# Third sound velocity in thin films of 3He-4He mixtures at low temperature

J.P. Laheurte, J.C. Noiray, J.P. Romagnan

Classification
*Physics Abstracts*
67.60 — 67.70

# Third sound velocity in thin films of $^3$He-$^4$He mixtures at low temperature

J. P. Laheurte, J. C. Noiray and J. P. Romagnan

Laboratoire de Physique de la Matière Condensée (*), Université de Nice, parc Valrose, 06034 Nice cedex, France

(*Reçu le 16 février 1981, accepté le 20 mars 1981*)

**Résumé.** — Nous présentons des mesures de vitesse du 3e son dans des films minces de mélanges $^3$He-$^4$He jusqu'à 0,1 K. Le comportement de cette vitesse à basse température suggère l'existence d'une séparation de phase perpendiculaire aux parois même pour des épaisseurs de film de 3,7 couches.

**Abstract.** — We present third sound velocity measurements in thin films of $^3$He-$^4$He mixtures down to 0.1 K. The low temperature behaviour of this velocity suggests the existence of a phase separation perpendicular to the walls even for a film mixture of 3.7 layer thickness.

In bulk $^3$He-$^4$He mixtures the phase diagram including the superfluid transition line in the homogeneous mixture and the phase separation line is well known [1]. In that case the phase separation appears through a stratification. The influence of dimensionality on the superfluid transition has already been studied both theoretically [2] and experimentally [3-4]. Its effect on the phase separation has not yet fully been investigated. The existence of a phase separation in a two dimensional (2D) $^3$He-$^4$He mixture has theoretically been predicted [5-6]. The diagrams are quite different from the bulk ones : for instance a mixture of average $^3$He molar concentration $\overline{X}$ larger than 0.12 is not supposed to be superfluid unless it separates between two phases. However for thin film we should take into account concentration gradients. The $^4$He atoms are preferentially adsorbed at the walls [7], while the $^3$He atoms are more concentrated near the free surface [8]. A model describing a two layers thick film in which boundaries conditions at walls and free surface directly affect the concentrations in each layer has also recently been proposed [9]. In that case the phase diagram in such system is more complex. To our knowledge, the phase separation, either in strictly 2D mixtures or in thin mixtures films has not yet been experimentally observed. Recent third sound measurements [4] did not show any significant effect which could prove without ambiguity the occurrence of phase separation in such thin films down to 0.5 K. The goal of this work is to extend third sound measurements below 0.5 K in order to detect any special feature coming from a possible phase separation.

Our experimental cell is the same as that described in the work of Laheurte *et al.* [4]. However this cell was cooled down in a $^3$He-$^4$He dilution refrigerator in order to reach a temperature lower than 0.1 K. Also we have introduced a low temperature valve so that the cell can be isolated once the helium mixture film has been put inside. The cell contains a glass substrate (G) on which the third sound velocity is measured by the usual technique [4], and a porous material (P). Due to its large adsorption surface $S$, we can accurately define [4] on the porous material, the number $n_P$ of atoms adsorbed per unit area and the average concentration $\overline{x}_P$ of the film. In our previous experiments [4] the porous material consisted of millipores filters [10], and the two parameters $n_G$ and $\overline{x}_G$ characterizing the film adsorbed on the glass substrate were obtained using a continuum model [11]. To get rid of this indirect determination we use in the present experiment crushed glass as porous material so that $n_P = n_G$ and $\overline{x}_G = \overline{x}_P$. The average size of the grains is about one micron, and the surface adsorption area measured by the classical Brunauer, Emmett and Teller [12] method with nitrogen adsorption is $S = 67\ m^2$. With such a surface, we find that when the temperature is decreased below 0.6 K the number of atoms which condense on the adsorbed film from the vapour phase is negligible, so that the coverage and average concentration of the adsorbed film can be considered as remaining constant in this range of temperature

(*) Laboratoire associé au C.N.R.S. (L.A. 190).

within an accuracy of one percent. Finally we mention that we observe the occurrence of capillary condensation with pure $^4$He when the coverage is larger than a critical value $n_{P_c} = 0.56$ at./Å$^2$. Of course all our experiments are made with coverage values smaller than $n_{P_c}$.

