlators [in case that $\vec{x}_j$ coincides with the center-of-mass position] also become simpler due to Eq. (61). $J^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q})$ is still given by Eqs. (60a) and (60b) for $(\alpha,\alpha') \neq (R,R)$, and for $\alpha = \alpha' = R$ one gets from Eqs. (60c) and (10),

$$J'^{R\mu,R\mu'}_{\kappa\kappa'}(\vec{q}) = \begin{cases} k_BT/I\,\delta^{\mu\mu'}\delta_{\kappa\kappa'}, & (\mu,\mu') \neq (0,0), \\ J'^{R0,R0}_{\kappa\kappa'}(\vec{q}), & (\mu,\mu') = (0,0), \end{cases} \tag{67}$$

where $J'^{R0,R0}_{\kappa\kappa'}(\vec{q})$ depends explicitly on $\{\omega_j'^{\mu=0}\}$ which are constant due to $L_j'^{\mu=0} = \text{const}$. In principle, this should exclude $\{\omega_j'^{\mu}\}$ from canonical averaging. If one is only interested in the slow dynamics of supercooled liquids the rotation around the symmetry axis is not particularly interesting. Therefore, let us take $\omega_j'^{\mu=0}(t) \equiv 0$ that leads to

$$j'^{R\mu=0}_{\kappa}(\vec{q},t) \equiv 0 \tag{68}$$

and, therefore,

$$J'^{R0,R0}_{\kappa\kappa'}(\vec{q}) \equiv 0. \tag{69}$$

Taking Eqs. (67) and (69) into account we get in the SFF,

$$J^{R\mu,R\mu'}_{\kappa\kappa'}(\vec{q}) = \frac{k_BT}{I}(\delta^{\mu\mu'}\delta_{\kappa\kappa'} - C^{\mu\mu'}_{\kappa\kappa'}) \tag{70}$$

with

$$C^{\mu\mu'}_{\kappa\kappa'} = \mathcal{R}^{\mu0*}_{\kappa\kappa''}\mathcal{R}^{\mu'0}_{\kappa'\kappa''}. \tag{71}$$

The unitarity of $\mathcal{R}$ yields immediately

$$\mathbf{C}^2 = \mathbf{C}, \tag{72}$$

i.e., $\mathbf{C}$ and therefore $\mathbf{1} - \mathbf{C}$, too, is a projector. Equations (53), (68), and (71) leads to

$$C^{\mu\mu'}_{\kappa\kappa'} j^{R\mu'}_{\kappa'}(\vec{q},t)^* = \mathcal{R}^{\mu0*}_{\kappa\kappa''} j'^{R0}_{\kappa''}(\vec{q})^* \equiv 0, \tag{73}$$

i.e., $\mathbf{C}$ projects out the rotational current density in the SFF. The choice Eq. (68) that has led to Eq. (70), introduces a problem since the inverse of $[J^{R\mu,R\mu'}_{\kappa\kappa'}(\vec{q})]$ and, therefore, the inverse of $[J^{\alpha\mu,\alpha'\mu}_{\kappa\kappa'}(\vec{q})]$ does not exist. This is because $\mathbf{1} - \mathbf{C}$ is a projector. However, $\mathbf{J}^{-1}(\vec{q})$ has been used in P_j for the derivation of the Mori-Zwanzig equations, only. In that case we have to choose $P_j = P_{j^T} + P_{j^R}$ as follows:

$$P_{j^T} = \sum_{\kappa,\mu} |j^{T\mu}_{\kappa}(\vec{q})\rangle \frac{M}{Nk_BT}\langle j^{T\mu}_{\kappa}(\vec{q})^*|, \tag{74a}$$

$$P_{j^R} = \sum_{\substack{\kappa\kappa' \\ \mu,\mu'}} |j^{R\mu}_{\kappa}(\vec{q})\rangle \frac{I}{Nk_BT}(\delta^{\mu\mu'}\delta_{\kappa\kappa'} - C^{\mu\mu'}_{\kappa\kappa'})\langle j^{R\mu'}_{\kappa'}(\vec{q})^*|. \tag{74b}$$

Note that the explicit form of P_{j^T} only holds if the reference point $\vec{x}_j$ coincides with the center-of-mass position. It is easy to prove that P_{j^R} is a projector. It simplifies due to Eq. (73) to

$$P_{j^R} = \sum_{\kappa,\mu} |j^{R}_{\kappa}(\vec{q})\rangle \frac{I}{Nk_BT}\langle j^{R\mu}_{\kappa}(\vec{q})^*|, \tag{74c}$$

such that

$$P_j = \sum_{\substack{\kappa\kappa' \\ \alpha\mu,\alpha'\mu'}} |j_\kappa^{\alpha\mu}(\vec{q})\rangle([\mathbf{J}^{(0)}(\vec{q})]^{-1})_{\kappa\kappa'}^{\alpha\mu,\alpha'\mu'}\langle j_{\kappa'}^{\alpha'\mu'}(\vec{q})^*|, \tag{75}$$

with

$$J_{\kappa\kappa'}^{(0)\alpha\mu,\alpha'\mu'}(\vec{q}) = N\frac{k_BT}{I_\alpha}\delta^{\alpha\alpha'}\delta_{\mu\mu'}\delta_{\kappa\kappa'} \tag{76}$$

and

$$I_\alpha = \begin{cases} M, & \alpha = T, \\ I, & \alpha = R. \end{cases} \tag{77}$$

Using P_ρ from Eq. (37) and P_j from Eq. (75) one gets the Eqs. (40), (56), and (57), where $\mathbf{J}(\vec{q})$ is replaced by $\mathbf{J}^{(0)}(\vec{q})$. The MCT approximation for $\mathbf{m}(\vec{q},t) = [\mathbf{J}^{(0)}(\vec{q})]^{-1} \mathbf{M}(\vec{q},t)[\mathbf{J}^{(0)}(\vec{q})]^{-1}$ is identical to the result Eqs. (58a), (58b), i.e., the explicit expression for $\mathbf{m}(\vec{q},\vec{t})$ does not depend on whether we ignore the conservation law $\omega_j'^{\mu=0}$ or not.

