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Estimation of the Cancellation Effects in Interaction Induced Far-IR Spectra of Liquid Mixtures via MD Simulation. Application to the CCl_4/CS_2 Mixture at Different Mole Fractions

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A molecular dynamics simulation study of the far-infrared spectra in liquid mixture CCl_4/CS_2 at three mole fractions of CCl_4 is reported. The total induced dipole correlation function $C_M(t)$ was separated into the total $C_s(t)$ self- and total $C_+(t)$ cross-correlation functions. Furthermore, the functions $C_s(t)$ were analyzed in terms of the partial self-correlation functions $C_s^A(t)$, $C_s^B(t)$ ($A = CCl_4$, $B = CS_2$), and they have been calculated separately at different concentrations of CCl_4. A more detailed separation of the correlation function $C_M(t)$ into the many-body microcorrelation functions revealed large cancellation effects for the contributions of these functions, which have been found to be dependent on the mole fraction. The obtained far-infrared spectra lie within the frequency range of the corresponding experimental results.

Introduction

In a previous paper[1] we have studied by molecular dynamics (MD) simulation the macroscopic properties of the liquid mixture CCl_4/CS_2. Also, in a subsequent study we have shown that the proposed potential model enables a quantitative prediction of the liquid structure which has been obtained by using "neutron diffraction" experimental techniques.[2] Moreover, the predicted thermodynamic and single-molecule dynamical properties, such as translational and rotational motion,[1,3] were found to be in quite satisfactory agreement with experiment.

The purpose of the present work is to investigate the cancellation effects in interaction induced correlation functions (CF) and to calculate the corresponding far-infrared spectra of this liquid mixture, using MD simulation techniques. Since this potential model can predict certain properties of the liquid mixture, it is of particular interest to investigate its usefulness in analogous studies of interpreting spectral line shapes.[4–7,11]

In certain respects, the nature of such an absorption spectrum is more complicated than the corresponding spectra of the pure liquids CCl_4 and CS_2.

However, it is well-known that the far-IR spectra of nonpolar molecules arise when two or more molecules encounter and acquire induced dipoles through electrical polarization of their electron clouds. In this case the absorption coefficient $a(\omega)$ is a measure of the absorption of radiation due to the time dependence of the total induced dipole $\mathbf{M}(t)$ in the sample.

Usually, in MD treatments the employed mechanism is the interaction of the electrostatic field gradient due to the multipoles on the molecules possessing dipole polarizabilities. On the other hand, a better approximation would have to include higher order multipole and polarizability terms. However, the use of the higher distributed multipoles and polarizabilities has a meaning only when their molecular values are known. Unfortunately, this is not the case for most of the molecular liquids due to several difficulties which arise in relevant experiments and analytical theories.

In the course of this study we are interested in examining the induced dipole CF and the absorption spectrum which results from the two most important induction mechanisms in the mixture, namely, the quadrupolar field of the linear CS_2 and the octupolar field of the tetrahedral CCl_4 molecule. We true to answer the fundamental question of how the immediate neighboring molecules of a molecule determine the induced dipoles at different compositions of the mixture. The results of this study can lead to a better understanding of the behavior of the absorption characteristics.

Theoretical Section

The total induced dipole correlation function (CF) $C_M(t)$ of the sample is defined by the following relation

$$C_M(t) = \langle \mathbf{M}(0) \cdot \mathbf{M}(\mathrm{t}) \rangle \tag{1}$$

The corresponding absorption coefficient $a(\omega)$, or the absorption spectrum, is related to the Fourier transform of the CF $C_M(t)$ by the equation

$$\alpha(\omega) = L \int_0^\infty C_M(t) \cos \omega t \, dt \tag{2}$$

The factor L contains the well-known Polo–Wilson local field correction $[(n^2+2)/3]^2/n$ and the most frequently used empirical quantum mechanical correction $\tanh(\hbar\omega/2KT)/\hbar$:

$$L = \frac{2}{27} \frac{(n^2 + 2)^2 \, \omega \tanh\left(\frac{\hbar\omega}{2KT}\right)}{n\epsilon_0 V c \hbar} \tag{3}$$

where n is the refractive index assumed frequency independent. In the far-infrared spectral range n is generally equal to its high-frequency value. Also, ϵ_0 is the vacuum dielectric permittivity, V is the volume of the molecular system, and c is the speed of light. In order to calculate the absorption spectrum $\alpha(\omega)$, one needs to calculate the total induced dipole moment $\mathbf{M}(t)$ as a function of time. This quantity can be written as a sum of the induced dipoles of pairs μ_{ij}, triplets μ_{ijk}, etc.

