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Viscoelastic behavior of dense microemulsions

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We have performed extensive measurements of shear viscosity, ultrasonic absorption, and sound velocity in a ternary system consisting of water–decane–sodium di(2-ethylhexyl)sulfosuccinate(AOT), in the one-phase region where it forms a water-in-oil microemulsion. We observe a rapid increase of the static shear viscosity in the dense microemulsion region. Correspondingly the sound absorption shows unambiguous evidence of a viscoelastic behavior. The absorption data for various volume fractions and temperatures can be reduced to a universal curve by scaling both the absorption and the frequency by the measured static shear viscosity. The sound absorption can be interpreted as coming from the high-frequency tail of the viscoelastic relaxation, describable by a Cole-Cole relaxation formula with unusually small elastic moduli.

The study of multicomponent supermolecular liquids, consisting of mixtures of oil, water, and surfactant, forming microemulsions, has recently become a very active field of research, both experimentally and theoretically. Some microemulsions can be effectively described as a collection of semimacroscopic surfactant-coated water droplets dispersed in a continuous oil medium having a weak interparticle attraction.[1,2] A wide variety of physical phenomena can be observed in these supramolecular liquids, for example, the critical[3-7] and percolation[8-13] phenomena or the glasslike behavior in dense phases.[14,15]

The behavior of microemulsions near the consolute point has been studied by static and dynamic light scattering.[3-7] Both the scattered intensity and the order parameter relaxation rate show a typical behavior which can be interpreted in terms of the well-established theories of critical phenomena. The critical indices have values showing that microemulsions belong to the same universality class as multicomponent liquid mixtures near a consolute point. The only difference between multicomponent molecular liquids and microemulsions lies in the fact that the smallest length scale in microemulsions is the size of the droplets, a feature that becomes significant for the interpretation of the static and dynamic light scattering data, when the system is not very close to the consolute point.[6,7]

The other striking property of microemulsions that has been carefully studied is the electrical conductivity percolation phenomena.[8-13] Both static and dynamic percolation phenomena hae been investigated by means of electrical conductivity and dielectric constant measurements. The results show a scaling behavior of the dielectric relaxation in terms of frequency, composition, and temperature. The overall percolation behavior is now accurately known.[13] When below the percolation threshold, a dynamic percolation picture holds, which relates the effect to hopping of surfactant molecules within transient clusters formed by microemulsion droplets. However, above the threshold the static percolation picture holds.[12]

Finally, a glasslike behavior in dense microemulsions has been shown by dynamic light-scattering experiments.[14,15] These measurements give the time correlation function of the droplet density fluctuations, which deviates from an exponential decay. The density correlation function follows, instead, a stretched exponential law of the Kohlrausch-Williams-Watts[16,17] type. This latter fact, which is characteristic of glassy systems, is related to the existence of a constrained diffusion,[14] which may be similar to the conduction by hopping of the surfactant molecules hypothesized in the dynamic percolation picture.

In this work we show that sound propagation in microemulsions has a novel feature that is distinct from the behavior of a typical dense molecular liquid. In fact it shows viscoelasticity, which is related to the anomalous increase of the static shear viscosity and/or to the rather low elastic moduli in these systems. This effect can be found in the ultrasonic frequency range and is sometimes detected in fluids of relatively small viscosity like systems forming micelles,[18-20] and in highly viscous systems such as polymer solutions, polymers, gels,[21,22] and supercooled liquids.[23] Ultrasonic investigations of not too dense microemulsions have been made and they show anomalous ultrasonic absorption effects related either to critical phenomena[24,25] or to the droplet-shape fluctuations.[26] Volume fraction dependent viscoelastic effects in sodium di92-ethylhexyl)sulfosuccinate (AOT)-decane inverted micellar systems have been recently reported by Ye *et al.*,[27] who performed ultrasonic and light-scattering experiments. These authors observed a frequency-dependent sound velocity and absorption, a behavior which was attributed to the existence of viscoelastic effects.

