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LIQUID-GAS PHASE TRANSITION IN ^3He

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Résumé.- Nous étudions le changement de phase liquide-gaz de ^3He par une méthode de champ moyen. Cette dernière donne des prédictions raisonnables pour la pression de vapeur saturante à toute température. Nous discutons également les prédictions de cette méthode concernant le point critique et les variations de densités du liquide et du gaz en fonction de la température le long de la ligne de coexistence.

Abstract.- A mean field approach is used to investigate the liquid-gas phase transition in ^3He . The method provides a reasonable prediction for the vapour pressure in the whole region of temperatures. The predictions for the critical point and the temperature dependence of the liquid and gas densities along the coexistence line are also discussed.

Introduction

The properties of the liquid-gas phase transition in helium systems are significantly affected by quantum effects. This makes the investigation of the problem particularly interesting from a theoretical point of view. One needs in fact a consistent formalism based on quantum mechanics able to describe the liquid and the gas in a wide range of conditions going from the degenerate limit to the classical regime.

Most of the properties of the liquid-gas phase transition have been measured in both ^3He and ^4He [1-2]. The availability in the near future of experimental data [3] on the liquid-gas phase transition in polarized ^3He is expected to open new and stimulating perspectives in the study of the thermodynamic behaviour of quantum liquids.

In this work we use a mean field approach for investigating the thermodynamic properties of the liquid-gas phase transition in ^3He . A similar method has been already employed to investigate the thermodynamic properties of nuclear systems, in particular of phase transitions [4-5].

The formalism

A convenient thermodynamics potential to study

the behaviour of the system is the free energy density

$$f(\rho, T) = h(\rho, T) - Ts(\rho, T) \quad (1)$$

where T is the temperature, ρ the particle density and h and s are the energy and entropy densities per unit volume respectively. It is convenient to write equation (1) in the following form

$$f(\rho, T) = f_{\text{ni}}(\rho, T) + f_v(\rho, T) \quad (2)$$

where $f_{\text{ni}}(\rho, T)$ is the free energy density of a non-interacting Fermi gas [6] and f_v is the contribution due to the interaction. The quantity f_v can be evaluated using an effective interaction in the context of a mean field approach, as for example the Hartree-Fock theory [7]. An especially simple result is obtained using a local zero range interaction. In this case f_v is T independent. In particular, the use of a local Skyrme interaction yields the following expression for f_v [5].

$$f_v = \frac{1}{2} b \rho^2 + \frac{1}{2} c \rho^{2+\sigma} \quad (3)$$

The term b originates from an attractive two body contact force, while the term in c comes from a repulsive density dependent interaction, such a dependence being characterized by the parameter σ .

From equations (2-3) one gets the following equa-

tion of state (EOS) at finite temperature :

$$P(\rho, T) = P_{ni}(\rho, T) + \frac{1}{2}b\rho^2 + \frac{1}{2}c(1 + \sigma)\rho^{2+\sigma} \quad (4)$$

where P_{ni} is the pressure of the non-interacting fermion gas.

The parameters of the force entering equations (3-4) have been fixed in order to reproduce the experimental values of the saturation density ($\rho_0 = 0.0165$ part/ \AA^3), binding energy ($E/N = -2.49$ K) and incompressibility ($K^{-1} = 0.20$ $K\text{\AA}^{-3}$) of liquid ^3He at zero temperature. In this limit, the energy per particle becomes

$$E/N = \frac{\hbar^2}{2m} \frac{3}{5} (3\pi^2)^{2/3} \rho^{2/3} + \frac{1}{2}b\rho + \frac{1}{2}c\rho^{1+\sigma} \quad (5)$$

where the first term in the right hand side of equation (5) represents the kinetic energy of the free Fermi gas. One can also derive the EOS and the incompressibility at $T=0$:

$$P = \frac{\hbar^2}{2m} \frac{2}{5} (3\pi^2)^{2/3} \rho^{5/3} + \frac{1}{2}b\rho^2 + \frac{1}{2}c(1 + \sigma)\rho^{2+\sigma} \quad (6)$$

and

$$K^{-1} = \frac{\hbar^2}{2m} \frac{2}{3} (3\pi^2)^{2/3} \rho^{5/3} + b\rho^2 + \frac{1}{2}c(1 + \sigma)(2 + \sigma)\rho^{2+\sigma}. \quad (7)$$