$$\mathbf{M}(t) = \sum_{j>i} \mu_{ij} + \sum_{k>j>i} \mu_{ijk} + ... \tag{4}$$

where μ_{ij} denotes the dipole moment that the pair of molecules i and j induce in each other through a given induction mechanism. In an approximation, it has been assumed that the three-body μ_{ijk} and higher terms contribute insignificantly to the total induced dipole $\mathbf{M}(t)$. However, the importance of these terms in liquids at high densities is unclear and remains an interesting question.

For this liquid mixture one has four distinct molecular induction schemes, i.e.

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(a) $CS_2 \leftarrow CS_2$

(b) $CS_2 \leftarrow CCl_4$

(c) $CCl_4 \leftarrow CCl_4$

(d) $CCl_4 \leftarrow CS_2$

In other words, the first term in eq 4 contains induced dipoles μ_{ij} of all possible molecule pairs i (CS_2 or CCl_4) and j (CS_2 or CCl_4)

$$\mathbf{M}(t) = \sum_i \sum_j \mu_{ij}(t) \tag{5}$$

The sum $\sum_j \mu_{ij}$ of the partial induced dipoles determines the total induced dipole on the molecule i

$$\mu_i(t) = \sum_j \mu_{ij}(t) \tag{6}$$

The dynamical variable $\mu_i(t)$ gives information about the relative motion of the reference molecule i with respect to its neighboring molecules, its reorientation in a laboratory frame of reference, and the dynamic fluctuations of the cage molecules.

In a mixture of certain mole fraction, the total number of molecules N contains N_A (A = CCl_4) CCl_4 and N_B (B = CS_2) CS_2 molecules. In this case the total induced dipole moment can be split into a sum of two terms given by the equation

$$\mathbf{M}(t) = \mathbf{M}^A(t) + \mathbf{M}^B(t) \tag{7}$$

where $\mathbf{M}^A(t)$ and $\mathbf{M}^B(t)$ are given by the following equations

$$\mathbf{M}^A(t) = \sum_{i=1}^{N_A} \mu_i^A(t) \tag{8a}$$

$$\mathbf{M}^B(t) = \sum_{i=1}^{N_B} \mu_i^B(t) \tag{8b}$$

The molecular induced dipoles μ_i^A and μ_i^B arise from the induction schemes AA, AB, BB, and BA, respectively. In other words, these quantities may be represented as follows:

$$\mu_i^A(t) = \sum_{j\neq i}^{N_A} \mu_{ij}^{AA}(t) + \sum_{k\neq i}^{N_B} \mu_{ik}^{AB}(t) \tag{9a}$$

$$\mu_i^B(t) = \sum_{m\neq i}^{N_B} \mu_{im}^{BB}(t) + \sum_{n\neq i}^{N_A} \mu_{in}^{BA}(t) \tag{9b}$$

On the basis of this analysis, the total induced dipole CF of the sample may be expressed as a sum of four different CFs

$$C_M(t) = \langle \mathbf{M}^A(0)\cdot\mathbf{M}^A(t)\rangle + \langle \mathbf{M}^A(0)\cdot\mathbf{M}^B(t)\rangle + \langle \mathbf{M}^B(0)\cdot\mathbf{M}^A(t)\rangle + \langle \mathbf{M}^B(0)\cdot\mathbf{M}^B(t)\rangle = C_M^{AA}(t) + C_M^{AB}(t) + C_M^{BA}(t) + C_M^{BB}(t) \tag{10}$$

Each CF in eq 10 may be further analyzed in four CFs of the general form

$$C^{FG,LM}(t) = \sum_{i=1}^{N_F} \sum_{j\neq i}^{N_G} \sum_{k=1}^{N_L} \sum_{l\neq k}^{N_M} \langle \mu_{ij}^{FG}(0)\cdot\mu_{kl}^{LM}(t)\rangle \tag{11}$$