After this more general discussion we can now apply Eq. (66) to Eqs. (58a) and (58b) in order to get $m_{\kappa\kappa'}^{\alpha\mu,\alpha'\mu'}(\vec{q},t)$ for axially symmetric molecules. The result is as follows:

$$\begin{aligned} m_{\kappa\kappa'}^{\alpha\mu,\alpha'\mu'}(\vec{q},t) = \frac{1}{2N}\sum_{\vec{q}_1\vec{q}_2}\sum_{\substack{\lambda_1\lambda_1' \\ \lambda_2\lambda_2'}} & [(\cdots)S_{\lambda_1\lambda_1'}(\vec{q}_1,t)S_{\lambda_2 n,\lambda_2' n'} \\ & \times(\vec{q}_2,t) + (\cdots)S_{\lambda_1 n,\lambda_1' n'}(\vec{q}_1,t)S_{\lambda_2\lambda_2'}(\vec{q}_2,t) \\ & + (\cdots)S_{\lambda_1 0,\lambda_1' n'}(\vec{q}_1,t)S_{\lambda_2 n,\lambda_2' 0}(\vec{q}_1,t) \\ & + (\cdots)S_{\lambda_1 n,\lambda_1' 0}(\vec{q}_1,t)S_{\lambda_2 0,\lambda_2' n'}(\vec{q}_2,t)]. \end{aligned} \tag{78}$$

Here we used Eq. (64). $(\cdots)$ stands for the corresponding vertices. The first observation is that $m_{\kappa\kappa'}^{\alpha\mu,\alpha'\mu'}(\vec{q},t)$ is obviously not diagonal in n and n', because we are not allowed to assume that Eq. (63) also holds for the solutions $\mathbf{S}^{MCT}(\vec{q},t)$ of the MCT equations. The diagonal elements decompose into two types. The elements with $n = n' = 0$ only involve a bilinear product $S_{\lambda_1\lambda_1'}(\vec{q}_1,t)\cdot S_{\lambda_2\lambda_2'}(\vec{q}_2,t)$, i.e., correlators for which $n_1 = n_1' = 0$ and $n_2 = n_2' = 0$ whereas the elements with $n = n' \neq 0$ contain, e.g., $S_{\lambda_1\lambda_1'}(\vec{q}_1,t)S_{\lambda_2 n,\lambda_2' n}(\vec{q}_2,t)$. The nondiagonal elements also contain nondiagonal correlators (in n and n') that in principle should vanish because of Eq. (63). Indeed, we will show in Appendix B that the MCT equations (40) with $\mathbf{m}(\vec{q},t)$ from Eq. (78) imply that $S_{\lambda n,\lambda' n'}^{MCT}(\vec{q},t) = 0$ for $n \neq n'$ is a solution and we will prove under certain conditions that it is the only solution. Therefore, we assume that $S_{\lambda n,\lambda' n'}(\vec{q},t) \equiv 0$ for $n \neq n'$ is the only solution. Then it follows that

$$m_{\lambda n,\lambda' n'}^{\alpha\mu,\alpha'\mu'}(\vec{q},t) = m_{\lambda n,\lambda' n}^{\alpha\mu,\alpha'\mu'}(\vec{q},t)\,\delta_{nn'}. \tag{79}$$

Hence the MCT equations become diagonal in n and n'. This has the important consequence [cf. Eq. (78)] that the MCT equations for $S_{\lambda\lambda'}(\vec{q},t)$, which involve $m_{\lambda 0,\lambda' 0}^{\alpha\mu,\alpha'\mu'}(\vec{q},t)$ only, are closed, i.e., $S_{\lambda n\lambda' n}$ for $n \neq 0$ do not influence $S_{\lambda\lambda'}(\vec{q},t)$. But in contrast $S_{\lambda n,\lambda' n}(\vec{q},t)$ for $n \neq 0$ depend on $S_{\lambda\lambda'}(\vec{q},t)$.

The MCT equations for $S_{\lambda\lambda'}(\vec{q},t)$ for *axially symmetric* molecules then are given by Eqs. (40a), (40b), and (57) where all n, n', n'', etc., has to be set to zero and by the memory matrix

$$m_{\lambda 0,\lambda' 0}^{\alpha\mu,\alpha'\mu'}(\vec{q},t) \equiv (2\pi)^2 m_{\lambda\lambda'}^{\alpha\mu,\alpha'\mu'}(\vec{q},t) \tag{80}$$

with

$$\begin{aligned} m_{\lambda\lambda'}^{\alpha\mu,\alpha'\mu'}(\vec{q},t) = \frac{1}{2N}\sum_{\vec{q}_1\vec{q}_2}\sum_{\substack{\lambda_1\lambda_1' \\ \lambda_2\lambda_2'}} & V^{\alpha\mu,\alpha'\mu'}(\vec{q}\lambda\lambda'|\vec{q}_1\lambda_1\lambda_1'; \\ & \times \vec{q}_2\lambda_2\lambda_2')S_{\lambda_1\lambda_1'}(\vec{q}_1,t)S_{\lambda_2\lambda_2'}(\vec{q}_2,t), \end{aligned} \tag{81a}$$

$$\begin{aligned} & V_{\lambda\lambda'}^{\alpha\mu,\alpha'\mu'}(\vec{q}\lambda\lambda'|\vec{q}_1\lambda_1\lambda_1';\vec{q}_2\lambda_2\lambda_2') \\ & = \left(\frac{\rho_0}{4\pi}\right)^2 \sum_{\lambda''\lambda_1''}[q_{\lambda''\lambda_1''}^{\alpha\mu}(\overrightarrow{q_1})b_{\lambda_1''\lambda_2\lambda}c_{\lambda''\lambda_1}(\overrightarrow{q_1}) \\ & \quad + (1\leftrightarrow 2)]\sum_{\lambda'''\lambda_1'''}[q_{\lambda'''\lambda_1'''}^{\alpha'\mu'}(\overrightarrow{q_1})b_{\lambda_1'''\lambda_2'\lambda'}c_{\lambda'''\lambda_1'}(\overrightarrow{q_1}) \\ & \quad + (1\leftrightarrow 2)]^*, \end{aligned} \tag{81b}$$

and

$$q_{\lambda\lambda'}^{\alpha\mu}(\vec{q}) = q_{\lambda 0,\lambda' 0}^{\alpha\mu}(\vec{q}), \tag{82a}$$

$$b_{\lambda\lambda'\lambda''} = b_{\lambda 0,\lambda' 0,\lambda'' 0}. \tag{82b}$$

V. COVARIANT MCT EQUATIONS FOR LINEAR MOLECULES

In this section we will investigate two points. First of all we will derive Mori-Zwanzig equations and an MCT expression for the corresponding memory kernel for linear molecules that are covariant under a shift of the reference point. Second, we will show that these equations can be obtained from those for arbitrary molecules by taking into account the axial symmetry.

