Molecular dynamics of liquid water in a circularly polarized external field

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(Received 1 June 1987; accepted 4 August 1987)

~~The dipole moment of the water molecule has~~ been subjected to an external circularly polarized field of force and the effect on the molecular dynamics evaluated through novel ~~laboratory and moving frame cross~~ correlation functions (ccf 's) using molecular dynamics computer simulation. The field symmetry is such as to make possible the existence in both ~~frames off diagonal elements~~ of the ccf 's which disappear at field off equilibrium. The laboratory frame ccf 's involve simultaneously the rotational and translational motion of the molecule and are not describable with contemporary theories of "rotational" or "translational" diffusion. The simulation shows laboratory frame birefringence in the orientational and rotational velocity autocorrelation functions (acf 's) in the y (or z) and x axes for a field applied in the x axis. This birefringence is linked to the appearance of ccf elements induced by the circularly polarized field, and its experimental measurement would provide indirect information on the time dependence of these ccf 's.

I. INTRODUCTION

The computer simulation of molecular dynamics in the liquid state of water[1–10] is extended in this work to include the effect of a circularly polarized external electric field applied to the sample in the x axis of the laboratory frame. The field induces a corkscrew motion in each water molecule and can be applied experimentally, either with a rotating electric field, or with the electric field component of a laser field at various frequencies in the electromagnetic spectrum. The first theoretical study of a rotating electric field applied to a molecular liquid was carried out by Born,[11] and the first experimental study by Lertes.[12] Born interpreted the effect with the Debye theory of dielectric relaxation,[13] and related the macroscopic torque induced by the rotating electric field to the Debye relaxation time. The Born theory was later greatly extended by Dahler[14] using hydrodynamic theory of structured fluid media.[15] With the computer facilities available to contemporary investigators the phenomenon first described by Born can be studied by simulation on the single molecule level. The approach used in this paper is to construct time correlation functions at field-on equilibrium and to study the symmetry of correlation functions of the type

$$\mathbf{C}_{AB}(t) = \langle \mathbf{A}(t)\mathbf{B}^T(0)\rangle \tag{1}$$

in the laboratory frame, and in the moving frame of reference defined by the three principal moments of inertia of the water molecule. This allows us to interpret the effect of the circularly polarized electric field in terms of symmetry patterns on the fundamental single molecule level. With enough computer power these symmetry patterns could be related to the essentially hydrodynamic theory of Dahler and a correspondence established with the molecular dynamics as interpreted through correlation functions of Eq. (1).

II. COMPUTER SIMULATION METHODS

A standard constant volume computer simulation algorithm (TETRA)[1–8] was modified to include the effect of a right circular polarized electric field of the type

$$E_x = 0; \quad E_y = E_0 \cos \omega t; \quad E_z = E_0 \sin \omega t; \tag{2}$$

on the net molecular dipole moment $\boldsymbol{\mu}$. The interaction between field and dipole moment was assumed to generate the torque

$$T_q = -\boldsymbol{\mu} \times \mathbf{E} \tag{3}$$

for each molecule in the ensemble. The net effect on the mean internal energy was to increase this form -35.2 kJ/mol at field-off equilibrium to -21.0 kJ/mol at field-on equilibrium. Both quantities were evaluated over 6000 time steps of 0.5 fs each. The torque, Eq. (3), was coded into TETRA at the point where the net torque on each molecule was evaluated from the atom–atom intermolecular forces. The resultant torque on each molecule was therefore a sum of internal and external terms. The algorithm then proceeds to compute dynamical quantities such as the molecular angular velocity, linear velocity, orientation, rotational velocity, and so on using the standard methods of computer simulation.[14] The temperature of the sample was kept near the input level by temperature rescaling, which effectively removed the extra deterministic contribution to the rotational and translational temperature introduced by the externally applied torque (3). This method is similar to that used elsewhere in the literature to simulate the effect of a uniaxial electric field applied in the z axis of the laboratory frame.[15–20]

The intermolecular potential was a five by five site–site type with Lennard-Jones atom–atom terms for the hydrogen and oxygen interactions and charge–charge terms consisting of positive charges on the hydrogens and negative charges near the lone pair sites. The potential parameters (in standard notation) were

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0021-9606/87/226040-06$02.10

$$\epsilon/k(\mathrm{O{-}O}) = 58.4\ \mathrm{K};\quad \sigma(\mathrm{O{-}O}) = 2.8\ \text{Å};$$
$$\epsilon/k(\mathrm{H{-}H}) = 21.1\ \mathrm{K};\quad \sigma(\mathrm{H{-}H}) = 2.25\ \text{Å};$$
$$q_O = -q_H = -0.231|e|.$$

After equilibration the auto- and cross-correlation functions were evaluated over 6000 time steps of 0.5 fs each recorded every two steps, providing 3000 configurations of 108 molecules each. Running time averages were constructed over the complete 3000 records for each correlation function described in this paper.