TABLE 1: Values of the CFs $C^{FG,LM}(t)$ at $t = 0$ per Molecule and at Different Mole Fraction $X(CCl_4)$ from This MD Study[a]

		X				
CFs	n-body micro-CFs	0	0.25	0.5	0.75	1
$C^{AA,AA}(0)$	2a, 2b, 3a, 3b, 3c, 4c		2.4	8.8	14.4	19.9
$C^{AA,BA}(0)$	3a, 3b, 4c		0.04	1.8	0.6	
$C^{AA,AB}(0)$	3b, 3c, 4c		−2.0	−6.9	−3.6	
$C^{AA,BB}(0)$	4c		0.3	−1.6	−0.4	
$C^{BA,BA}(0)$	2a, 3a, 3c, 4c		7.2	6.1	3.4	
$C^{BA,AB}(0)$	2b, 3b, 4c		−8.4	−11.8	−4.9	
$C^{BA,BB}(0)$	3b, 3c, 4c		−7.8	−5.7	−2.3	
$C^{AB,AB}(0)$	2a, 3a, 3c, 4c		23.5	32.0	20.1	
$C^{AB,BB}(0)$	3b, 3c, 4c		1.2	1.8	−0.1	
$C^{BB,BB}(0)$	2a, 2b, 3a, 3b, 3c, 4c	18.7	14.3	7.1	2.3	
$C_M(0)/N$		18.7	30.7	31.6	29.9	19.9
$\mu_{eff}/10^{-31}$ C m		4.32	5.5	5.6	5.4	4.5
$\mu_{eff}^{ex}/10^{-31}$ C m[b]		3.44	4.0	4.4	4.5	4.35
density[b] (g/cm³)		1.253	1.362	1.449	1.520	1.583
refr index[b]		1.623	1.561	1.517	1.483	1.457

[a] A stands for molecule CCl_4 and B for molecule CS_2. The employed parameters for CS_2 are $Q = 9.5 \times 10^{-40}$ C m², $\alpha = 9.96 \times 10^{-40} \times$ C² m² J⁻¹, and $\gamma = 11.18 \times 10^{-40}$ C² m² J⁻¹ and for CCl_4 are $\Omega = 416 \times 10^{-51}$ C m³ and $\alpha = 11.68 \times 10^{-40}$ C² m² J⁻¹. [b] Experimental values are taken from ref 11.

where F, G, L, and M indicate the molecule A or B. It may be easily seen that the functions $C^{FG,LM}(t)$ and $C^{LM,FG}(t)$ are identical. With the above analysis and eq 11, the CF $C_M(t)$ may be rewritten in the following form

$$C_M(t) = C^{AA,AA}(t) + 2C^{AA,AB}(t) + C^{AB,AB}(t) + 2C^{AA,BB}(t) + 2C^{AA,BA}(t) + 2C^{AB,BB}(t) + 2C^{AB,BA}(t) + C^{BA,BA}(t) + 2C^{BB,BA}(t) + C^{BB,BB}(t) \tag{12a}$$

More analytically, following the procedure which was first introduced in the study of the total induced dipole CF of the pure liquids N_2, CS_2, and CO_2,[8-10] each term in eq 12a may be decomposed into the n-body CFs with n = 2–4. In this case we obtain the micro-CFs 2a $C_{2a}^{FG,LM}(t)$, 3a $C_{3a}^{FG,LM}(t)$, 2b $C_{2b}^{FG,LM}(t)$, 3b $C_{3b}^{FG,LM}(t)$, 3c $C_{3c}^{FG,LM}(t)$, and 4c $C_{4c}^{FG,LM}(t)$. The exact definition of these "microcorrelation functions" of the type 2a, 3a, 2b, 3b, 3c, and 4c have been introduced in a previous paper.[8] A procedure similar to this one has been used also in ref 9. Thus one can see that the CF $C^{AA,AA}(t)$ may be written in terms of the above n-body CFs as follows