Fitting the experimental data yields the following values for the parameters : $b = -823.35$ $K \text{\AA}^3$, $c = 871.4874 \times 10^4$ $K (\text{\AA}^3)^{1+\sigma}$ and $\sigma = 2.66$. Equations (6-7) then provide a very accurate description of the EOS of liquid ^3He up to the solidification pressure. Notice that expressions (5-7) differ from the ones in reference [8] where the effective interaction, introduced to investigate the equation of state at zero temperature, includes also a non local term responsible for a density dependent effective mass. The use here of a local effective interaction is justified by the fact that in the range of relatively high temperatures explored in the present work, the effective mass is expected to be closer to the bare value than to the one exhibited in the low temperature regime ($T \leq 0.2$ K).

In the limit of high temperature where the Fermi gas is completely non-degenerate and thus equivalent to a classical gas, the EOS (4) gets the simplified form :

$$P(\rho, T) = \rho T + \frac{1}{2}b\rho^2 + \frac{1}{2}c(1 + \sigma)\rho^{2+\sigma} \quad (8)$$

Results and discussions

We have investigated the EOS of ^3He using equation (4). Typical isotherms are plotted in figure 1 for different values of T . The present approach predicts instability regions ($\partial P/\partial \rho < 0$), revealing the existence of a liquid-gas phase transition. One can see from this

figure that our calculated equation of state for the liquid (solid lines) is in fair agreement with the experimental values [1] which are represented by dots.

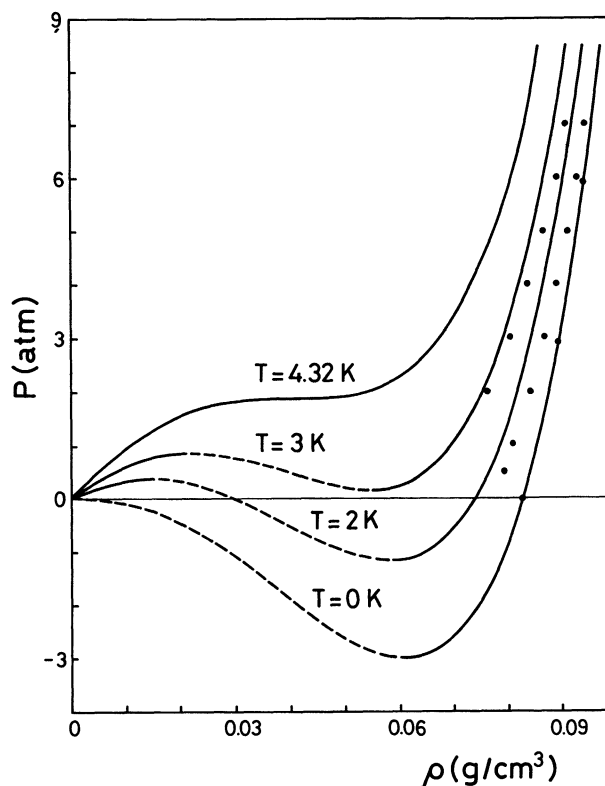


Fig.1.- ^3He isotherms for different temperatures. The dashed lines correspond to the negative compressibility regions. Dots are experimental values from [1].

Our model is able to reproduce the critical point characteristics only in a semiquantitative way. The critical point occurs at $T_c = 4.32$ K, $P_c = 1425$ mm Hg and $\rho_c = 0.0404$ g/cm^3 . These values should be compared with the experimental data [2,9] $T_c = 3.32$ K, $P_c = 873$ mm Hg and $\rho_c = 0.0418$ g/cm^3 .