We have studied film mixtures for different average concentrations and coverages, and we report in this letter the most significant results. We present in figure 1, the temperature dependence, below 0.6 K,

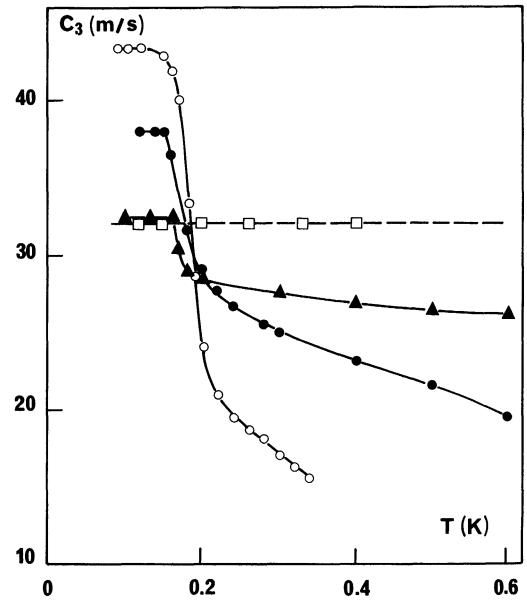


Fig. 1. — Temperature dependence of the third sound velocity at constant coverage ($n_G = 0.50$ at. Å$^{-2}$) for four different average concentrations $\bar{x}$. Pure $^4$He (open square), $\bar{x} = 0.156$ (full triangle), $\bar{x} = 0.33$ (full circle) and $\bar{x} = 0.45$ (open circle).

of the third sound velocity $C_3$ at a given constant coverage $n_G = 0.50$ at./Å$^2$ (which corresponds roughly to a film thickness of 5 atomic layers), for four different average concentrations $\bar{x}_G$. Since the coverage remains constant during the experiment, $C_3$ is expected to depend only on the average superfluid density $\langle \rho_S \rangle$ of the film. For pure $^4$He we know that $\langle \rho_S \rangle$ reaches a constant value at low temperature and indeed experimentally we observe a constant third sound velocity $C_3$. However for mixtures we observe a very striking feature i.e. a sharp rise of $C_3$ in a narrow range of temperature (0.20 K to 0.16 K) to reach a saturated value $C_{3S}$ which remains constant down to the lowest measurable temperature. Moreover we notice that this saturated value $C_{3S}$ increases with the average concentration of the film. This behaviour is quite surprising as it would mean that the average superfluid density increases with the average concentration : to our knowledge there is no mechanism leading to an enhancement of the superfluid density when the $^3$He concentration is increased. Thus it appears that our experimental results below 0.2 K are difficult to understand as third sound velocity measurements in homogeneous superfluid mixture films.

A first possible explanation is that at low temperature we observe a new mode different from the isothermal third sound. We know that when isothermal third sound propagates, evaporation and condensation exchanges between the film and the vapour play an essential role. Now, in our experiments, an estimation of the mean free path in the vapour phase at 0.2 K is about 1 mm which is comparable with the height of the dead free space above the glass substrate. So one could think that, below 0.2 K the evaporation-condensation effects are reduced and that we observe an adiabatic wave motion or temperature mode whose velocity $C_A$, larger than the isothermal one $C_3$, is given by [13]

$$C_A^2 = C_3^2 + \frac{C_5^2}{1 + K^2}. \qquad (1)$$

The last term in (1) is the fifth sound contribution, with $K$ proportional to the amount of evaporation-condensation exchange.