$$C^{AA,AA}(t) = C_{2a}^{AA,AA}(t) + C_{3a}^{AA,AA}(t) + C_{2b}^{AA,AA}(t) + C_{3c}^{AA,AA}(t) + 2C_{3b}^{AA,AA}(t) + C_{4c}^{AA,AA}(t) \tag{12b}$$

while the CF $C^{AA,AB}(t)$ contains the terms $C_{3b}^{AA,AB}(t)$, $C_{3c}^{AA,AB}(t)$, and $C_{4c}^{AA,AB}(t)$ only. Similar relations can be obtained for the other terms in eq 12a. Table 1 gives information about these relations. Thus, there are 36 n-body CFs of different types which determine the total induced dipole CF $C_M(t)$ of the mixture.

All CFs $C^{FG,LM}(t)$ have been calculated at time zero and for the three mole fractions of the mixture. The results are shown in Table 1. The cancellation between different contributions may be also observed.

However, we split the sum in eq 10 into a self- and cross-CF, $C_s(t)$ and $C_+(t)$, defined as follows:

$$C_M(t) = C_s(t) + C_+(t) \tag{13a}$$

$$C_s(t) = \sum_i \langle \mu_i(0)\cdot\mu_i(t)\rangle \tag{13b}$$

$$C_+(t) = \sum_{\substack{i,j\\ i\neq j}} \langle \mu_i(0)\cdot\mu_j(t) + \mu_j(0)\cdot\mu_i(t)\rangle \tag{13c}$$

The molecular induced dipole $\mu_i(t)$ is defined by eq 6. These functions have also been calculated and compared to each other.

Especially, the self-CF $C_s(t)$ results from the partial self-CSs $C_s^A(t)$ and $C_s^B(t)$

$$C_s(t) = C_s^A(t) + C_s^B(t) \qquad (14)$$

Both CFs may be written in terms of the *n*-body 2a and 3a micro-CFs as follows

$$C_s^A(t) = C_{2a}^{AA,AA}(t) + C_{3a}^{AA,AA}(t) + C_{2a}^{AB,AB}(t) + C_{3a}^{AB,AB}(t) + 2C_{3a}^{AA,AB}(t) \qquad (15a)$$

$$C_s^B(t) = C_{2a}^{BB,BB}(t) + C_{3a}^{BB,BB}(t) + C_{2a}^{BA,BA}(t) + C_{3a}^{BA,BA}(t) + 2C_{3a}^{BB,BA}(t) \qquad (15b)$$

The CFs $C_s^A(t)$ and $C_s^B(t)$ have been obtained and discussed at each composition of the mixture.

MD Simulation

All MD simulations were carried out in the microcanonical ensemble (N,V,E). The number of molecules was 500 in a box with the well-known periodic boundary conditions. The starting configuration in all runs was an FCC lattice. The analytical description of the used potential model and the atom–atom potential parameters has been determined in a previous work.[1] The molecules with the smallest concentration in the mixture were located in the free sites randomly created within the FCC lattice. Most of the computational details have been published in our previous paper.[1] The relevant CFs studied in this work were obtained using a method of direct calculation (DC). The length of the simulation runs over which averages were performed was approximately 140 ps for each mixture and 90 ps for the pure liquids.

The calculation of the induced dipoles μ_{ij} in a given configuration was based on the induction mechanism of a quadrupole moment and an anisotropic polarizability on the linear molecule[8] and of an octupole moment and an isotropic polarizability on the tetrahedral molecule.[10] The four induction schemes AA, AB, BA, and BB in analytical form are given as follows:

$$\mathrm{B} \leftarrow \mathrm{B} \quad \mu_{ij}^{BB} = \alpha_i^B \cdot T^{(3)}(r_{ij}) : Q_j^B \qquad (16a)$$

$$\mathrm{B} \leftarrow \mathrm{A} \quad \mu_{ij}^{BA} = \alpha_i^B \cdot T^{(4)}(r_{ij}) \vdots \Omega_j^A \qquad (16b)$$

$$\mathrm{A} \leftarrow \mathrm{B} \quad \mu_{ij}^{AB} = \alpha_i^A \cdot T^{(3)}(r_{ij}) : Q_j^B \qquad (16c)$$

$$\mathrm{A} \leftarrow \mathrm{A} \quad \mu_{ij}^{AA} = \alpha_i^A \cdot T^{(4)}(r_{ij}) \vdots \Omega_j^A \qquad (16d)$$

$T^{(3)}(r_{ij})$ and $T^{(4)}(r_{ij})$ are the third and fourth rank tensors defined by the expressions $\nabla^3(1/r_{ij})$ and $\nabla^4(1/r_{ij})$, respectively.