III. RESULTS AND DISCUSSION

The circularly polarized electric field induces birefringence in the orientational autocorrelation function (acf) as illustrated in Fig. 1. This is the correlation function (cf) of the dipole unit vector normalized in the x, y, and z directions:

$$\mathbf{C}_1^{ij}(t)_{xyz} = \frac{\langle \mu_i(t)\mu_j(0)\rangle}{\langle \mu_i^2\rangle^{1/2}\langle \mu_j^2\rangle^{1/2}}.$$

The phenomenon illustrated in Fig. 1 is in the class of birefringence phenomena typified by the Kerr and Faraday effects,[21] but has not been reported for a circularly polarized field of the Eq. (2). Figure 1 also suggests that birefringence would also be induced by a circularly polarized electromagnetic field, and this would be akin to the Buckingham effect.[22] The birefringence effect of Fig. 1 is observable experimentally, and its full interpretation requires consideration of the simultaneous rotation and translation of a diffusing asymmetric top such as water. This is possible only with computer simulation, because contemporary analytical theories of diffusion[23–28] are intractable. This point is amply illustrated in this paper by reference to ccf's which are obtainable by simulation but which diffusion theory cannot describe. The z and y components of the orientational acf have a different time dependence in the laboratory frame which we ascribe to artifact or residual noise due to small sample size. This may be reflected furthermore in the small negative intercept at $t = 0$ in Fig. 2. The orientational acf components are more noisy than the equivalents in the other ccf's observed in this work.

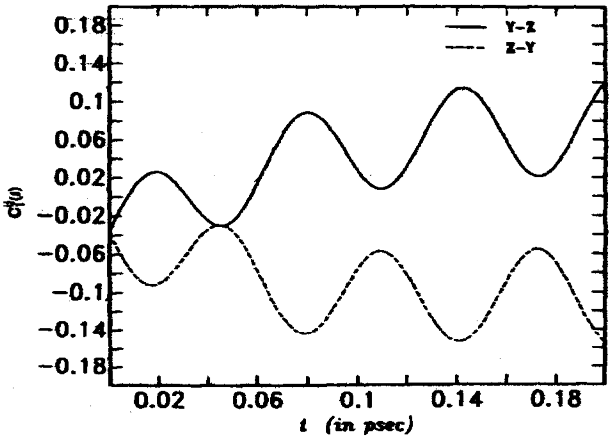

FIG. 2. (y,z) and (z,y) elements of orientational correlation tensor $\mathbf{C}_1(t)$.

Figure 2 is an example of laboratory frame ccf elements of this type, for the off diagonal (y,z) and (z,y) elements of the orientational ccf tensor $\mathbf{C}_1$. At field-off equilibrium these elements should vanish, and any analytical theory of birefringence of the type illustrated in Fig. 1 would have to describe the symmetrical time dependence of the off-diagonal elements of Fig. 2. The other off-diagonal elements of $\mathbf{C}_1$ are much smaller in amplitude, so that the complete symmetry effect of the R circularly polarized electric field is

$$\mathbf{C}_1(t) = \begin{bmatrix} + & 0 & 0 \\ 0 & + & 0 \\ 0 & 0 & + \end{bmatrix}_{(x,y,z)} \rightarrow \begin{bmatrix} + & \delta & \delta \\ \delta & + & + \\ \delta & - & + \end{bmatrix}_{(x,y,z)},$$

where δ represents an element with a small normalized amplitude and the + or − signs indicate that the off-diagonal elements exist and are antisymmetric in time dependence.

Figure 3 shows that the birefringence also appears in the rotational velocity correlation tensor

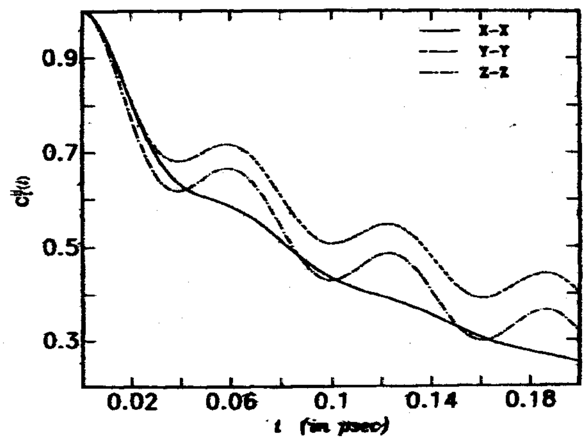

FIG. 1. The diagonal elements of the orientational correlation tensor $\mathbf{C}_1(t)$.