We have studied the system water–decane–AOT along a line in the ternary phase diagram where the molar ratio of water to surfactant X is held constant to the value 40.8. The volume fraction ϕ of the dispersed phase (water plus surfactant) is determined assuming additivity of the volumes of the three components. For these systems it has been shown by neutron scattering[1,2] that the microemulsion behaves effectively as a two-component mixture of AOT-coated water droplets in oil. When X is kept constant of 40.8, it was shown that the droplets are spherical particles having a constant water core radius of 50 Å. We have measured the static shear viscosity, the sound velocity, and absorption for volume fractions $\phi=0.10$, 0.15, 0.30, 0.50, 0.65, and 0.75 and at two temperatures 23.5 and 36.5 °C. The system shows a lower consolute critical point at the critical volume fraction $\phi_c=0.098$ and the critical temperature T_c is around 40 °C. Below this temperature the microemulsion is in the one-phase region. The choice of ϕ and T has been guided by the precise knowledge of the percolation line of the system, which has been determined at the same time by means of static electrical conductivity.[13] For the two temperatures investigated, the percolation point is at $\phi_p=0.63$ for $T_p=23.5\,^{\circ}\mathrm{C}$ and 0.11 for 36.5 °C. The low-conductivity region occurs at low temperatures and the high-conductivity region at high temperatures.

The static kinematic viscosity η_s/ρ measurements have been performed using a Ubbelhode capillary viscometer immersed in a thermostatic bath. The mass density of the mixtures ρ was measured for some compositions by means of a piknometer in order to establish the validity of a linear law of variation of ρ with ϕ. This fact has been also verified in the same system, and under the same conditions, in an independent measurement.[28] The accuracy of the shear viscosity measurements, which range from 0.6 to 100 cP, is about ±0.5%.

The static shear viscosity η_s measurements are reported in Fig. 1, and show a rapid increase as a function of ϕ, for both temperatures we investigated. These results are in very good agreement with the available experimental data.[15,28,29] We analyzed our measurements in terms of the empirical law due to Krieger[30]

$$\frac{\eta_s}{\eta_d}=\left[1-\frac{\phi}{\phi_0}\right]^{-1/\nu}, \qquad (1)$$

with η_d the measured shear viscosity of decane, and ν and ϕ_0 two free parameters. It has been possible to fit our data and those of Refs. 15 and 29 for the low temperature (23.5 °C) with the choice $\nu=0.4$ and $\phi_0=0.65$, while the same numerical value of $\nu=0.4$ and $\phi_0=0.33$ are the best parameters for the high-temperature measurements (36.5 °C) and the ones of Ref. 29. In Fig. 2 we report a universal plot of the scaled shear viscosity $(\eta_s/\eta_d)^{-\nu}$ as a function of the scaled volume fraction ϕ/ϕ_0. The empirical law is represented here as a straight-line intersecting the axes at 1. The experimental points follow this law up to $\phi/\phi_0\approx 0.6$. We note from Fig. 1 that there is no clear evidence of an anomaly of the shear viscosity as a function of ϕ due to the crossing of the percolation line, both for low and high temperature. On the other hand, the absence of scaling of the shear viscosity, Fig. 2, could be related to percolation; in fact, the scaled volume fraction at percolation is $\phi_p/\phi_0\approx 0.9$ and 0.3 at low and high temperatures, respectively. The viscosity increase is attributed to the percolation behavior of the system in a recent study of shear viscosity in microemulsions by Peyrelasse, Moha-Ouchane, and Boned.[11] These authors show that the anomaly in viscosity can be interpreted in terms of power laws, like electrical conductivity, only when the difference in viscosity of the two-liquid components of the microemulsion is very large, as it is the case in some waterless microemulsions.

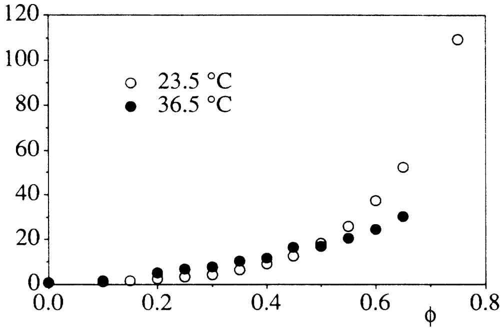

FIG. 1. Measured static shear viscosity for the two temperatures we investigated (23.5 and 36.5 °C).