Linear molecules have $I' = 0$. This property makes the angular velocity $\vec{\omega}_j(t)$ always perpendicular to $\vec{e}_j(t)$, the unit vector along the symmetry axis of the jth molecule. In the

BFF where $\vec{e}_j(t)$ is presented by $\vec{e}_j\,' \equiv (0,0,1)$ we, therefore, get

$$\omega_j'^{\,\mu=0}(t) \equiv 0. \tag{83}$$

The tensorial density $\rho_\lambda(\vec{q},t)$ and current density $j_\lambda^{\alpha\mu}(\vec{q},t)$ (as used in Ref. [16]) for linear molecules are related to $\rho_\kappa(\vec{q},t)$ [Eq. (4)] and $j_\lambda^{\alpha\mu}(\vec{q},t)$ [Eq. (7)] by

$$\begin{aligned} \rho_\lambda(\vec{q},t) &= \rho_{\lambda 0}(\vec{q},t), \\ j_\lambda^{\alpha\mu}(\vec{q},t) &= j_{\lambda 0}^{\alpha\mu}(\vec{q},t), \end{aligned} \tag{84}$$

i.e., we have to put $n=n'=0$ (almost) everywhere for quantities of arbitrary molecules in order to get the corresponding quantities for linear molecules, provided $\vec{x}_j$ is chosen on the axis of the linear molecule. This rule does not apply, e.g., to the rotational current density in the BFF since Eqs. (53), (54), and (84) imply that $j_{\lambda,n}'^{\,R\mu}(\vec{q},t) \sim \delta_{n,-\mu}$. Due to Eq. (83) it must be

$$j_{\lambda n}'^{\,R0}(\vec{q},t) = 0 \tag{85}$$

for all n.

The rotational part of the static current density correlator can also be calculated without the detour via the BFF as done in Sec. III [cf. Eq. (60d)]. The calculation is straightforward but tedious. Therefore, we give the final result (again for $\vec{x}_j$, the center-of-mass position), only

$$J_{\lambda\lambda'}^{R\mu,R\mu'}(\vec{q}) = k_B T I^{-1}[\delta^{\mu\mu'}\delta_{\lambda\lambda'} - C_{\lambda\lambda'}^{\mu\mu'}], \tag{86}$$

where

$$C_{\lambda\lambda'}^{\mu\mu'} = \mathcal{R}_{\lambda\lambda''}^{\mu 0 *}\mathcal{R}_{\lambda'\lambda''}^{\mu' 0} \tag{87}$$

with

$$\mathcal{R}_{\lambda\lambda'}^{\mu\mu'} = \mathcal{R}_{\lambda 0,\lambda' 0}^{\mu\mu'} \tag{88}$$

and $\mathcal{R}_{\kappa\kappa'}^{\mu\mu'}$ from Eq. (54). Note that Eq. (54) leads to

$$\mathcal{R}_{\lambda 0,\lambda' n}^{\mu 0} = \mathcal{R}_{\lambda\lambda'}^{\mu 0}\delta_{n0}. \tag{89}$$

It is easy to see that the results for $J_{\lambda\lambda'}^{R\mu,R\mu'}(\vec{q})$, $C_{\lambda\lambda'}^{\mu\mu'}$ and $\mathcal{R}_{\lambda\lambda'}^{\mu\mu'}$ for linear molecules are identical to the corresponding quantities for axially symmetric particles but with $n=n'=0$. Furthermore, we can apply Eq. (89) to prove that

$$C_{\lambda\lambda''}^{\mu\mu''}C_{\lambda''\lambda'}^{\mu''\mu'} = C_{\lambda\lambda'}^{\mu\mu'} \tag{90}$$

and

$$C_{\lambda\lambda'}^{\mu\mu'} j_{\lambda'}^{R\mu}(\vec{q},t)^* \equiv 0, \tag{91}$$

where we also used Eq. (85). This demonstrates that $\mathbf{C} = (C_{\lambda\lambda'}^{\mu\mu'})$ and, therefore, $\mathbf{1}-\mathbf{C}$, too, is a projector. Now we can follow directly the argument in Sec. IV in order to show that the projector P_j is given by

$$P_j = \sum_{\substack{\lambda,\lambda' \\ \alpha\mu,\alpha'\mu'}} |j_\lambda^{\alpha\mu}(\vec{q})\rangle([\mathbf{J}^{(0)}(\vec{q})]^{-1})_{\lambda\lambda'}^{\alpha\mu,\alpha'\mu'}\langle j_{\lambda'}^{\alpha'\mu'}(\vec{q})^*| \tag{92}$$

with

$$J_{\lambda\lambda'}^{(0)\alpha\mu,\alpha'\mu'}(\vec{q}) = N\frac{k_B T}{I_\alpha}\delta^{\alpha\alpha'}\delta^{\mu\mu'}\delta_{\lambda\lambda'}. \tag{93}$$

We note that the result (92) can also be obtained from P_j [cf. Eq. (38)] for arbitrary molecules by choosing first $I_1 = I_2 = I$ and taking the limit $I_3 = I'$ to zero, and second using Eq. (91) and finally replacing κ by λ. Use of P_ρ from Eq. (37) with $\kappa \rightarrow \lambda$ and P_j from Eq. (92) we get the Mori-Zwanzig equations for linear molecules when the reference point $\vec{x}_j$ coincides with the center-of-mass position,

$$\dot{S}_{\lambda\lambda'}(\vec{q},t) + \int_0^t dt' K_{\lambda\lambda''}(\vec{q},t-t')[\mathbf{S}^{-1}(\vec{q})]_{\lambda''\lambda'''}S_{\lambda'''\lambda'}(\vec{q},t') = 0, \tag{94a}$$

$$K_{\lambda\lambda'}(\vec{q},t) = q_{\lambda\lambda''}^{\alpha\mu}(\vec{q})k_{\lambda''\lambda'''}^{\alpha\mu,\alpha'\mu'}(\vec{q},t)q_{\lambda'\lambda'''}^{\alpha'\mu'}(\vec{q})^*, \tag{94b}$$

$$\dot{k}_{\lambda\lambda'}^{\alpha\mu,\alpha'\mu'}(\vec{q},t) + \int_0^t dt' M_{\lambda\lambda''}^{\alpha\mu,\alpha''\mu''}(\vec{q},t-t') \times([\mathbf{J}^{(0)}(\vec{q})]^{-1})_{\lambda''\lambda'''}^{\alpha''\mu'',\alpha'''\mu'''}k_{\lambda'''\lambda'}^{\alpha'''\mu''',\alpha'\mu'}(\vec{q},t') = 0. \tag{94c}$$

