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THERMAL MEASUREMENTS ON SUPERCOOLED LIQUIDS AND EMULSION BY "MIRAGE" DETECTION

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Résumé - La diffusivité thermique de l'eau surfondu et d'émulsions d'eau et d'heptane est déterminée en sondant optiquement la propagation d'ondes thermiques.

Abstract - Thermal diffusivity of supercooled water and water-heptane emulsion is determined by optically probing thermal waves propagation.

Let us recall that when a plane, periodically heated at the frequency \( f = \omega / 2\pi \) is in thermal contact with a medium (thermal conductivity \( k \), density \( \rho \), specific heat \( c \)) whose thermal diffusion length is given by \( \mu = (2k/\rho c\omega)^{1/2} \), the periodic temperature distribution in this medium is given by \( T(x) = T_0 \cos(\omega t - \pi) \cos(\omega t - x/\mu) \) (plane wave), \( T_0 \cos \omega t \) being the periodic temperature at the interface and \( x \) the distance from the heated surface /1/.

If the medium is transparent (refractive index \( n \)), one can probe easily the thermal waves by using the deflection \( \Theta \) of a probe beam propagating parallel to the interface plane over a path length \( \ell \) :

\[
\Theta = \frac{\rho}{n} \frac{dn}{dT} \frac{dT}{dx} = -\sqrt{\frac{\ell}{\mu \pi}} T_0 \cos(\omega t - x/\mu) e^{-x/\mu}
\]

A careful determination of \( \Theta \) (amplitude or phase) as a function of \( x \) leads to the determination of \( \mu /2/\).

We have used this method to measure the thermal diffusion length of water in a small cell (1 x 1 mm² section) which allows us to keep water supercooled down to \(-18^\circ C\). Fig. 1a shows a schematic diagram of the experimental set-up, the cell being in thermal contact with a copper bloc cooled by a flow of gaseous nitrogen.

A typical result obtained with the phase signal is shown on Fig. 2. The experimental values of \( \mu \) are deduced from such curves by using a least square linear regression.

In the case of water, the thermal conductivity temperature dependence is unknown and can be deduced from the temperature dependence of \( \mu \). It is interesting to use this method to study the thermal conductivity of water in supercooled regime.

Actually, a controversy subsists about the physical interpretation of the thermal and dynamic properties of metastable water. Most of these properties exhibit an anomalous temperature dependence which has been interpreted as manifestation of a critical behaviour near \(-45^\circ C\) /3/.

Dynamic properties are particularly affected in critical regime and measurements of viscosity and different relaxation times are in good agreement with the critical

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behaviour interpretation.

It is however possible to take into account the anomalies of water with a geometrical picture where the formation of hydrogen bonds enhance the formation of low density/low mobility patches of water molecules \(^1/4\). In such explanation the anomalies of supercooled water must vanish at some temperatures low enough.

A good test for both theories is the behaviour of thermal conductivity, because it is well known that near a critical point, thermal conductivity has a singular behaviour increasing sharply with decreasing temperature.

Measurements of the thermal conductivity below 0°C have never been performed before, probably because of the incompatibility between the classical methods and the maintenance of the metastable state which needs extremely small volumes.

Our methods seem consequently well adapted and non perturbative. Fig. 3 illustrates this dependence, from this result which constitutes the first measurement of \(k\) below 0°C, one can conclude that \(k\) does not exhibit any critical behaviour down to \(-18°C\). This observation does not preclude of another behaviour at lower temperature.

In order to reach much lower temperature \((-35°C)\) we have recently undertaken the study of the thermal conductivity of micro-emulsions of water in heptane. Because the optical properties of this highly diffusing material prevent us from probing directly the thermal waves in the liquid we have used a slightly different experimental set-up (Fig. 1b). The thermal wave is probed in an adjacent transparent medium constituted by the walls of the glass cell containing the emulsion.

This experiment is more difficult than the preceding one. On the one hand, the larger distance between the heater and the probe beam reduces the signal; on the other hand the alignment of the cell, the probe beam and the resistor are more difficult.

The preliminary results obtained by varying the distance between the probe and the heater are in good accordance with the values which can be calculated using the model of thermal conductivity for a simple granular medium (Fig. 4) \(^5/\).

In conclusion, we would like to outline that the technique which has been used here to probe the emulsions may be easily extended to a large number of applications including thermal measurements and spectroscopy of heterogeneous materials. By probing the thermal waves in a solid of large \(dn/dT\) values, the sensitivity of the method exhibits the large improvement which has already been found by interfacing solids samples with liquids when probing with the mirage effect \(^6/\).

REFERENCES


Fig. 1 - Exponential set-up for supercooled water (a) and emulsion (b).

Fig. 2 - Phase of the signal versus cell displacement for a modulation frequency of 18 Hz.
Fig. 3 - Thermal conductivity temperature dependence for supercooled water.

Fig. 4 - Amplitude and phase of the signal versus heater displacement for emulsion (water 70%, heptane 30%). The modulation frequency was 8 Hz and the temperature 23°C.