It is also interesting to comment on the adimensional quantity $P_c/(\rho_c T_c)$. The experimental value is 0.30 while the present calculation gives 0.39. On the other hand if one had used expression (8) for the EOS, one would have obtained the result [5] $P_c/(\rho_c T_c) = (\sigma+1)/2(\sigma+2)$, yielding 0.39. The fact that the classical limit yields the same result as the quantum calculation seems to indicate that the combination $P_c/(\rho_c T_c)$ is very little affected by quantum effects. This fact has been already pointed out in the investigation of critical phenomena for different noble gases using the principle of corresponding states [6]. Notice that our value for $P_c/(\rho_c T_c)$ is very close to the one given by the Van der Waals equation ($P_c/(\rho_c T_c) = 3/8$).

The instability of the homogeneous phase for $T <$

T_c reveals the occurrence of a phase separation. The coexistence line can be obtained by imposing the equilibrium conditions between the liquid (l) and vapour (v) phases, i.e. $p_l = p_v$ and $\mu_l = \mu_v$, where μ is the chemical potential.

Figure 2 reports the vapour pressure as a function of $1/T$. The agreement between our calculation and the experimental data [2] is satisfactory. Notice that the present model correctly reproduces the asymptotic behaviour of the vapour pressure when T goes to zero

$$P_v(T) = \frac{1}{4\pi^{3/2}} \left(\frac{2m}{\hbar^2} \right)^{3/2} T^{5/2} e^{(E/N)/T}. \quad (9)$$

In fact, in the $T \rightarrow 0$ limit the gas in equilibrium with the liquid behaves as a free classical gas and the chemical potential of the liquid approaches the binding energy per particle E/N at saturation, which is one of the quantities used to construct the effective interaction.

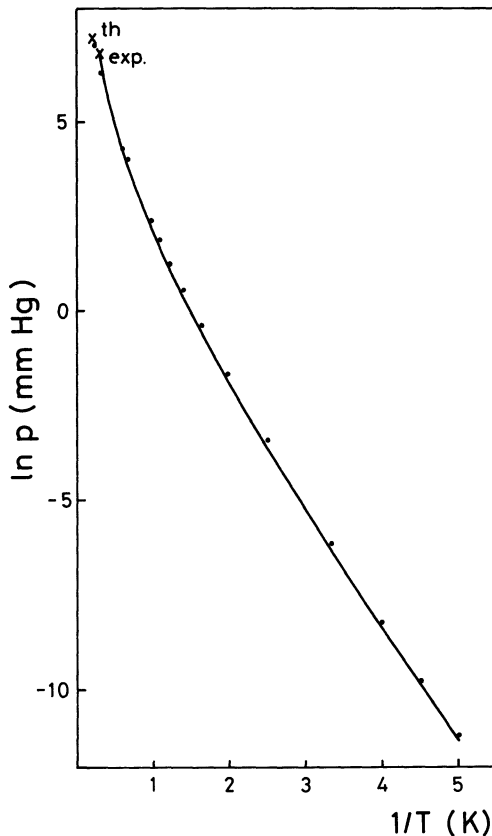


Fig.2.- Vapour pressure *versus* $1/T$. The continuous line corresponds to a fit to the experimental results [2]. The points are our calculated values. The corresponding experimental (Exp) and the theoretical (Th) critical points are indicated by crosses.

In figure 3 we report the value of the liquid (ρ_l) and vapour (ρ_v) densities computed along the coexis-

tence curve. One can see that the vapour and liquid densities are predicted to be almost symmetric with respect to ρ_c in agreement with the experimental findings [9-11].

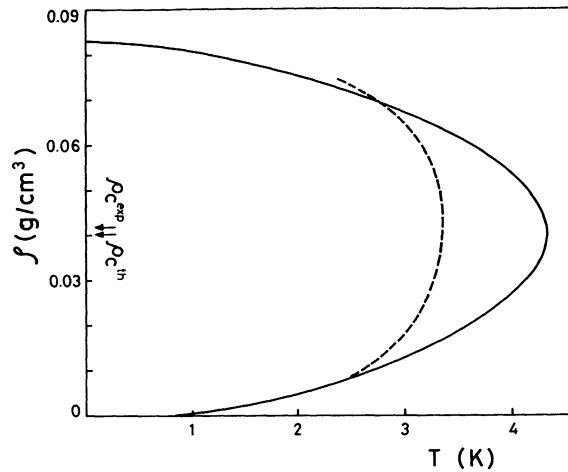


Fig.3.- ³He liquid-vapour coexistence line. The arrows indicate the critical densities. The dashed line corresponds to the experimental values of reference [11].