Results and Discussion

Induced Dipole CFs. The induced dipole CFs $C_M(t)$ for the three mole fractions of the mixture as well as the corresponding self- and cross-CFs $C_s(t)$ and $C_+(t)$, obtained from eqs 12a–c, are shown in Figure 1. They are normalized to the self-CF at each mole fraction. We may observe that the function $C_M(t)$ has a small amplitude in contrast to the self-term $C_s(t)$. This is due to the large negative value of the cross-CF. On the other hand, if we compare these results with those of the pure liquids CS_2 [8] and CCl_4,[10] we observe that the relaxation of the cross-CFs of the mixtures is faster than those of the pure liquids. This effect leads to the long time tails (LTT) of the total CFs since the CFs $C_s(t)$ decay very slowly. In the zero time shift, the ratio $C_s(0)/C_M(0)$ shows a clear concentration dependence; increasing the concentration of CS_2 in the mixture increases the above ratio.

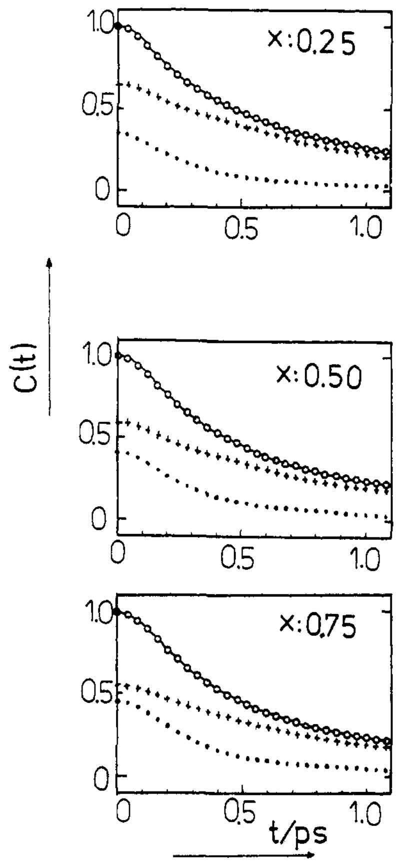

Figure 1. Comparison of the simulated total CF (···), self-CF (○), and cross-CF (+) at three mole fractions X(CCl_4). All CFs are normalized to the self-CFs.

The calculated CFs $C_s(t)$, $C_s^A(t)$, and $C_s^B(t)$ are illustrated in Figure 2. From Figure 2a we may conclude that increasing the CS_2 in the mixture causes the relaxation of the CFs $C_s(t)$ to become faster at short times and up to 0.3 ps, while from this point on and throughout the long time region the relaxations become slower.

A more clear situation appears in the case of the partial self-CFs, $C_s^A(t)$ and $C_s^B(t)$. From Figure 2b and 2c it is easy to see that decreasing the CS_2 concentration in the mixture causes both CFs to decay faster and consequently the corresponding correlation times to become smaller.

Although the partial self-CFs may contain information about molecular collective motion, in a first approximation they are related to the characteristic motions of the individual molecules. In the previous MD simulation study of liquid CS_2,[8] we found that the self-CFs $C_s(t)$ and the two-body CF $C_{2a}(t)$ are almost identical. This is due to the three-body CF $C_{3a}(t)$, which in the total time domain has been found to have a much smaller amplitude compared to the amplitude of the $C_{2a}(t)$ CF.

If one applies the above finding to the discussion here, we may conclude that the $C_s^A(t)$ and $C_s^B(t)$ more or less will be formed by contributions of the CFs $C_{2a}^{AA,AA}(t)$, $C_{2a}^{AB,AB}(t)$, and $C_{2a}^{BB,BB}(t)$, $C_{2a}^{BA,BA}(t)$, respectively.

