Comment on "Nuclear Magnetic Ordering of ^{3}He Clusters in Solid ^{4}He"

Schrenk, König, and Pobell (SKP) [1] have reported several effects relating to magnetic ordering of ^{3}He droplets in solid ^{4}He including higher ordering temperatures, at lower pressures, than for bulk solid ^{3}He and a history-dependent transition temperature. Well below the phase-separation temperature, the separated droplets are known to be almost pure ^{3}He (see [1], and references therein). SKP compare properties of the droplets with those of bulk ^{3}He, leading to apparent inconsistencies. However, as we explain, the effects seen by SKP can be understood on the basis of known differences between the equations-of-state of bulk ^{3}He and of ^{3}He in confined geometries [2–4]. These differences, which include nucleation below bulk melting pressure of solid surface layers in which the density varies with distance from the surface [4], hysteresis between melting and freezing, and incomplete melting of ^{3}He in confined geometries [2,3], result from the interaction between the ^{3}He and the substrate.

For ^{3}He in porous glass [2], the confined-geometry effects are attributed to homogeneous nucleation resulting from the mismatch between the interatomic spacing in the dense surface layer and that of bulk solid. In the droplets studied by SKP, a similar mismatch must occur at the interface with the hcp ^{4}He which has a significantly higher density than that of ^{3}He at the same pressure. As a result of the van der Waals attraction to the ^{4}He surface, the density of the solid in the droplet would be higher at the interface than in the interior [4]. The existence of solid in the droplets at pressures below the bulk melting pressure is thus well understood.

The pressure of the bulk ^{4}He is not the appropriate variable to describe inhomogeneous solid droplets in which magnetic interactions are determined by density. Consequently, there is no reason to expect that magnetic ordering temperature versus pressure of the droplet would lie on the same curve as for bulk ^{3}He, as SKP have plotted in their Fig. 4. Moreover, the error bars of this figure do not allow such a conclusion to be drawn.

The transition of SKP (Fig. 1) is much broader than that of Greywall and Busch [5] and has no latent heat. These are well-known effects of density gradients in the sample. Their pressure drop on freezing (Fig. 2) has a much smaller slope than that of the bulk melting curve, and there is a significant tail on the high-temperature side. Additionally, high-resolution measurements by Haley *et al.* [3] of melting and freezing in droplets show a hysteresis in temperature. All of these are characteristics observed for melting of ^{3}He in confined geometries [2].

It is clear from the behavior of the heat capacity of SKP (Fig. 3) that their observed history dependence cannot be attributed to a higher transition temperature when samples have not been cooled to as low a minimum temperature as those giving a lower transition temperature. Although the peak which they observe does indeed appear at a higher temperature if $T_{\min}$ is higher, this peak cannot be claimed to indicate T_N. If the droplets were cooled through the ordering temperature, then there would be a decrease in entropy (extrapolated to $T = 0$) of $R \ln 2$, where R is the gas constant. There is clearly a considerable missing entropy reduction when samples were not cooled as low as those giving a lower transition temperature. For $p = 34.0$ bars, $T_N = 0.92$ mK, the entropy reduction in cooling to just above T_N is actually greater than that found by Greywall and Busch [5] for bulk ^{3}He, probably indicative of a broadening of the transition in the droplets. The entropy reduction upon cooling to 1 mK for $T_{\min} = 922\ \mu$K is only about 50% of that for $T_{\min} = 783\ \mu$K. This suggests strongly that in these cases all of the solid in the droplets was not cooled through T_N, as would occur for a transition broadened by density gradients.

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