The critical exponents β and δ characterizing the behaviour of the thermodynamic variables near the critical point have been also evaluated. We find the characteristic values of a mean field theory [12] $\beta = 0.5$ and $\delta = 3$. The same is true in the case that we start from the classical limit for the EOS, equation (8). These critical exponents are significantly different from the experimental ones [9] : $\beta = 0.32$ and $\delta = 4.2$.

It is also interesting to investigate the liquid density along the coexistence line in the low temperature regime. While the theoretical curve is monotonously decreasing with T , the experimental values [13] exhibits a smooth maximum at $T \sim 0.5$ K. The occurrence of the maximum implies a negative value of the expansion coefficient for lower values of T [14]. The fact that our model does not show such an effect is due to the absence of non local potential terms in the effective interaction. Indeed, the main origin of the negative value of the expansion coefficient at very low T is the density dependence of the effective mass [15]. Notice that such an effect, being important only at very low temperatures ($T \leq 0.5$ K), has small influence on the analysis of the vapour pressure P_v (and consequently on the vaporization heat L , see Eq. (10) below). In fact, at low temperatures P_v is essentially determined by the value of the energy per particles (see Eq. (9)). For example at $T = 0.5$ K, the experimental value of P_v differs from the value predicted by equation (9) only by 15 % .

We finally compare the theoretical prediction for the latent vaporization heat

$$L = T \left(\frac{1}{\rho_v} - \frac{1}{\rho_L} \right) \frac{dP}{dT} \Big|_{\text{sat}} \quad (10)$$

with the experimental values [2] (see Fig.4). This quantity is proportional to dP/dT and hence rather sensitive to the fine details of the vapour pressure. The agreement is satisfactory in the low temperature region.

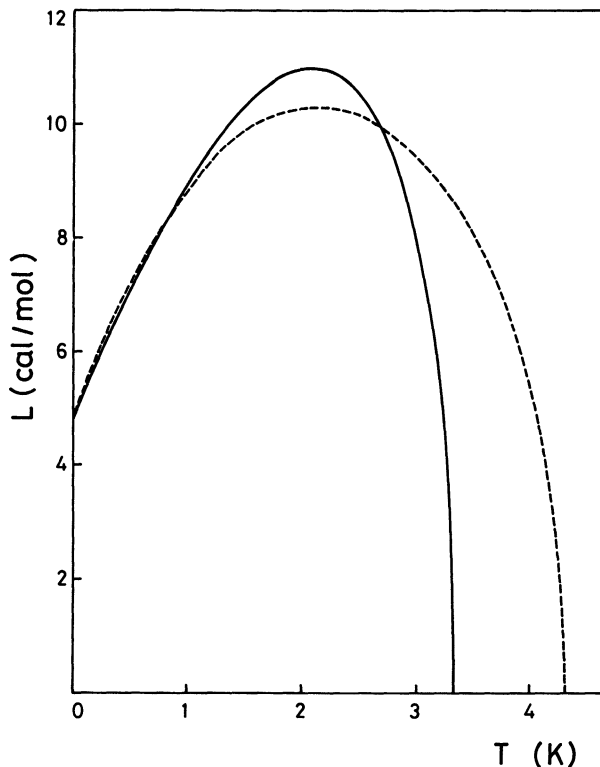


Fig.4.- Latent heat as a function of T . The continuous and dashed lines correspond respectively to the experimental and theoretical values.

In summary, we have provided a consistent quantum mechanical description of the liquid-gas phase transition in ^3He . The method is based on an extremely simplified treatment of the interaction effects in the framework of a mean field approach. Nevertheless, it provides a good quantitative description of the phase transition, in particular of the vapour pressure in the whole range of temperatures. The present approach has been generalized in reference [16] to investigate the properties of the liquid-gas phase transitions in polarized ^3He .

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