Heat capacity investigation of two dimensional isotopic fluid helium mixtures

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Abstract

The properties of a helium fluid mixture, adsorbed as the second atomic layer on the surface of graphite plated by a monolayer of ^{4}He, have been investigated in the temperature range 2 - 80 mK. Using a small tracer of ^{3}He, we find support for the theoretically predicted phase diagram for the second layer of ^{4}He ; a self bound liquid, followed by a significant coverage range over which the fluid uniformly covers the surface, before solidification. Adding ^{3}He up to a coverage of 1.0 nm^{-2} to the uniform ^{4}He film, we initially observe an anomalous temperature dependence of the heat capacity, which we attribute to virtually complete phase separation within the layer, giving rise to the formation of two dimensional ^{3}He fluid nanoclusters. We also find evidence for a small solubility of ^{3}He in the ^{4}He liquid, with a density dependence similar to that predicted.

Key words:
Two dimensional helium; helium mixtures; nanoclusters

The phase separation of isotopic liquid helium mixtures in bulk is well understood and technologically important. However in the case of two dimensions fundamental differences are expected [1] , and experiments are needed. We investigate a monolayer mixture on the surface of graphite, plated with a completed solid monolayer of ^{4}He (i.e. a second layer mixture).

How might 2D mixtures differ from bulk? It is widely predicted that a second layer ^{4}He film has a liquid-gas transition, with a liquid density of around 4.5 nm^{-2}. Thus at low temperatures, up to a second layer ^{4}He coverage of 4.5 nm^{-2}, it is expected that the state of the film is a liquid-gas (L+G) coexistence, so at the lowest temperatures the film consists of self-bound 2D puddles of liquid at zero pressure. The most recent theoretical phase diagram [2] predicts a region over which the uniform liquid is stable, in contrast with the reported experimental phase diagram [3]. On the other hand the corresponding ^{3}He film is not expected to be self bound, as confirmed experimentally, i.e. 2D ^{3}He is a gas with finite pressure at $T = 0$, and no liquid-gas transition. Furthermore, previous work, at temperatures above possible phase separation has shown that the L-G transition of a helium mixture is suppressed at sufficiently high ^{3}He concentrations [4].

Thus, at the lowest temperatures, in the case of complete phase separation, the ground state should consist of a coexistence of liquid ^{4}He and gaseous ^{3}He. The liquid ^{4}He will be compressed somewhat, with the densities of the two phases determined by the condition that their pressures are equal.

What is the solubility of ^{3}He in ^{4}He in two dimensions? Recent theoretical work [5] has found a maximum solubility of 2.5% around a ^{4}He density of 5.5 nm^{-2}, with the solubility tending to zero as the ^{4}He density is reduced towards its zero pressure value.

Experimental details are given in a companion paper in these proceedings, which also gives details of our coverage scale. In the first part of the experiment the objective was to clarify the phase diagram of the second ^{4}He layer. A small amount of ^{3}He, coverage 0.1 nm^{-2}, was introduced as a “tracer” and the total ^{4}He

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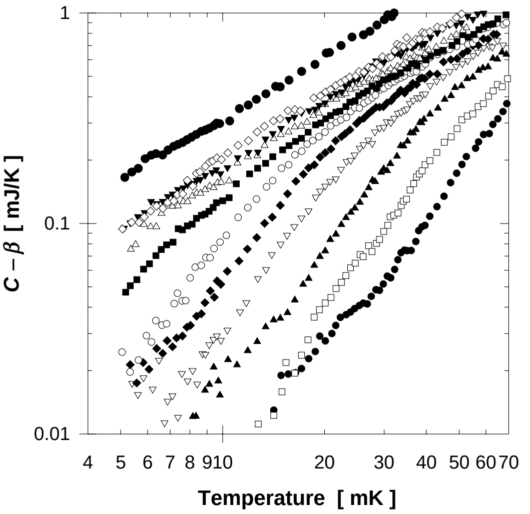