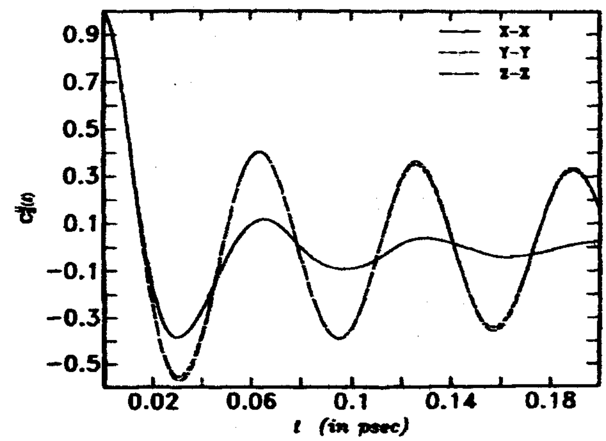

FIG. 3. Diagonal elements of $\mathbf{C}_2(t)$.

$$C_2^{ij}(t)_{xyz} = \frac{\langle \dot{\mu}_i(t)\dot{\mu}_j(0)\rangle}{\langle \dot{\mu}_i^2\rangle^{1/2}\langle \dot{\mu}_j^2\rangle^{1/2}},$$

where $\dot{\mu}$ is the time derivative of the dipole unit vector. This implies that the R circularly polarized electric field would induce birefringence in the far infrared spectrum of liquid water, providing an optical birefringence in addition to the dielectric birefringence of Fig. 1, and another experimental method of investigation. The rotational velocity correlation tensor is dominated in the off-diagonal elements by the (y,z) and (z,y) components which are antisymmetric (Fig. 4). The (x,z) and (z,x) elements are also antisymmetric but of much lower amplitude, and the remaining two are indistinguishable from noise. The symmetry effect of the field is therefore as follows:

$$\mathbf{C}_2(t) = \begin{bmatrix} + & 0 & 0 \\ 0 & + & 0 \\ 0 & 0 & + \end{bmatrix}_{(x,y,z)} \rightarrow \begin{bmatrix} + & \delta & - \\ \delta & + & + \\ + & - & + \end{bmatrix}_{(x,y,z)}.$$

In contrast, as shown in Fig. 5, there is no laboratory frame anisotropy in the center of mass velocity acf's so that the corkscrew effect of the applied electric field does not affect the isotropy of the laboratory frame diagonal elements of the tensor:

$$C_3^{ij}(t)_{xyz} = \frac{\langle v_i(t)v_j(0)\rangle}{\langle v_i^2\rangle^{1/2}\langle v_j^2\rangle^{1/2}}.$$

The (y,z) and (z,y) off-diagonal elements of this tensor in the laboratory frame are again antisymmetric in time dependence, but barely rise above the background noise. The other four off-diagonal elements are indistinguishable from noise. The overall effect of the right circularly polarized field is therefore

$$\mathbf{C}_3(t) = \begin{bmatrix} + & 0 & 0 \\ 0 & + & 0 \\ 0 & 0 & + \end{bmatrix}_{(x,y,z)} \rightarrow \begin{bmatrix} + & \delta & \delta \\ \delta & + & - \\ \delta & + & + \end{bmatrix}_{(x,y,z)}.$$

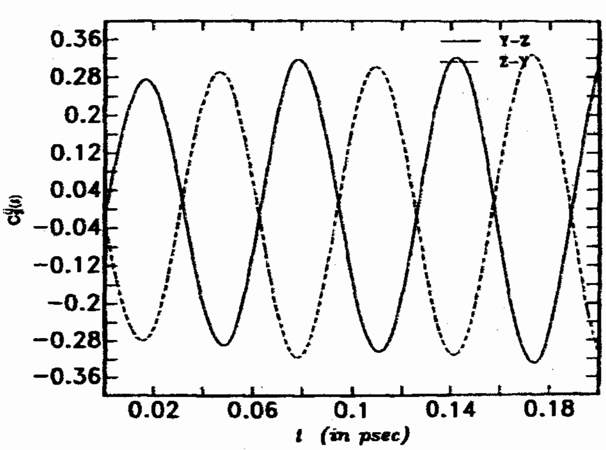

FIG. 4. (y,z) and (z,y) elements of $\mathbf{C}_2(t)$.