In order to estimate the different contributions to the total induced dipole CF and to the total self-CF, we have calculated the micro-CFs at $t = 0$, and after that, the corresponding amplitudes of the CFs $C^{FG,LM}(t)$ (see Table 1) were obtained. These results give information about the cancellation effects which take place between different kinds of CFs in the mixture. As we see in Table 1, the amplitudes of some of these 10 CFs $C^{FG,LM}(t)$ are positive and the rest are negative. The CFs with upper indices (AA,AA), (AB,AB), (BA,BA), and (BB,BB) are positive at zero time shift and predominate over the strong negative CFs of the type (AA,AB), (BA,AB), and (BA,BB). We also observe that

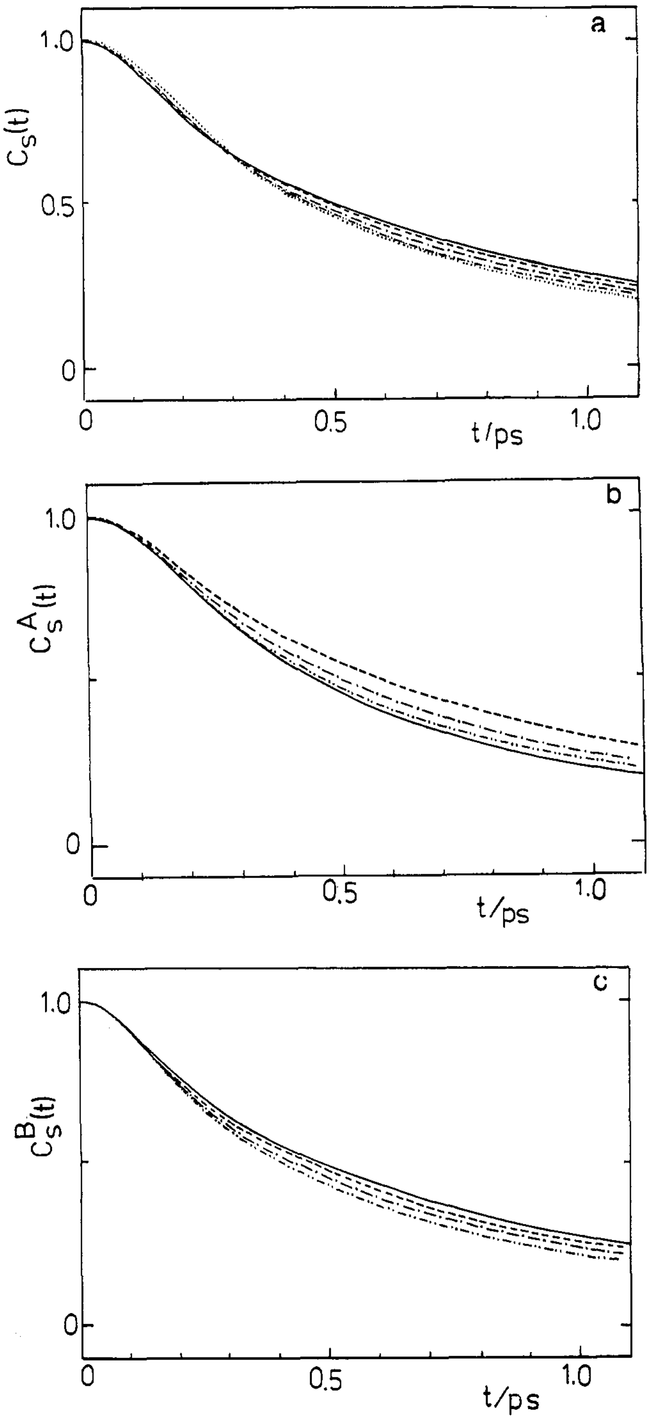

Figure 2. Comparison of the self-CFs at different concentrations of CCl_4: (a) total self-CFs $C_s(t)$ at mole fractions (—) 0.0, (- - -) 0.25, (-·-) 0.5, (-··-) 0.75, and (···) 1.0; (b) partial self-CFs $C_s^{CCl_4}(t)$ at mole fractions (—) 1.0, (-··-) 0.75, (-·-) 0.5, and (- - -) 0.25; (c) partial self-CFs $C_s^{CS_2}(t)$ at mole fractions (—) 0.0, (- - -) 0.25, (-·-) 0.50, and (-··-) 0.75.

all negative terms, as in the case of the CF $C^{AB,BA}(t)$, contain micro-CFs of type two, three, and four body. As in the case of pure CS_2, some of these functions have negative values at $t = 0$.