Fig. 1. Heat capacity for increasing ^{3}He coverage; 0.1, 0.15, 0.2, 0.25, 0.3, 0.35, 0.4, 0.5, 0.6, 0.7, 1.0 nm^{-2}

coverage increased from 13.1 nm^{-2} through to solidification of the layer. Note that on our coverage scale the uncompressed first ^{4}He layer has density 11.4 nm^{-2}; adding helium this is eventually compressed to around 12.0 nm^{-2}. Heat capacity isotherms below 30 mK show an approximately linear decrease from 13.1 nm^{-2}, with a break at 16.2 nm^{-2}, which we identify with the end of the L+G coexistence regime. This implies a self-bound ^{4}He density around 4 nm^{-2}, consistent with prediction.

In a second series of experiments the ^{4}He coverage was kept fixed at 16.2 nm^{-2} and the ^{3}He coverage, n_3, increased from 0.1 to 1.0 nm^{-2}. The evolution of the temperature dependence of the heat capacity, after removing a low temperature offset β, is shown in Fig 1. Data at lower ^{3}He coverages is clearly not linear in T at the lowest temperatures. This may be attributed to phase separated ^{3}He nanoclusters, giving rise to finite size effects when $T < \delta \sim E_F/N$. In this picture the cluster size increases with increasing n_3, so that at the highest coverages we recover a linear temperature dependence over the entire temperature window. We use the theoretical $T = 0$ equations of state of 2D ^{4}He and ^{3}He [6], to estimate the densities and relative areas of the two components.

Subsequently the ^{4}He coverage was increased from 16.2 nm^{-2}, until the layer began to solidify. We fit the heat capacity, over the path in coverage followed (first increasing $n_3 > 0.5$ nm^{-2}, then n_4), to the form $c = \beta + \gamma T$, and find that β shows an interesting coverage dependence. This may arise from a small concentration, x_3, of ^{3}He dissolved in the ^{4}He. Since conditions are such that this solution should be non-degenerate, we can infer this concentration from β. In Fig. 2 this is plotted against the density of ^{4}He, estimated as above, where we continue to assume that both isotopes are confined to the monolayer. The result is qualitatively and quantitatively consistent with theoretical prediction [5], shown as the dashed line. In particular the result places an upper bound for the solubility of ^{3}He in ^{4}He for our system of near 2%.

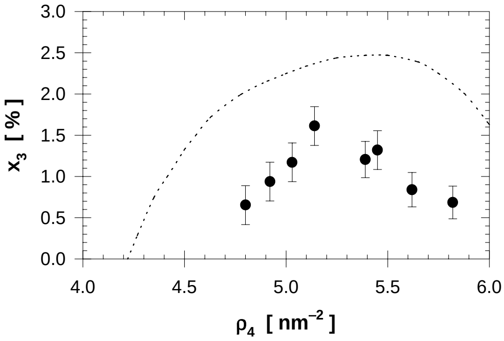

Fig. 2. Solubility of ^{3}He in 2D ^{4}He

Since $\gamma = \pi k_B^2 m^* A_3 / 3\hbar^2$, we calculate the effective mass ratio of ^{3}He within the clusters from the experimental value of γ and a value for A_3, the area occupied by the ^{3}He, inferred from the theoretical equations of state. In this model we find $m^*/m \sim 9$, significantly higher than the result for a second layer pure ^{3}He film at comparable densities. This would suggest that the Fermi liquid properties of 2D ^{3}He nanoclusters, show new effects, possibly arising from a cut-off in the spectrum of spin fluctuations at long wavelengths. An alternative scenario to phase separation within the layer is that for $n_3 > 0.5$ nm^{-2} the ^{3}He occupies a surface state, floating above the ^{4}He film, and hence filling the entire substrate area. In this case we infer an m^*/m increasing from 1.0 to 1.9 as n_3 is increased from 0.5 to 1.0 nm^{-2}. Subsequently increasing the ^{4}He coverage has little effect on m^*/m. Theoretical work to help resolve these two scenarios would be very helpful.

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