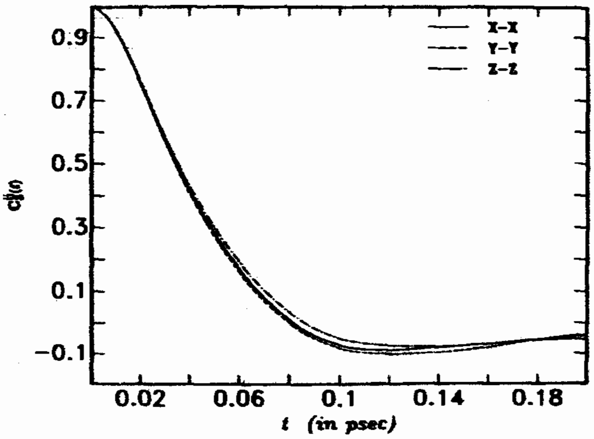

FIG. 5. Diagonal elements of $\mathbf{C}_3(t)$.

The diagonal elements of the angular velocity correlation tensor

$$C_4^{ij}(t)_{xyz} = \frac{\langle \omega_i(t)\omega_j(0)\rangle}{\langle \omega_i^2\rangle^{1/2}\langle \omega_j^2\rangle^{1/2}}$$

are evidently anisotropic, although smaller than that in either the orientational or rotational velocity correlation tensor. However, the (y,z) and (z,y) off-diagonal elements are much greater in amplitude than the other four off-diagonal terms. The (x,z) and (z,x) elements are finite in time dependence but disappear in the background noise after 0.06 ps. The (y,x) and (x,y) elements are symmetric, but of much lower amplitude. The overall symmetry effect of the right circularly polarized field is therefore

$$\mathbf{C}_4(t) = \begin{bmatrix} + & 0 & 0 \\ 0 & + & 0 \\ 0 & 0 & + \end{bmatrix}_{(x,y,z)} \rightarrow \begin{bmatrix} + & + & + \\ - & + & + \\ - & - & + \end{bmatrix}_{(x,y,z)}.$$

The intricate interrelation between rotational and center of mass translational molecular diffusion has been demonstrated recently in terms of the molecular noninertial accelerations and is evidenced in Figs. 6 and 7 for the ccf of the

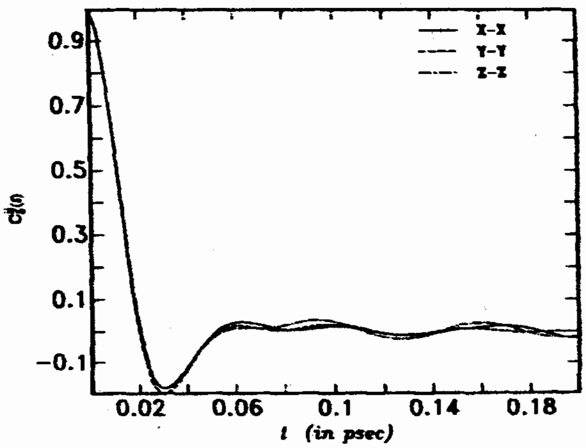

FIG. 6. Diagonal elements of $\mathbf{C}_3(t)$.

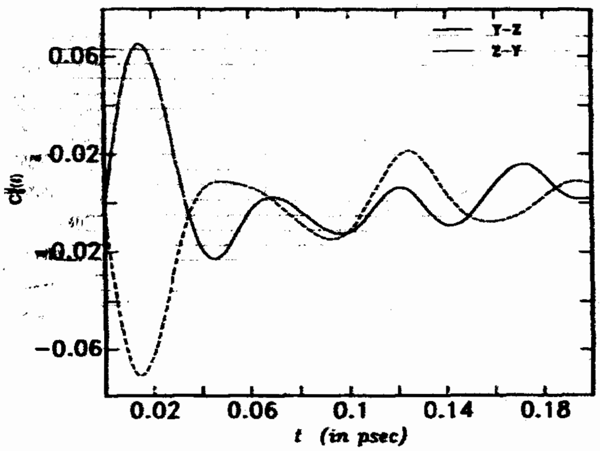

FIG. 7. (y,z) and (z,y) elements of $C_5(t)$.

Coriolis acceleration

$$C_5^{ij}(t)_{xyz} = \frac{\langle [\mathbf{v}(t)\times\boldsymbol{\omega}(t)]_i [\mathbf{v}(0)\times\boldsymbol{\omega}(0)]_j\rangle}{\langle[\mathbf{v}(0)\times\boldsymbol{\omega}(0)]_i^2\rangle^{1/2}\langle[\mathbf{v}(0)\times\boldsymbol{\omega}(0)]_j^2\rangle^{1/2}}$$

whose diagonal (acf) elements are clearly isotropic in the laboratory frame of reference. Again the (y,z) and (z,y) elements are dominant in the off-diagonal terms and antisymmetric (Fig. 7). The other four off-diagonal elements are indistinguishable from noise. The overall effect of the field on the Coriolis acceleration tensor is therefore

$$C_5(t) = \begin{bmatrix} + & 0 & 0 \\ 0 & + & 0 \\ 0 & 0 & + \end{bmatrix}_{(x,y,z)} \to \begin{bmatrix} + & \delta & \delta \\ \delta & + & + \\ \delta & - & + \end{bmatrix}_{(x,y,z)}.$$