These equations are identical to those for $S_{\lambda\lambda'}(\vec{q},t) \equiv S_{\lambda 0,\lambda' 0}(\vec{q},t)$ of axially symmetric particles. But they differ from the corresponding equations in Ref. [16], due to the presence of the μ and μ' dependence in Eqs. (94b) and (94c). MCT approximates $[\mathbf{M}(\vec{q},t)]_{\text{slow}}$ by $\mathbf{M}^{MCT}(\vec{q},t)$ where $[\mathbf{M}^{MCT}(\vec{q},t)]_{\lambda\lambda'}^{\alpha\mu,\alpha'\mu'}$ is given by Eqs. (45) but all κ replaced by λ. The corresponding vertices $W^{\alpha\mu,\alpha'\mu'}(\vec{q}\lambda\lambda'|\vec{q}_1\lambda_1\lambda_1';\vec{q}_2\lambda_2\lambda_2')$ involve $\langle[Lj_\lambda^{\alpha\mu}(\vec{q})]^* Q\rho_{\lambda_1}(\vec{q_1})\rho_{\lambda_2}(\vec{q_2})\rangle$ that can be calculated by using (i) $L_j'^{\,\mu=0}Y_\lambda(\Omega_j) = q_{\lambda\lambda'}'^{\,R0}Y_{\lambda'}(\Omega_j) \equiv 0$, since the zero component of the angular momentum vanishes for linear molecules in the BFF and (ii) the approximation for the static three-point correlator $\langle\rho_{\lambda_1}(\vec{q_1})^*\rho_{\lambda_2}(\vec{q_2})\rho_{\lambda_3}(\vec{q_3})\rangle$ as used in Ref. [16]. Note that this approximation also follows from the result Eq. (47) for arbitrary molecules by $\kappa \rightarrow \lambda$. Taking all this together one finds for

$$\mathbf{m}(\vec{q},t) = [\mathbf{J}^{(0)}(\vec{q})]^{-1}\mathbf{M}^{MCT}(\vec{q},t)[\mathbf{J}^{(0)}(\vec{q})]^{-1} \tag{95}$$

that

$$m^{\alpha\mu,\alpha'\mu'}_{\lambda\lambda'}(\vec{q},t)=\frac{1}{2N}\sum_{\vec{q}_1\vec{q}_2}\sum_{\substack{\lambda_1\lambda_1'\\ \lambda_2\lambda_2'}}V^{\alpha\mu,\alpha'\mu'}\times(\vec{q}\lambda\lambda'|\vec{q}_1\lambda_1\lambda_1';\vec{q}_2\lambda_2\lambda_2')S_{\lambda_1\lambda_1'}(\vec{q}_1,t)\times S_{\lambda_2\lambda_2'}(\vec{q}_2,t), \tag{96a}$$

with the new vertices

$$V^{\alpha\mu,\alpha'\mu'}(\vec{q}\lambda\lambda'|\vec{q}_1\lambda_1\lambda_1';\vec{q}_2\lambda_2\lambda_2')=\left(\frac{\rho_0}{4\pi}\right)^2\sum_{\lambda''\lambda_1''}[q^{\alpha\mu}_{\lambda''\lambda_1''}(\vec{q_1})b_{\lambda_1''\lambda_2\lambda}c_{\lambda''\lambda_1}(\vec{q_1})+(1\leftrightarrow 2)]\sum_{\lambda'''\lambda_1'''}[q^{\alpha'\mu'}_{\lambda'''\lambda_1'''}(\vec{q_1})b_{\lambda_1'''\lambda_2'\lambda'}c_{\lambda'''\lambda_1'}(\vec{q_1})+(1\leftrightarrow 2)]^* \tag{96b}$$

with $q^{\alpha\mu}_{\lambda\lambda'}(\vec{q})$ and $b_{\lambda\lambda'\lambda''}$ from Eqs. (82a) and (82b), respectively. Comparison with the result Eqs. (81a) and (81b) for axially symmetric particles and $n=n'=0$ reveals complete agreement.

What remains is to prove the covariance of the MCT equations for linear molecules under a shift [cf. Eqs. (14) and (15)] of the reference point. For linear molecules we restrict the shift $\vec{u}_j(t)$ for simplicity along the symmetry axis. One can also consider an *arbitrary* shift. But in that case one must use the tensorial quantities $\rho_\kappa(\vec{q},t)$ and $j^{\alpha\mu}_\kappa(\vec{q},t)$ with $n\geqslant 0$, even for linear molecules. Choosing $\vec{u}_j(t)\equiv\vec{e}_j(t)$ we get as a special case of Eqs. (25) and (26),

$$\tilde{\rho}_\lambda(\vec{q},t)=T_{\lambda\lambda'}(\vec{q};a)\rho_{\lambda'}(\vec{q},t), \tag{97}$$

$$\tilde{j}^{\alpha\mu}_\lambda(\vec{q},t)=Y^{\alpha\mu,\alpha'\mu'}_{\lambda\lambda'}(\vec{q};a)j^{\alpha'\mu'}_{\lambda'}(\vec{q},t). \tag{98}$$

Since $\vec{u}'=(0,0,1)$ it follows from Eq. (27),

$$T_{\lambda\lambda'}(\vec{q};a)\equiv T_{\lambda 0\lambda' 0}(\vec{q};a,\vec{u}')=\sqrt{4\pi}\sum_{l''m''}j_{l''}(aq)Y_{l''m''}(\Omega_q)^*b_{\lambda''\lambda\lambda'} \tag{99}$$

and from Eqs. (28),

$$Y^{\alpha\mu,\alpha'\mu'}_{\lambda\lambda'}(\vec{q};a)=T_{\lambda\lambda''}(\vec{q};a)U^{\alpha\mu,\alpha'\mu'}_{\lambda''\lambda'}(a), \tag{100a}$$

$$U^{\alpha\mu,\alpha'\mu'}_{\lambda\lambda'}(a)=\begin{cases}\delta^{\mu\mu'}\delta_{\lambda\lambda'}, & \alpha=\alpha',\\ U^{\mu\mu'}_{\lambda\lambda'}(a), & \alpha=T,\quad \alpha'=R,\\ 0, & \alpha=R,\quad \alpha'=T,\end{cases} \tag{100b}$$

$$U^{\mu\mu'}_{\lambda\lambda'}(a)=3^{-1/2}aq^{R\mu'*}_{1\mu,\lambda''}b_{\lambda''\lambda\lambda'}. \tag{100c}$$

It is obvious that $\mathbf{T}(a)$ and $\mathbf{Y}(a)$ have all the same properties like the corresponding quantities for arbitrary molecules.