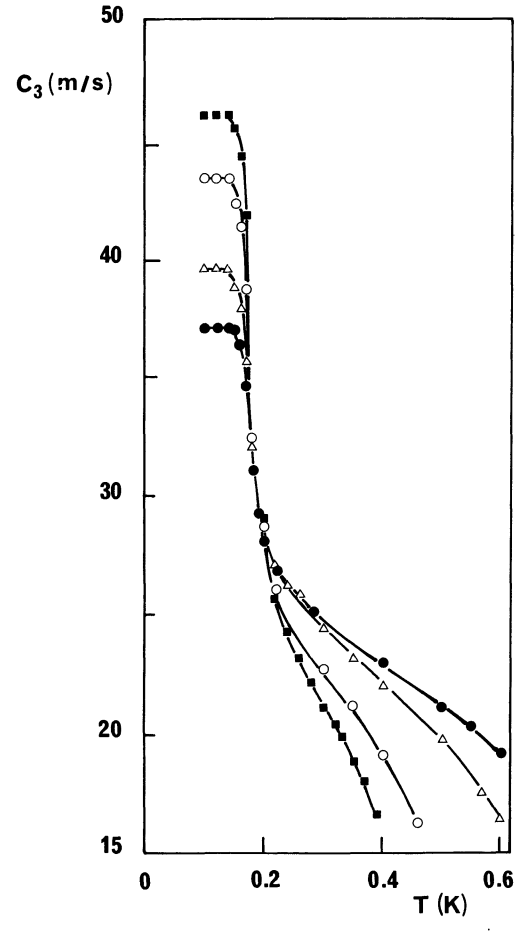


Fig. 2. — Temperature dependence of the third sound velocity at constant average concentration $\bar{x} = 0.33$ for four different coverages. $n_G = 0.40$ at. Å$^{-2}$ (full square), $n_G = 0.43$ at. Å$^{-2}$ (open circle), $n_G = 0.47$ at. Å$^{-2}$ (open triangle), $n_G = 0.50$ at. Å$^{-2}$ (full circle).

In the case of pure $^4$He, this temperature mode has recently been observed on a flat waveguide geometry for thick films [13]; for thin films the fifth sound contribution becomes negligible compared to $C_3^2$. As far as we are concerned no experimental observations are reported for mixture films. So we have repeated some of our experiments using the flat waveguide geometry described in reference [13]; the two flat substrates are separated by 15 μm. The experimental results remain unchanged, leading us to the conclusion that in thin mixture films the fifth sound contribution is negligible and cannot explain the sharp rise of the sound velocity at low temperature.

A second possible explanation, suggested by the dependence of $C_{3S}$ on average concentration, is that the mixture film does not remain homogeneous during the whole run in temperature [14]. We would then observe the occurrence of a phase separation perpendicular to the walls, or stratification, with the superfluid $^4$He rich phase and the normal $^3$He rich phase located respectively near the walls and at the free surface. In that case, for a given coverage, the larger is the average concentration, the thinner is the superfluid phase. As $C_3$ increases when the superfluid phase thickness is reduced, such a mechanism can effectively explain the measured values of $C_{3S}$. To get further information, we have studied the influence of coverage at a given average concentration (0.33). The same striking observation can be made (Fig. 2) and the largest saturated value $C_{3S}$ at low temperature is obtained for the lowest coverage. A very naive interpretation of these results can be made using the relation between the third sound velocity $C_3$ and the average superfluid density $\langle \rho_S \rangle$ [4] :

$$C_3^2 = \frac{\langle \rho_S \rangle}{\langle \rho \rangle} \frac{3\,\theta_G}{d_G^3} \qquad (2)$$

where $\langle \rho \rangle$ is the average density of the superfluid phase of thickness $d_G$ while $\theta_G$ is the Van der Waals interaction constant between a helium atom and the walls. The thickness $d_G$ should of course depend on the concentration of the superfluid phase. However at very low temperatures it is quite plausible that this phase is pure $^4$He or is at least, $^3$He. Thus the real thickness $d_G$ of the superfluid phase should be equal or proportional to $d_4$, the thickness of an $^4$He film that we evaluate assuming all $^4$He atoms are in this superfluid phase. Using $d_G = d_4$ we can get $\langle \rho_S \rangle / \langle \rho \rangle$ at 0.1 K from relation (2). Plotting as usual $\frac{\langle \rho_S \rangle}{\langle \rho \rangle} d_4$ *versus* $d_4$ for two different average concentrations (Fig. 3), we observe a linear behaviour; moreover the intercept with the horizontal axis gives a value of 1.5 atomic layers as in pure $^4$He films [3]. These two points are consistent with the idea of a stratification. However we must point out that the slopes of the two lines, presented in figure 3 are less than one, the value corresponding to pure $^4$He; clearly this result is not consistent with the idea of a superfluid phase constituted of pure $^4$He or dilute in $^3$He. A possible answer to this puzzling question is that, the superfluid phase being not directly in contact with the vapour but with a normal phase, this new boundary condition affects the average superfluid density.