The recent discovery[29-31] of ccf's direct in the laboratory frame has emphasized the interrelation between rotational and translational diffusion, even for spherical top symmetries. Figure 8 illustrates the isotropy of the diagonal elements of the correlation tensor

$$C_6^{ij}(t)_{xyz} = \frac{\langle [\mathbf{v}(t)\times\boldsymbol{\omega}(t)]_i [\mathbf{F}(0)\times\boldsymbol{\omega}(0)]_j\rangle}{\langle[\mathbf{v}(0)\times\boldsymbol{\omega}(0)]_i^2\rangle^{1/2}\langle[\mathbf{F}(0)\times\boldsymbol{\omega}(0)]_j^2\rangle^{1/2}}$$

in the laboratory frame of reference. Here $\mathbf{F}$ is the net force on the molecule. In this case the six off-diagonal elements vanish in the noise of the simulation, so that the overall effect of the field is

$$C_6(t) = \begin{bmatrix} + & 0 & 0 \\ 0 & + & 0 \\ 0 & 0 & + \end{bmatrix}_{(x,y,z)} \to \begin{bmatrix} + & \delta & \delta \\ \delta & + & \delta \\ \delta & \delta & + \end{bmatrix}_{(x,y,z)}.$$

In contrast the right circularly polarized laser field affects the symmetry of the ccf matrix

$$C_7^{ij}(t)_{xyz} = \frac{\langle [\mathbf{v}(t)\times\boldsymbol{\omega}(t)]_i [\mathbf{T}_q(0)\times\mathbf{v}(0)]_j\rangle}{\langle[\mathbf{v}(0)\times\boldsymbol{\omega}(0)]_i^2\rangle^{1/2}\langle[\mathbf{T}_q(0)\times\mathbf{v}(0)]_j^2\rangle^{1/2}}$$

as shown in Figs. 9 and 10. In this case the (y,z) and (z,y) off-diagonal elements exist above the noise, and are antisymmetric (Fig. 10) in time dependence. The overall effect of the circularly polarized laser field is therefore

$$C_7(t) = \begin{bmatrix} + & 0 & 0 \\ 0 & + & 0 \\ 0 & 0 & + \end{bmatrix}_{(x,y,z)} \to \begin{bmatrix} + & \delta & \delta \\ \delta & + & + \\ \delta & - & + \end{bmatrix}_{(x,y,z)}.$$

The importance of the noninertial[32-35] velocities and accelerations in the molecular dynamics of liquid water is illustrated in Fig. 11 by reference to the tensor ccf of $\mathbf{r}\times\boldsymbol{\omega}$ in the laboratory frame (x,y,z). Here $\mathbf{r}$ is the position vector of the molecular center of mass and $\boldsymbol{\omega}$ is the molecular angular velocity. The cross product is a linear velocity generated through the fact that the motion of the molecule is "noninertial," dynamically speaking, with respect to the laboratory frame of reference, and vice versa. The diagonal elements of the tensor

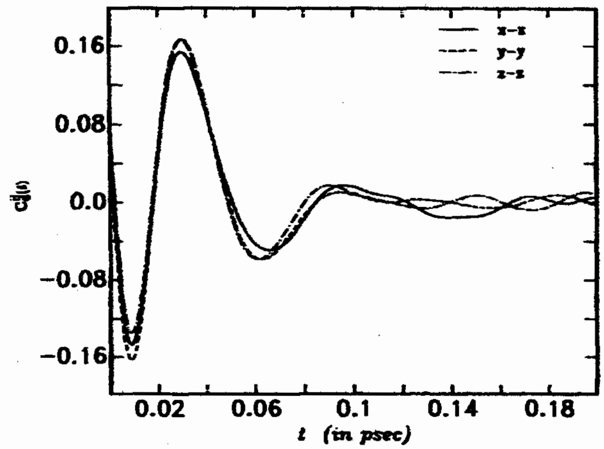

FIG. 8. Diagonal elements of $C_6(t)$.

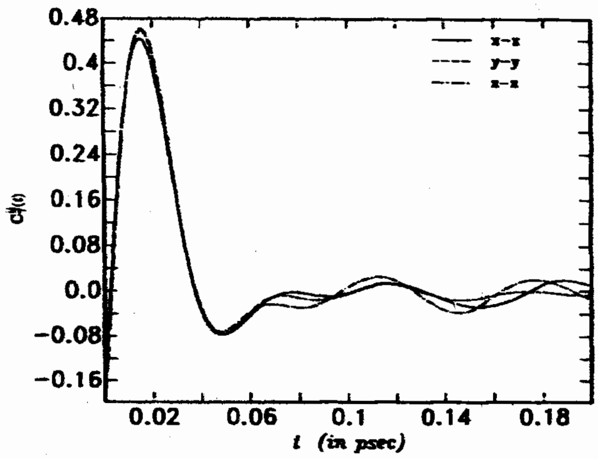

FIG. 9. Diagonal elements of $C_7(t)$.