Finally, in Table 1 we have tabulated the calculated effective dipole moments μ_{eff} per molecule, obtained from the total induced dipole CFs $C_M(0)$ and from the corresponding total self-terms $C_s(0)$, at each mole fraction of the mixture. The corresponding experimental effective dipole moments have been estimated previously.[11]

Far-IR Absorption Spectra. Figure 3 displays the calculated far-infrared spectra of this liquid mixture at three mole fractions. The spectra have been calculated by Fourier transformation of the three-variable Mori fits of the simulated total induced dipole CFs $C_M(t)$. In order to check the model used, we have compared the results of this work with the corresponding experimental spectra.[11] Inspecting these figures, we see that the calculated spectra lie within the range of the experimental values. However, the amplitudes of the experimental spectra lie somewhat below the calculated spectra. Also, the position of the calculated absorption maximum is located at some lower frequency values relative to the experimental data. On the other hand, increasing the CS_2 concentration in the mixture shifts the peak positions of the experimental spectra to higher frequencies. This spectral feature is qualitatively reproduced by our MD simulated spectra.

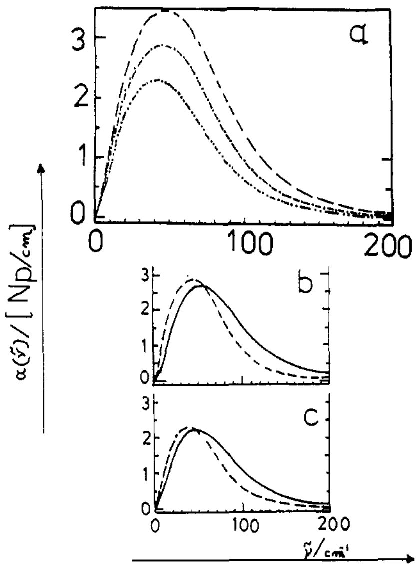

Figure 3. Comparison between experimental and simulated spectra of liquid CCl_4/CS_2 mixtures: (a) calculated spectra at mole fractions (- - -) 0.25, (-·-) 0.50, and (-··-) 0.75. In parts b (X = 0.50) and c (X = 0.75), the solid line represents experimental and the dashed line represents MD results.

The observed discrepancy remains an open question, and it may be explained on the basis of the truncation of the employed first-order induction mechanisms. It is possible that higher multipoles, polarizabilities, and field gradients could account in part for this discrepancy. However, in order to establish this, we need to know accurate values for the higher order electric moments and polarizabilities like hexadecapole moments of both species, which at this stage unfortunately are unavailable.

Summary

In the present work, an MD simulation technique was used to study the interaction induced far-IR absorption spectrum of the liquid mixture CCl_4/CS_2 at three mole fractions of CCl_4. The main results are summarized as follows.

(a) The most interesting induced dipole CFs $C_M(t)$, $C_s(t)$, and $C_+(t)$ were calculated and compared. Furthermore, the CF $C_s(t)$ was separated into the partial self-CFs $C_s^A(t)$ and $C_s^B(t)$. These functions show a noticeable concentration dependence.

(b) A further analysis of the CF $C_M(t)$, as the sum of 10 different CFs of the general type $C^{FG,LM}(t)$, given by eqs 11 and 12, has been presented. This analysis was completed by the evaluation of the various n-body micro-CFs, using the same procedure as applied in the case of the pure liquid CS_2.[8] In this way, it was possible to estimate more precisely the well-known "cancellation effects" affecting each CF of the above type and the total induced dipole CF $C_M(t)$ at $t = 0$.

(c) The effective dipole moments as well as the far-IR spectra have been calculated and compared with the corresponding experimental results. Although the MD results show some deviations from the experimental spectra, they lie within the experimental frequency range.

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