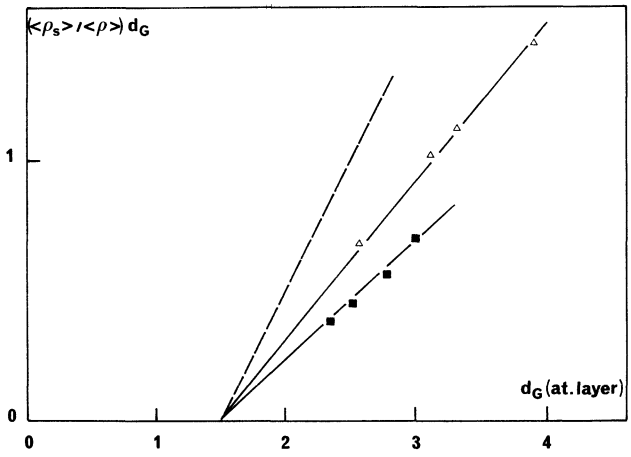


Fig. 3. — $\frac{\langle \rho_S \rangle}{\langle \rho \rangle} d_G$ *versus* the estimated superfluid phase thickness $d_G$ at 0.1 K for two average concentrations $\bar{x} = 0.15$ (triangle) and $\bar{x} = 0.33$ (circle). The dashed line represents the experimental results for pure $^4$He at 0.5 K from reference [15].

Finally we mention that all our results were reproducible, but were always obtained when cooling down the mixture. When the mixture films were heated from the lowest temperature we observed an hysteresis in a temperature range between 0.18 K and 0.28 K.

In conclusion we have presented in this letter third sound velocity measurements on thin mixture films which exhibit a very unusual temperature dependence around 0.2 K. A possible explanation of the behaviour observed is the occurrence of a phase separation in the film. However the mechanism responsible for this stratification is still not clearly established.

## References


[1] TACONIS, K. W. and DE BRUYN OUBOTER, R., *Progress in Low Temperature Physics*, edited by C. J. Gorter (North Holland, Amsterdam) 1964, vol. **IV**, Chap. 2.

[2] KOSTERLITZ, J. M. and THOULESS, D. J., *J. Phys. C* **6** (1973) 1181.

[3] RUDNICK, I., *Phys. Rev. Lett.* **40** (1978) 1454.
BISHOP, D. J. and REPPY, J. D., *Phys. Rev. Lett.* **40** (1978) 1727.
WEBSTER, E., WEBSTER, G. and CHESTER, M., *Phys. Rev. Lett.* **42** (1979) 243.

[4] LAHEURTE, J. P., NOIRAY, J. C., ROMAGNAN, J. P. and DANDACHE, H., *Phys. Rev. B* **22** (1980) 4307.

[5] BERKER, A. N. and NELSON, D. R., *Phys. Rev. B* **19** (1979) 2488.

[6] CARDY, J. L. and SCALAPINO, D. J., *Phys. Rev. B* **19** (1979) 1428.

[7] LAHEURTE, J. P., *Phys. Rev. A* **6** (1972) 2452.

[8] SAAM, W. F., *Phys. Rev. A* **4** (1971) 1278.

[9] MON, K. K. and SAAM, W. F., to be published.

[10] Filtres Millipore V.M.W.P. 02500.

[11] CHESTER, M., LAHEURTE, J. P. and ROMAGNAN, J. P., *Phys. Rev. B* **14** (1976) 2812.

[12] BRUNAUER, S., EMMETT, P. H. and TELLER, E., *J. Am. Chem. Soc.* **60** (1938) 309.

[13] JELAITIS, G., ROTH, J. A. and MAYNARD, J. D., *Phys. Rev. Lett.* **42** (1979) 1285.

[14] A phase separation in clusters as predicted in references [5-6, 9] would reduce $\langle \rho_s \rangle$ without changing the thickness and thus cannot explain the low temperature behaviour of the third sound velocity.

[15] SCHOLTZ, J. H., MCLEAN, E. D. and RUDNICK, I., *Phys. Rev. Lett.* **32** (1974) 147.


Commission paritaire N° 59.024



*Directrice de la Publication :* Jeanne BERGER

*Imprimé en France.* — Imprimerie JOUVE, 17, rue du Louvre, 75001 PARIS
Dépôt légal : 2e trimestre 1981