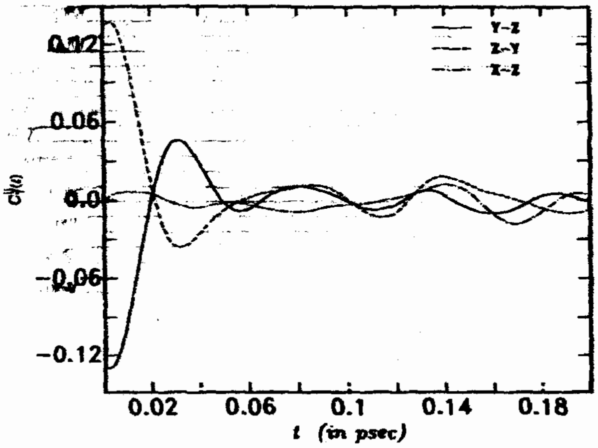

FIG. 10. (y,z) and (z,y) elements of $C_7(t)$.

$$C_8^{ij}(t)_{xyz}=\frac{\langle[\mathbf{r}(t)\times\boldsymbol{\omega}(t)]_i[\mathbf{r}(0)\times\boldsymbol{\omega}(0)]_j\rangle}{\langle[\mathbf{r}(0)\times\boldsymbol{\omega}(0)]_i^2\rangle^{1/2}\langle[\mathbf{r}(0)\times\boldsymbol{\omega}(0)]_j^2\rangle^{1/2}}$$

are slightly anisotropic in the laboratory frame (Fig. 11), and the off-diagonal elements (y,z) and (z,y) are antisymmetric [Fig. 11(b)]. The other two pairs of off-diagonal elements are symmetric in time dependence, but are much smaller in amplitude. The overall effect of the R polarized field is therefore

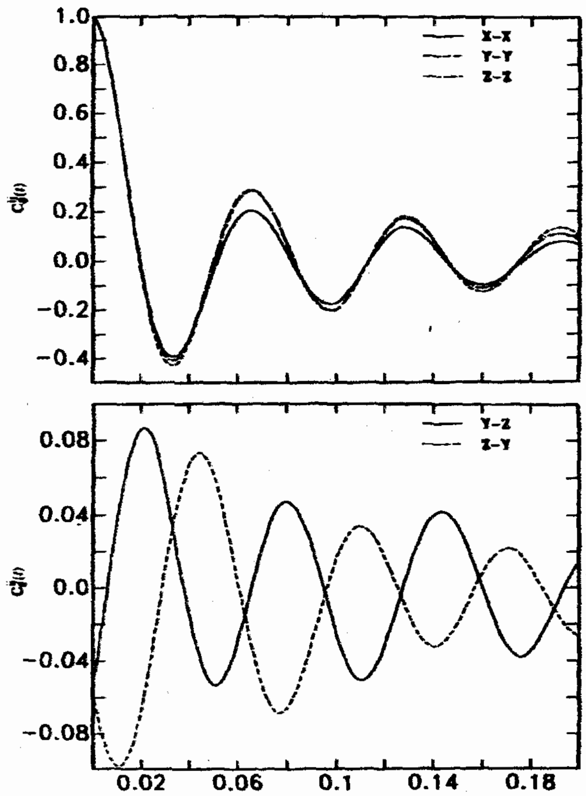

FIG. 11. (a) Diagonal elements of $C_8(t)$; (b) off-diagonal elements (y,z) and (z,y) of $C_8(t)$.

$$C_8(t)=\begin{bmatrix}+&0&0\\0&+&0\\0&0&+\end{bmatrix}_{(x,y,z)}\rightarrow\begin{bmatrix}+&-&-\\-&+&+\\-&-&+\end{bmatrix}_{(x,y,z)}.$$

Switching to the moving frame (1,2,3) defined by the three principal moments of inertia 1, 2, and 3 provides a considerable amount of supplementary information about the molecular dynamics and also about the specific effect of the R polarized laser field on these dynamics.

In Fig. 12 the moving frame anisotropy in the diagonal elements of the ccf tensor

$$C_9^{ij}(t)_{123}=\frac{\langle\omega_i(t)\omega_j(0)\rangle}{\langle\omega_i^2\rangle^{1/2}\langle\omega_j^2\rangle^{1/2}}$$

are illustrated. The anisotropy in the moving frame elements of the molecular center of mass velocity is similarly made more pronounced by the frame transformation, which also affects the time dependence of the three elements of the Coriolis acceleration.