For the derivation of the equations of motion for the shifted reference point we have to be cautious again with the projector $P_{\tilde{j}}$. As seen before, P_j for linear molecules can be obtained from P_j for arbitrary molecules by the procedure described below Eq. (93). We also use this approach to determine $\tilde{P}_j\equiv P_{\tilde{j}}$. This yields

$$\tilde{P}_j=\sum_{\substack{\lambda,\lambda'\\ \alpha\mu,\alpha'\mu'}}|\tilde{j}^{\alpha\mu}_\lambda(\vec{q})\rangle([\tilde{\mathbf{J}}^{(0)}(\vec{q})]^{-1})^{\alpha\mu,\alpha'\mu'}_{\lambda\lambda'}\langle\tilde{j}^{\alpha'\mu'}_{\lambda'}(\vec{q})^*|, \tag{101}$$

where

$$\tilde{\mathbf{J}}^{(0)}(\vec{q})=\mathbf{Y}(\vec{q};a)^*\mathbf{J}^{(0)}(\vec{q})\mathbf{Y}(\vec{q};a)^t. \tag{102}$$

$\tilde{P}_\rho=P_{\tilde{\rho}}$ is obviously given by

$$\tilde{\mathbf{P}}_\rho=\sum_{\lambda,\lambda'}|\tilde{\rho}_\lambda(\vec{q})\rangle[\tilde{\mathbf{S}}^{-1}(\vec{q})]_{\lambda\lambda'}\langle\tilde{\rho}_{\lambda'}(\vec{q})|^*. \tag{103}$$

Now it is easy to prove (like for arbitrary molecules) that the projectors are invariant, i.e.,

$$\tilde{P}_\rho=P_\rho,\quad \tilde{P}_j=P_j. \tag{104}$$

This fact together with the transformation rules Eqs. (97) and (98) can be used in close analogy to Sec. II B to prove that the Mori-Zwanzig equations Eqs. (94a), (94b), and (94c) are covariant under a shift of the reference point.

That $\mathbf{M}^{MCT}(\vec{q},t)$ and $\mathbf{m}(\vec{q},t)$ for linear molecules are also covariant can be proved by use of Eqs. (97) and (98), and

$$b_{\lambda_1\lambda_2\lambda_3}=T_{\lambda_1'\lambda_1}(\vec{q}_1;a)^*T_{\lambda_2'\lambda_2}(\vec{q}_2;a)^*T_{\lambda_3'\lambda_3}(\vec{q}_3;a)b_{\lambda_1'\lambda_2',\lambda_3'}, \tag{105}$$

which follows directly from Eq. (49). The proof of the covariance of $\mathbf{M}^{MCT}(\vec{q},t)$ and $\mathbf{m}(\vec{q},t)$ follows exactly the same steps as in Sec. III for arbitrary molecules. Accordingly the MCT equations (94a), (94b), and (94c) with the memory matrix given by Eqs. (95), (96a), and (96b) including *all* approximations are covariant in contrast to the MCT equations presented in Refs. [15,16].

VI. SUMMARY AND CONCLUSIONS

We have derived equations of motion for the correlators $S_{\kappa\kappa'}(\vec{q},t)$ and $S_{\lambda\lambda'}(\vec{q},t)$ in the *tensorial representation* for a liquid of arbitrary and linear molecules, respectively. These equations do not change their form under a shift of the reference point. The form invariance also holds for the MCT approximation of the memory matrix $M^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)$ and $M^{\alpha\mu,\alpha'\mu'}_{\lambda\lambda'}(\vec{q},t)$ including all approximations for the vertices. This so-called covariance means that if one has obtained a solution $\mathbf{S}^{MCT}(\vec{q},t)$ for one choice of the reference point, one obtains the solution $\tilde{\mathbf{S}}^{MCT}(\vec{q},t)$ for another reference point just from

$$\tilde{\mathbf{S}}^{MCT}(\vec{q},t)=\mathbf{T}(\vec{q};a,\vec{u}')^{*}\mathbf{S}^{MCT}(\vec{q},t)\mathbf{T}(\vec{q};a,\vec{u}')^{t}. \tag{106}$$

As shown in Sec. II, the original MCT equations for arbitrary molecules [17] are indeed covariant, but this is not true for the linear molecule in an isotropic liquid [15] and the liquid of linear molecules [16], as already stressed in Ref. [19]. Since the covariance guarantees that the physical properties do not depend on the reference frame it is important for molecular liquids of linear particles to use Eqs. (94a), (94b), and (94c) together with the memory matrix from Eqs. (95), (96a), and (96b). The modification for a single linear molecule in an isotropic liquid is straightforward.

The ideal glass transition is an example for which the physical behavior should be independent of the reference point. The MCT equations first derived by Götze and coworkers [4–6] for *simple liquids* yield a transition at a critical temperature T_c or a critical density n_c from an ergodic (liquid) to a nonergodic phase (glass). This transition is interpreted as an ideal glass transition. The extension to *molecular* liquids has been done in Ref. [20] within a site-site description and in Refs. [15–17] by use of the tensorial representation. In this respect we mention that the reported formulas in Ref. [20] did not yield the correct equations in the isotropic limit, i.e., where the particles become spherical. The corrected equations were presented in Ref. [19].

The nonergodicity parameters $F_{\kappa\kappa'}(\vec{q})=\lim_{t\to\infty}S_{\kappa\kappa'}(\vec{q},t)$ and $F_{\lambda\lambda'}(\vec{q})=\lim_{t\to\infty}S_{\lambda\lambda'}(\vec{q},t)$ change at T_c or n_c discontinuously from zero in the liquid to a nonzero value in the glass. Since it is for, e.g., arbitrary molecules,

$$\lim_{t\to\infty}S_{\kappa\kappa'}(\vec{q},t)=\begin{cases}0, & T>T_c \quad \text{or} \quad n<n_c,\\ F_{\kappa\kappa'}(\vec{q})>0, & T\leq T_c \quad \text{or} \quad n\geq n_c,\end{cases}$$

we get from Eq. (106) that $\tilde{F}_{\kappa\kappa'}(\vec{q})=\lim_{t\to\infty}\tilde{S}_{\kappa,\kappa'}(\vec{q})$ changes from zero to nonzero at the *same* critical point. This demonstrates that the covariance leads to a glass transition that does not depend on the reference point.