The ultrasonic absorption and sound velocity measurements have been performed in the frequency range from 5 to 300, and 5 to 45 MHz, respectively. For the absorption measurements we used a standard pulse comparison technique, while for the sound velocity a phase comparison

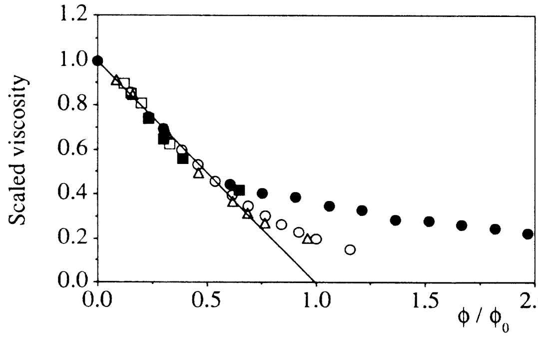

FIG. 2. Scaled viscosity of Eq. (1). The open circle and the solid one refer, respectively, to low and high temperatures. The open square and the solid one to Ref. 29 and the open triangles to Ref. 15.

method has been employed. Both techniques have already been described in detail elsewhere.[31] The ultrasonic cell was a 2.5-cm-diam stainless-steel cylinder with two 5-MHz X-cut quartz transducers. Sample volumes were about 50 ml and the temperature was controlled within ±0.1 °C. The overall accuracy of the sound velocity measurements is of the order of 0.1%, while for sound absorption it is better than 5%.

For all the investigated compositions and temperatures we observed no discernible frequency dispersion in the sound velocity. As shown in Fig. 3 the sound velocity slightly decreases with ϕ in the low-ϕ range (up to 0.20) and then increases at high ϕ for both temperatures. The positions of the sound velocity minima seems to bear no relationship with the corresponding percolation values ϕ_p. The presence of sound velocity minima as a function of ϕ has been previously noticed in Refs. 26 and 27.

The ultrasonic absorption measurements, as expressed by α/f^2, with α the amplitude absorption coefficient and f the frequency, show large frequency-dependent effects. We analyze the overall results according to a viscoelastic behavior assuming for the absorption coefficient the general form[32]

$$\frac{\alpha(f,\phi,T)}{f^2}=\frac{2\pi^2}{\rho c^3}\left[\tfrac{4}{3}\eta_s(f,\phi,T)+\eta_v(f,\phi,T)\right]+B\ , \quad (2)$$

where c the sound velocity and B the constant high-frequency absorption whose relaxation frequency is well above the ultrasonic range. It includes all the high-frequency relaxational processes not due to the frequency-dependent shear viscosity $\eta_s(f,\phi,T)$ and/or volume viscosity $\eta_v(f,\phi,T)$. The two viscosities relax due to structural effects, and we assume a relaxation process described by the well-known Cole-Cole relaxation formula, which physically represents a continuous distribution of processes around an average relaxation frequency[33]

$$\begin{aligned}\eta_s(f,\phi,T)&=\frac{G_\infty\tau_s}{1+(2\pi\tau_s f)^{2(1-\beta)}}\ ,\\ \eta_v(f,\phi,T)&=\frac{K_r\tau_v}{1+(2\pi\tau_v f)^{2(1-\beta)}}\ ,\end{aligned} \quad (3)$$

where β is a distribution parameter for the relaxation processes with characteristic times $\tau_s(\phi,T)$ and $\tau_v(\phi,T)$, respectively, for the shear and volume viscosities. $G_\infty(\phi,T)$ is the shear modulus and $K_r(\phi,T)$ the relaxational part of the compressional modulus.[22] They are related to the static shear and volume viscosities and the corresponding relaxation times through the relations $\eta_s(0,\phi,T)=G_\infty\tau_s$ and $\eta_v(0,\phi,T)=K_r\tau_v$.