The ccf between the molecular center of mass velocity and the molecular angular velocity in the moving frame of reference is illustrated in Fig. 13. The two nonvanishing terms are the (3,2) and (2,3) elements, which are slightly distorted by the right circularly polarized laser field in that the (2,3) element cuts the y axis below the zero level, and the maximum value reached by the function is less than the 0.15 reached in field-free water. The (3,2) element is more oscillatory than in field-free water and reaches a deeper negative minimum.

The inclusion of noninertial velocities and accelerations in the analysis of molecular diffusion in the liquid state leads to new ccf's for water at field-on equilibrium, in that the field seems to induce a different time dependence in each of the three elements of the ccf matrix

$$C_{10}^{ij}(t)_{123}=\frac{\langle[\mathbf{v}(t)\times\boldsymbol{\omega}(t)]_iv_j(0)\rangle}{\langle[\mathbf{v}(0)\times\boldsymbol{\omega}(0)]_i^2\rangle^{1/2}\langle v_j^2\rangle^{1/2}}$$

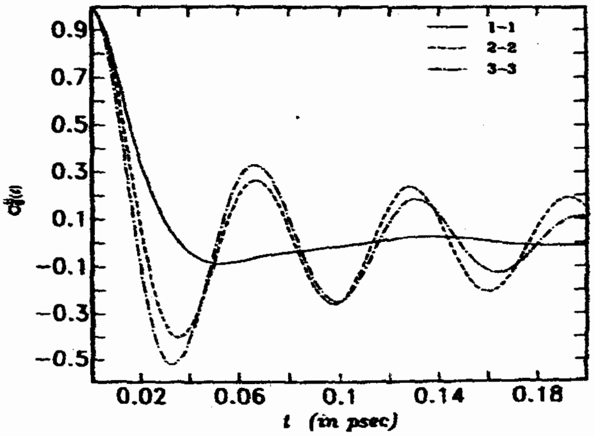

FIG. 12. Diagonal elements of the moving frame angular velocity tensor $C_9(t)$.

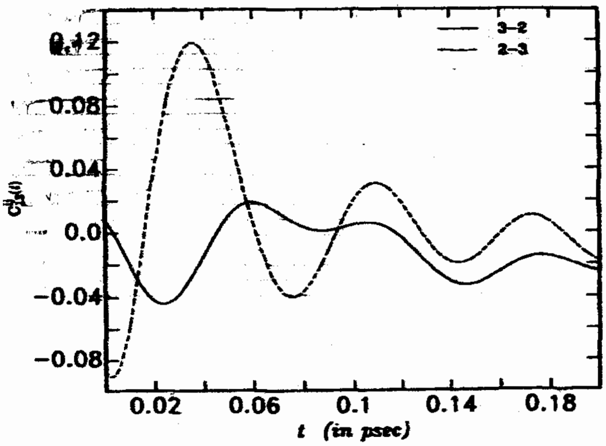

FIG. 13. (3,2) and (2,3) elements of the moving frame tensor $C^{ij}_{12}(t)_{123} = \langle v_i(t)\omega_j(0)\rangle / \langle v_i^2\rangle^{1/2}\langle \omega_j^2\rangle^{1/2}$.

and induces oscillations in two of the elements of the ccf matrix:

$$C^{ij}_{11}(t)_{123} = \frac{\langle [\mathbf{r}(0)\times\boldsymbol{\omega}(t)]_i r_j(0)\rangle}{\langle [\mathbf{r}(0)\times\boldsymbol{\omega}(0)]_i^2\rangle^{1/2}\langle r_j^2\rangle^{1/2}}.$$

The overall effect of the right circularly polarized field on moving frame $\mathbf{C}_9(t)$, $\mathbf{C}_{10}(t)$, and $\mathbf{C}_{11}(t)$ ccf's is

$$\begin{bmatrix} + & 0 & 0 \\ 0 & + & 0 \\ 0 & 0 & + \end{bmatrix}_{(x,y,z)} \rightarrow \begin{bmatrix} + & \delta & \delta \\ \delta & + & \delta \\ \delta & \delta & + \end{bmatrix}_{(x,y,z)},$$

i.e., there is no change of symmetry in the moving frame, in contrast to the results in the laboratory frame described already. In the moving frame the elements that exist for $t > 0$ do so both in the presence and the absence of the field.