Furthermore, the covariance assures that the same is true for the critical exponent *a*, the von Schweidler exponent *b* that describe the time and frequency dependence in the β-relaxation regime [5], and for the exponent γ that determines the power law divergence of the α-relaxation time by approaching T_c (or n_c) from above (or below) [5] provided, these scaling laws are valid for the molecular version of MCT, as well. These few examples demonstrate the importance of the covariance of the MCT equations.

In addition, the covariance also guarantees that the long-time dynamics and the glass transition is independent on the inertia parameters *M* and $\{I_j\}$. This means that the properties of the long-time dynamics obtained from the covariant MCT depend on the potential energy landscape only. This nicely agrees with recent numerical investigations for simple liquids [11–13]. It would be interesting to perform analogous numerical studies for a molecular liquid.

Here a comment may be in order. To obtain the explicit time dependence of $S_{\kappa\kappa'}(\vec{q},t)$ from the MCT equations they have to be solved numerically. This numerical treatment requires a truncation of the infinite-dimensional matrices at $l=l_{co}<\infty$. This truncation destroys the covariance for *arbitrary* shifts $a\vec{u}'$ of the reference point. Because $\mathbf{T}(\vec{q};a,\vec{u}')\to\mathbf{1}$ and $\mathbf{Y}(\vec{q};a,\vec{u}')\to\mathbf{1}$ for $a\to 0$ the physical properties, however, will be practically the same for "a" small enough. Or in other words: for given l_{co} and accuracy δ, of, e.g., the critical temperature T_c, there exists $a(l_{co},\delta)$ such that $|T_c(a\neq 0)-T_c(a=0)|<\delta$ for all $a<a(l_{co},\delta)$. It is obvious that $a(l_{co},\delta)$ increases with l_{co} and that $\lim_{l_{co}\to\infty}a(l_{co},\delta)=\infty$.

Now, the reader may ask: If one would be able for linear molecules to solve the original noncovariant MCT equations [16,18] *without* a *l* cutoff and would compare with the solution of the covariant equations with $l_{co}<\infty$, how could one disentangle the effect of noncovariance due to the incorrect structure of the former equations from that due to truncation? If l_{co} is rather small, e.g., $l_{co}=2$, it is not possible to disentangle both effects. But, we do not expect any *qualitative* difference for the glassy properties such as the glass transition point or the long-time dynamics. These features will differ from each other only *quantitatively*. On the other hand, e.g., the nonergodicity parameters $f_{lmn,l'm'n'}(\vec{q})$ converge to zero for increasing l and l'. Therefore, if one systematically increases l_{co} in the covariant theory, the quantitative error due to truncation can be made rather small. Although this has not been checked for a molecular liquid up to now, we can imagine that $l_{co}=10$ already yields rather accurate results. Hence, variation of l_{co} for the covariant set of equations allows us to disentangle both effects, although the present computational power is not large enough to study $l_{co}=10$.

The crucial point for the covariance for *linear* molecules has been the use of each individual component of the *rotational* current density as a slow variable. Although not necessary, this has also been done for the translational current density in order to allow for a coupling to transverse (translational) current density, as it was done in Ref. [18]. This choice for linear molecules is not only essential for the covariance of their MCT equations but also that one can derive the MCT equations for linear from those for arbitrary molecules by choosing $I_1=I_2=I$ and taking the limit $I_3=I'$ to zero. We have also shown that $S_{\lambda 0,\lambda' 0}(\vec{q},t)$ for axially symmetric molecules decouple from all the other correlators $S_{\lambda n,\lambda' n}(\vec{q},t)$, $n>0$, and obey the MCT equations for linear molecules, as one expects.

To conclude, we can say that the tensorial formalism of molecular dynamics allows us to derive covariant equations of motion even including the MCT approximation and that the corresponding equations of motion for linear molecules are a special case of those for arbitrary ones.

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APPENDIX A

In this appendix we will prove the identity Eq. (49). Let

$$\rho_\kappa^{(0)}(\vec{q},\vec{x},\Omega)=i^l(2l+1)^{1/2}e^{i\vec{q}\vec{x}}D^l_{mn}(\Omega)^* \qquad \text{(A1)}$$

and

$$\tilde{\rho}_\kappa^{(0)}(\vec{q},\vec{x},\Omega)=\rho_\kappa^{(0)}(\vec{q},\tilde{\vec{x}},\Omega), \qquad \text{(A2)}$$

then we have from Sec. II,

$$\tilde{\rho}_\kappa^{(0)}(\vec{q},\vec{x},\Omega)=T_{\kappa,\kappa'}(\vec{q};a,\vec{u}\,'),\rho_{\kappa'}^{(0)}(\vec{q},\vec{x},\Omega). \qquad \text{(A3)}$$

Then we get

$$\frac{1}{8\pi^2}\int d\Omega D^{l_1}_{m_1n_1}(\Omega)^*D^{l_2}_{m_2n_2}(\Omega)^*D^{l_3}_{m_3n_3}(\Omega)$$
$$=(-i)^{l_1+l_2-l_3}[(2l_1+1)(2l_2+1)(2l_3+1)]^{-1/2}b_{\kappa_1\kappa_2\kappa_3} \qquad \text{(A4)}$$

for the following expression:

$$\frac{1}{V}\frac{1}{8\pi^2}\int d^3x\int d\Omega\tilde{\rho}_{\kappa_1}^{(0)}(\vec{q_1},\vec{x},\Omega)$$
$$\times\tilde{\rho}_{\kappa_2}^{(0)}(\vec{q_2},\vec{x},\Omega)\tilde{\rho}_{\kappa_3}^{(0)}(\vec{q_3},\vec{x},\Omega)^*$$
$$=\delta_{\vec{q}_1+\vec{q}_2,\vec{q}}\quad b_{\kappa_1\kappa_2\kappa_3}. \qquad \text{(A5)}$$