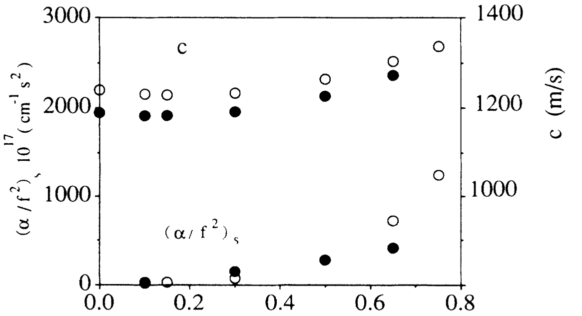

FIG. 3. Measured sound velocity and static absorption due to shear viscosity.

The combined measurements of sound velocity and static shear viscosity allow the evaluation of the static *classical* absorption coefficient due to the shear viscosity alone $[(\alpha/f^2)_s]$:

$$\left[\frac{\alpha}{f^2}\right]_s=\lim_{f\to 0}\frac{\alpha(f,\phi,T)}{f^2}=\frac{8\pi^2}{3\rho c^3}\eta_s(0,\phi,T)\ . \quad (4)$$

The most important feature of our experiments is observed by comparing the measured absorption coefficient $(\alpha/f^2)_{\mathrm{expt}}$ to $(\alpha/f^2)_s$ as a function of frequency (Fig. 4). For relatively small values of ϕ, up to 0.30, we find $(\alpha/f^2)_{\mathrm{expt}}$ greater than $(\alpha/f^2)_s$ while for $\phi\geq 0.30$, $(\alpha/f^2)_{\mathrm{expt}}$ is always smaller than $(\alpha/f^2)_s$. This fact is direct evidence of a viscoelastic behavior, i.e., the relaxation of both shear and volume viscosities with a relaxation frequency in the investigated range. This is an unusual property of the dense microemulsion as compared to the usual molecular liquids, where the bulk viscosity is relaxing in the case of structural relaxation. In order to stress this fact, we report in Fig. 5 the scaled absorption $(\alpha/f^2)_{\mathrm{expt}}/(\alpha/f^2)_s$ as a function of frequency, which shows, for large volume fractions, clear evidence of viscoelasticity.

The sound absorption for $\phi<0.30$ can be interpreted in a usual way as for sound propagation in complex liquids,[19] namely as the superposition of relaxation processes at low and high frequencies. The former are typically due to bulk viscosity, while the latter to various processes beside shear viscosity relaxation. They cause $(\alpha/f^2)_s$ in the low-frequency region to be less than the asymptotic absorption B. For example, in not too dense microemulsions, volume viscosity relaxation may be due to exchange processes of surfactant molecules[24] or to shape fluctuations in the spherical droplets.[26] As ϕ in-

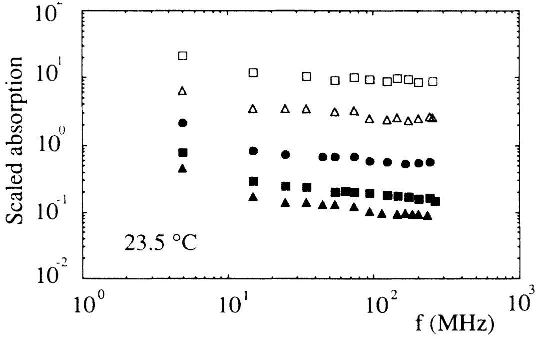

FIG. 4. Scaled absorption $(\alpha/f^2)_{\mathrm{expt}}/(\alpha/f^2)_s$ for low temperature. The open squares and triangles refer to $\phi=0.10$ and 0.30; the solid circles, squares, and triangles to $\phi=0.50$, 0.65, and 0.75, respectively.