The complete set of cross-correlation functions illustrated in these figures may be used to characterize in detail the effect of the R polarized field (2) on the molecular dynamics of water. These results pose a challenge to the theory of molecular diffusion and may be used to supplement at a fundamental level the essentially hydrodynamic approach used by Born, and later by Dahler, to explain the effect of a rotating electric field on a sample of molecular liquid. With sufficient computer power it may soon be possible to explain with computer simulation observable experimental consequences of treating a molecular liquid with a circularly polarized or rotating electric field. An example is the ability of a simple rotating electric field to impose a macroscopic torque on a sample of liquid suspended from a torsion wire in a thin walled glass vessel. The present simulation is an attempt to explore this effect using the torque imposed on each molecule in the water sample.

ACKNOWLEDGMENT

One of us (MWE) would like to thank IBM Kingston for a Visiting Professorship.

[1] G. C. Lie and E. Clementi, Phys. Rev. A 33, 2679 (1986).
[2] M. W. Evans, K. Refson, S. Kandadai, G. C. Lie, and E. Clementi, IBM Kingston Technical Report KGN-111.
[3] M. W. Evans, J. Mol. Liq. 32, 173 (1986).
[4] G. J. Evans, M. W. Evans, P. Minguzzi, C. J. Reid, G. Salvetti, and J. K. Vij, J. Mol. Liq. 34, 285 (1987).
[5] M. D. Morse and S. A. Rice, J. Chem. Phys. 76, 650 (1982).
[6] M. W. Evans, J. Mol. Liq. 33, 255 (1987).
[7] M. W. Evans, J. Mol. Liq. 33, 271 (1987).
[8] D. Bertolini, M. Cassettari, M. Ferrario, P. Grigolini, and G. Salvetti, in *Memory Function Approaches to Stochastic Problems in Condensed Matter*, edited by M. W. Evans, P. Grigolini, and G. Pastori Parravicini, Vol. 62 of the Advances in Chemical Physics series (Wiley, New York, 1985), Chap. VII.
[9] G. C. Lie and E. Clementi, Phys. Rev. A 33, 2679 (1986).
[10] M. W. Evans, K. Refson, K. N. Swamy, G. C. Lie, and E. Clementi, Phys. Rev. A 36, 3935 (1987).
[11] M. Born, Z. Phys. 1, 22 (1921).
[12] D. Lertes, Z. Phys. 2, 87 (1922).
[13] M. W. Evans, G. J. Evans, W. T. Coffey, and P. Grigolini, *Molecular Dynamics* (Wiley, New York, 1982), Chap. 2.
[14] D. W. Condiff and J. S. Dahler, J. Chem. Phys. 44, 3988 (1966).
[15] I. R. McDonald and J. P. Hansen, *Theory of Simple Liquids* (Academic, New York, 1976).
[16] D. Fincham and D. M. Heyes, in *Dynamical Processes in Condensed Matter*, edited by M. W. Evans (Wiley, New York, 1985), Chap. 6.
[17] M. W. Evans, J. Chem. Phys. 76, 5473 (1982).
[18] M. W. Evans, J. Chem. Phys. 76, 5480 (1982).
[19] M. W. Evans, J. Chem. Phys. 77, 4632 (1982).
[20] M. W. Evans, J. Chem. Phys. 78, 925 (1983).
[21] M. W. Evans, J. Chem. Phys. 78, 5403 (1983).
[22] M. W. Evans, Ref. 6, Chap. V.
[23] S. Kielich, in *Dielectric and Related Molecular Processes*, senior reporter M. Davies (Chemical Society, London, 1972), Vol 1.
[24] R. J. Abbott and D. Oxtoby, J. Chem. Phys. 72, 3972 (1980).
[25] For contemporary reviews, see Refs. 6 and 7.
[26] E. Nelson, *Dynamical Theories of Brownian Motion* (Princeton University, Princeton, 1967).
[27] *Organic Liquids, Structure, Dynamics, and Chemical Properties*, edited by A. D. Buckingham (Wiley, New York, 1979).
[28] B. J. Berne and R. Pecora, *Dynamical Light Scattering, with Application to Physics, Chemistry, and Biology* (Wiley, New York, 1976).
[29] *Selected Papers on Noise and Stochastic Processes*, edited by N. Wax (Dover, New York, 1954).
[30] P. G. Wolynes and J. M. Deutch, J. Chem. Phys. 67, 733 (1977).
[31] M. W. Evans, G. C. Lie, and E. Clementi, Z. Physik D (submitted).
[32] M. W. Evans, Phys. Rev. A 34, 2302 (1986).
[33] M. W. Evans, G. C. Lie, and E. Clementi, Chem. Phys. Lett. 138, 149 (1987).
[34] M. W. Evans, Phys. Rev. Lett. 55, 1551 (1985).
[35] M. W. Evans and G. J. Evans, Phys. Rev. Lett. 55, 818 (1985).
[36] M. W. Evans, Phys. Rev. A 33, 1903 (1986).
[37] M. W. Evans and G. J. Evans, Phys. Rev. A 34, 468 (1986).