On the other hand we can use Eq. (A3) to calculate the lhs of Eq. (A5) which yields

$$\text{lhs of Eq. (A5)}=T_{\kappa_1\kappa_1'}(\vec{q_1};a,\vec{u}\,')T_{\kappa_2\kappa_2'}(\vec{q_2};a,\vec{u}\,')T^*_{\kappa_3\kappa'_3}(\vec{q};a,\vec{u}\,')\frac{1}{V}\frac{1}{8\pi^2}$$
$$\times\int d^3x\int d\Omega\rho_{\kappa_1'}^{(0)}(\vec{q}_1,\vec{x},\Omega)\rho_{\kappa_2'}^{(0)}(\vec{q}_2,\vec{x},\Omega)\rho_{\kappa_3'}^{(0)}(\vec{q}_3,\vec{x},\Omega)^*$$
$$=[\mathbf{T}^{-1}(\vec{q_1};a,\vec{u}\,')]^*_{\kappa_1'\kappa_1}[\mathbf{T}^{-1}(\vec{q_2};a,\vec{u}\,')]^*_{\kappa_2'\kappa_2}[\mathbf{T}^{-1}(\vec{q_3};a,\vec{u}\,')]_{\kappa_3'\kappa_3}b_{\kappa_1'\kappa_2'\kappa_3'}, \qquad \text{(A6)}$$

where we used the unitarity of **T**. Comparison of the rhs of Eqs. (A5) and (A6) yields the identity Eq. (49).

APPENDIX B

In this appendix we will prove that under certain conditions the nondiagonal elements $S_{\lambda n,\lambda' n'}(\vec{q},t)$ and $k^{\alpha\mu,\alpha'\mu'}_{\lambda n,\lambda' n'}(\vec{q},t)(n\neq n'!)$ of the MCT solutions vanish for all *t* for axially symmetric molecules provided the reference point $\vec{x}_j$ coincides with the center-of-mass position.

First of all it is rather easy to prove that

$$\frac{\partial^\nu}{\partial t^\nu}S_{\lambda n,\lambda' n'}(\vec{q},t)|_{t=0}=0,$$

$$\frac{\partial^\nu}{\partial t^\nu}k^{\alpha\mu,\alpha'\mu'}_{\lambda n,\lambda' n'}(\vec{q},t)|_{t=0}=0(n\neq n'), \qquad \text{(B1)}$$

for all $\nu\geqslant 0$. This follows by induction from Eqs. (40a), (40b), and (57) with $m^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)$ from Eq. (78) by taking into account the initial condition given by Eq. (B1) for $\nu=0$ and 1. If solutions of the MCT equations were analytic, Eq. (B1) would hold for *all t*. Concerning the smoothness we will only assume continuity in *t* in the following, and not analyticity.

It is easy to prove that $S_{\lambda n,\lambda' n'}(\vec{q},t)\equiv 0$ for $n\neq n'$ is a solution of the MCT equations. We will prove that it is the only solution. To proceed we decompose the correlation matrices in their diagonal and nondiagonal part with respect to *n* and n', which will be denoted by $(')$ and $('')$, respectively. Then we get from Eqs. (40a) and (57),

$$\mathbf{S}''(\vec{q},t)=-\int_0^t dt'\int_0^{t'}dt''[\mathbf{K}'(\vec{q},t'-t'')\mathbf{S}'(\vec{q})^{-1}\mathbf{S}''(\vec{q},t'')$$
$$+\mathbf{K}''(\vec{q},t'')\mathbf{S}'(\vec{q})^{-1}\mathbf{S}'(\vec{q},t'-t'')$$
$$+\mathbf{K}''(\vec{q},t'-t'')\mathbf{S}'(\vec{q})^{-1}\mathbf{S}''(\vec{q},t'')], \qquad \text{(B2)}$$

$$\mathbf{k}''(\vec{q},t)=-\mathbf{J}'(\vec{q})\int_0^t dt'\int_0^{t'}dt''[\mathbf{m}'(\vec{q},t'-t'')\mathbf{k}''(\vec{q},t'')$$
$$+\mathbf{m}''(\vec{q},t'')\mathbf{k}'(\vec{q},t'-t'')$$
$$+\mathbf{m}''(\vec{q},t'-t'')\mathbf{k}''(\vec{q},t'-t'')]. \qquad \text{(B3)}$$

Here we have integrated Eqs. (40a) and (57) over time and used the initial condition (B1) for $\nu=0$. Furthermore, the static correlators $\mathbf{S}(\vec{q})$ and $\mathbf{J}(\vec{q})$ we used were diagonal in *n* and n'. We want to prove that the quantities on the lhs of Eqs. (B2) and (B3) are zero. For this we will make the following assumptions for the diagonal elements:

$$\sup_{t,\vec{q},n} \sum_{\lambda,\lambda'} |S'_{\lambda n,\lambda' n}(\vec{q},t)| = S_o < \infty, \tag{B4a}$$

$$\sup_{t,\vec{q},n} \sum_{\lambda,\lambda'} |[\mathbf{K}(\vec{q},t)\mathbf{S}'(\vec{q})^{-1}]_{\lambda n,\lambda' n}| = K_o < \infty, \tag{B4b}$$

$$\sup_{t,\vec{q},n} \sup_{\substack{\alpha,\alpha' \\ \mu,\mu'}} \sum_{\lambda,\lambda'} |k'^{\,\alpha\mu,\alpha'\mu'}_{\lambda n,\lambda' n}(\vec{q},t)| = k_o < \infty, \tag{B4c}$$

$$\sup_{t,\vec{q},n} \sup_{\substack{\alpha,\alpha' \\ \mu,\mu'}} \sum_{\lambda,\lambda'} |[\mathbf{J}'(\vec{q})\mathbf{m}'(\vec{q},t)]^{\alpha\mu,\alpha'\mu'}_{\lambda n,\lambda' n}| = m_o < \infty, \tag{B4d}$$

and for the nondiagonal elements,

$$\sup_{t\leqslant T} \sup_{\vec{q}} \sum_{\kappa,\kappa'} |S''_{\kappa\kappa'}(\vec{q},t)| = \sigma(T) < \infty, \tag{B5a}$$

$$\sup_{t\leqslant T} \sup_{\vec{q}} \sum_{\kappa,\kappa'} |[\mathbf{K}''(\vec{q},t)][\mathbf{S}'(\vec{q})^{-1}]_{\kappa\kappa'}| = \kappa(T) < \infty, \tag{B5b}$$