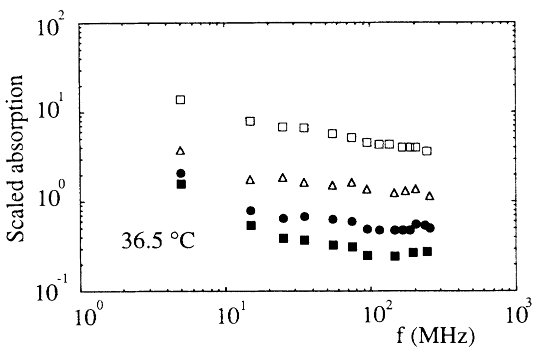

FIG. 5. Scaled absorption $(\alpha/f^2)_{\rm expt}/(\alpha/f^2)_s$ for high temperature. The symbols are the same as in Fig. 4.

creases, the static shear viscosity and the corresponding $(\alpha/f^2)_s$ show a large increase. According to the proportionality between static shear viscosity $\eta_s(0,\phi,T)$ and the relaxation time τ_s, assuming that the elastic moduli are slowly varying functions, we then expect a corresponding increase of τ_s, hence a lowering of the relaxation frequencies. This fact is well known from the theory and experiments on ultrasonic relaxation in viscous molecular liquids where it is often found $\tau_s \approx \tau_v \approx \tau$. We therefore tentatively assume this equality so that form Eq. (2) we derive

$$\frac{\alpha(f,\phi,T)}{f^2}=\frac{A(\phi,T)}{1+(2\pi\tau_s f)^{2(1-\beta)}}+B\ . \qquad (5)$$

The amplitude $A(\phi,T)$, according to our hypothesis, is given by

$$A=\frac{2\pi^2}{\rho c^3}[\tfrac{4}{3}\eta_s(0,\phi,T)+\eta_v(0,\phi,T)]\ . \qquad (6)$$

In order to estimate the relaxational properties of the system, we plot in Fig. 6 the scaled absorption due only to viscosities $[(\alpha/f^2)_{\rm expt}-B]/(\alpha/f^2)_s$ as a function of the frequency scaled with the relaxation frequency given by Eq. (4), i.e., we use a dimensionless scaling parameter $2\pi f\tau_s \propto f\eta_s(0,\phi,T)$.[34] We stress that the classical absorption coefficient is obtained from the combined measurements of shear viscosity, density, and sound velocity. The only parameter that is fixed by inspection of the raw data is the limiting value of the absorption which, together with the measured classical absorption, gives B. The values we get are rather smoothly varying among the different measurements and moreover a slight change of B does not affect the scaled absorption. Once this procedure for the evaluation of B is used, there are no other fitting parameters in our treatment and the data naturally follow a single scaled curve. The fact that the data points scale on a single curve, which is a straight line in a double-logarithmic plot, indicates that we are observing the high-frequency part of a relaxation process. This observation supports, in addition, the simplifying assumption that the shear and volume viscosities have similar relaxation frequencies and give rise to a relaxation with an amplitude which is related to the combination of the static values of the longitudinal viscosity. The relaxational behavior of the system could be interpreted in a different way by supposing a small contribution from the volume viscosity and a dominance of the shear one. However, such possibility is ruled out by the observation that in the low-frequency region of our investigation, the absorption attains values of the order of twice the values obtained from the *classical* absorption of Eq. (4).

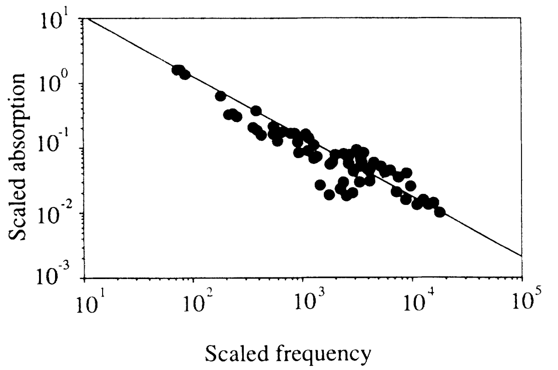

FIG. 6. Scaled absorption as a function of scaled frequency for the mixtures with volume fraction larger than 0.30. The straight line is the Cole-Cole formula of Eq. (5) with the values of the pararmeters given in the text.

In order to proceed further we then assume a relaxation process described by the Cole-Cole formula for frequencies high compared to the typical relaxation frequency. Looking at the double-logarithmic plot in Fig. 6, we see that all the data points, for any f and T and for high volume fractions, fall on a single straight line with a slope $2(1-\beta)=0.92$. This result leads us to exclude a high-frequency single relaxation process of the Debye type, which would imply $\beta=0$, and supports instead a high-frequency Cole-Cole relaxation with $\beta \approx 0.54$.

It is worth noting that since the data roughly fall on the single curve shown in Fig. 6, we are allowed to assume that for high volume fractions of the microemulsion, the parameters appearing in Eq. (6) do not depend strongly on ϕ and T. Therefore we can fit all the measured values, using Eqs. (4)–(6), with the Cole-Cole equation in the high-frequency limit with the relaxation times respectively 23.2, 58.3, and 122 ns for $\phi=0.50$, 0.65, and 0.75 at low temperature and 24.8 and 27.4 ns for $\phi=0.50$ and 0.65 at high temperature. The parameters of the fit are given by $\beta \approx 0.54$, $G_\infty \approx 0.8\times 10^7$ dyn/cm^2 and $K_r \approx 3\times 10^8$ dyn/cm^2. These values of the elastic moduli are small compared to those obtained in associated liquids ($\approx 10^{10}$ dyn/cm), but the same order of magnitude as those found in other dense micellar solutions.[19,20] The relative values of G_∞ and K_r indicate the presence of a very large static volume viscosity, of the order of 40 times the static shear viscosity. In addition, they can explain the lack of dispersion effects on the measured sound velocities. In fact, according to the viscoelastic theory,[22] the sound velocity dispersion is given by

$$\frac{c_\infty - c_0}{c_0}=\frac{2G_\infty}{3\rho c_0^2}\left[1+\frac{3K_r}{4G_\infty}\right]\ , \qquad (7)$$

and, with the values of the elastic moduli we obtain, turns out to be of the order of 10^{-2}, a dispersion which is not observable in the frequency range of our velocity measurements. In fact, our measurements show a dispersion of the order of 1 to 2 m/s in the frequency range from 5 to 45 MHz, which is well above the relaxation frequency and consistent with the previous estimate. The corresponding variation in the measurements of Ye *et al.*[27] is of the order of 4% from the ultrasonic to the hypersonic region. The difference between the two measurements is not very large and can be attributed to the different composition of the samples, i.e., to the presence of water in our system that can vary the elastic moduli and therefore the sound dispersion. The situation is completely different in the case of viscoelastic associated liquids, where the dispersion of sound velocity may be of the order of 50%.

In conclusion we emphasize that the viscoelastic behavior of this water-in-oil microemulsion is observed only for volume fractions of the droplets greater than 0.30. One can speculate on the mechanism underlying the viscoelastic behavior of the dense microemulsions. Since the measurement was made in a one-phase region in the vicinity of the percolation line,[13] as mentioned before, the viscoelastic behavior could be understood from the point of view of the increased connectivity among the droplets, which has been used to explain electrical conductivity in the percolating regime. Since the microemulsion droplet is electrically neutral as a whole, there is a weak, but long-range, attractive part of the interdroplet interaction which may become important when the volume fraction of the droplet is sufficiently high. A transient solidlike network may form, with a characteristic time τ of the order between 0.01 and 0.1 μs. On a time scale longer than τ the system behaves like a collection of isolated droplets undergoing Brownian motions, but in the opposite case one sees the system as formed by fractal clusters on a solidlike network having some finite elastic moduli. A similar picture has been proposed in the work of Ye *et al.*[27] on a dense reverse micellar system we mentioned before. Even though this picture is appealing, we are nevertheless unable to observe a sharp viscoelastic transition at the percolation line. There is an alternative mechanism that may explain the occurrence of viscoelastic behavior for volume fractions larger than 0.30. It is known from Ref. 15 that above volume fractions of 0.40 there is a transition from the isotropic microemulsion phase to an anisotropic lamellar phase at temperature above 35 °C. Thus the solidlike behavior at high volume fractions may be linked to a pretransitional ordering of the surfactant layers. Regardless of the model, the smallness of the elastic moduli reflects that the local solidlike order is rather fragile due to the interdroplet attractive interaction the order of magnitude of which is less than $k_B T$.

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[34] Figure 6 actually reports the scaling variable $2\pi\tau_s f$ since, as we describe in what follows, we are able to estimate the proportionality constant G_∞.