$$\sup_{t\leqslant T} \sup_{\vec{q}} \sup_{\substack{\alpha,\alpha' \\ \mu,\mu'}} \sum_{\kappa,\kappa'} |k''^{\,\alpha\mu,\alpha',\mu'}_{\kappa,\kappa'}(\vec{q},t)| = \gamma(T) < \infty, \tag{B5c}$$

and for the vertices,

$$\sup_{\vec{q}} \sup_{\substack{\alpha,\alpha' \\ \mu,\mu'}} \sup_{\substack{\kappa_1\kappa_1' \\ \kappa_2\kappa_2'}} \frac{1}{2N} \sum_{\substack{\vec{q}_1\vec{q}_2 \\ \kappa,\kappa'}} \sum_{\kappa'',\alpha'',\mu''} |J'^{\,\alpha\mu,\alpha''\mu''}_{\kappa\kappa''}(\vec{q})$$
$$\times V^{\alpha''\mu'',\alpha'\mu'}(\vec{q}\kappa''\kappa'|\vec{q}_1\kappa_1\kappa_1';\vec{q}_2\kappa_2\kappa_2')| = V_o < \infty. \tag{B6}$$

Note that in Eqs. (B5) the supremum is taken over $t\leqslant T$, only.

Due to the continuity of the correlators and due to Eq. (B1) for $\nu=0$, it is

$$\sigma(T)\to 0, \quad \kappa(T)\to 0, \quad \gamma(T)\to 0, \quad T\to 0. \tag{B7}$$

Now, taking the modulos of the nondiagonal part $m''^{\,\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)$ from Eq. (78) one gets with Eqs. (B4) and (B6),

$$\sup_{t\leqslant T} \sup_{\vec{q}} \sup_{\substack{\alpha,\alpha' \\ \mu,\mu'}} \sum_{\kappa,\kappa'} |[\mathbf{J}'(\vec{q})\mathbf{m}''(\vec{q},t)]^{\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}|$$
$$\leqslant V_o[S_o+\sigma(T)]\sigma(T). \tag{B8}$$

The upper bound on the rhs of Eq. (B8) converges to zero due to Eq. (B7). Next, making use of Eqs. (B4), (B5), and (B8) we find from Eqs. (B2) and (B3),

$$\sup_{t\leqslant T} \sup_{\vec{q}} \sum_{\kappa,\kappa'} |S''_{\kappa\kappa'}(\vec{q},t)|$$
$$\leqslant [K_o\sigma(T)+S_o\kappa(T)+\sigma(T)\kappa(T)]\frac{1}{2}T^2 \tag{B9}$$

and

$$\sup_{t\leqslant T} \sup_{\vec{q}} \sup_{\substack{\alpha,\alpha' \\ \mu,\mu'}} \sum_{\kappa,\kappa'} |k''^{\,\alpha\mu,\alpha'\mu'}_{\kappa\kappa'}(\vec{q},t)| \leqslant 6[m_o\gamma(T)$$
$$+V_o[S_o+\sigma(T)]\sigma(T)[k_o+\gamma(t)]]\frac{1}{2}T^2, \tag{B10}$$

where the factor 6 in Eq. (B10) originates from $\Sigma_{\alpha'',\mu''}1 = 6$.

Now let

$$\sigma(T)+\kappa(T)+\gamma(T)\equiv\mu(T)>0, \tag{B11}$$

then we get from Eqs. (B9), (B10), and (B11),

$$\mu(T)\leqslant\{[K_o+S_o+6m_o+6V_oS_ok_o+1]\mu(T)$$
$$+[1+6V_o(k_o+S_o)]\mu^2(T)+\mu^3(T)\}\frac{1}{2}T^2, \tag{B12}$$

where we used the relation, e.g., that $\sigma(T)\leqslant\mu(T)$, $\sigma(T)\kappa(T)\leqslant\mu^2(T)$, etc.

Since the coefficients of $\mu^\nu(T)$ on the rhs of (B12) and $\mu(T)$ are positive, and $\mu(T)\to 0$ for $T\to 0$ it is obvious that there is a finite $T_o>0$ such that for $T\leqslant T_o$ the rhs of (B12) becomes smaller than its lhs due to the factor T^2, which is a contradiction. Therefore, it must be $\mu(T)=0$, for all $T \leqslant T_o$. Since $\sigma(T)\geqslant 0$, $\kappa(T)\geqslant 0$, and $\gamma(T)\geqslant 0$ Eq. (B11) implies that

$$\sigma(t)=0, \quad \kappa(T)=0, \quad \gamma(T)=0, \quad \forall T\leqslant T_o,$$

and this in turn proves that the nondiagonal part (in n and n'!) of the correlators vanishes for $T\leqslant T_o$.

To prove that this result holds for all T one restricts Eqs. (40a), (40b), and (57) to $t\geqslant T_o$ with the new initial conditions,

$$S_{\lambda n,\lambda' n'}(\vec{q},T_o)=0, \quad k^{\alpha\mu,\alpha'\mu'}_{\lambda n,\lambda' n'}(\vec{q},T_o)=0, \quad n\neq n'. \tag{B13}$$

Introducing the "shifted" functions

$$\tilde{S}_{\lambda n,\lambda' n'}(\vec{q},t) := S_{\lambda n,\lambda' n'}(\vec{q},T_o+t), \quad \text{etc.}, \tag{B14}$$

the Eqs. (B2) and (B3) keep their form for $\tilde{\mathbf{S}}''(\vec{q},t)$ and $\tilde{\mathbf{k}}''(\vec{q},t)$ with the only difference that the $(t'-t'')$-dependent kernels are the old ones. Then, following again the procedure described above, one finds that there exists $\tilde{T}_o>0$ such that the nondiagonal part of the correlators vanish for $T\leqslant T_o T\leqslant T_o+\tilde{T}_o$. Now ‘‘shifting’’ again by $T_o+\tilde{T}_o$ the equations for $\tilde{\tilde{\mathbf{S}}}''(\vec{q},t)$ and $\tilde{\tilde{\mathbf{k}}}''(\vec{q},t)$ are the same as for $\tilde{\mathbf{S}}''(\vec{q},t)$ and $\tilde{\mathbf{k}}''(\vec{q},t)$. Therefore, it follows that the nondiagonal part of the correlators vanishes for $T\leqslant T_o+2\tilde{T}_o$. Iterating this procedure infinitely many times completes the proof.

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