Homogeneous Nucleation in Phase Separation of Solid ^{3}He-^{4}He Mixtures

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Abstract

NMR and pressure have been measured during phase-separation in solid ^{3}He-^{4}He mixtures. Spin-echoes were used to observe bounded diffusion and to estimate the diffusion coefficient, size and nuclei concentration in the ^{3}He-enriched phase. The characteristic phase separation time constant of the mixture was found from pressure measurements. The results argue convincingly for homogeneous nucleation. The surface tension of the nuclei is found independently from NMR and from pressure measurements; the two determinations agree well and yield a surface tension coefficient of 4.9×10^{-6}J m^{-2}.

Key words:
nucleation; solid helium; phase separation

1. Introduction

The kinetics of phase transitions is one of the fundamental problems in condensed matter physics. This has been investigated theoretically and experimentally for many decades but some aspects are still unclear. It has been emphasized frequently in the literature that helium and its isotopic mixtures hold much promise as model systems for studying phase transitions. However, these types of experiments and comparison with theoretical calculation are hampered by some difficulties. One of these is the realization of the conditions for homogeneous nucleation. There was a well-grounded hope for homogeneous nucleation in dilute liquid ^{3}He-^{4}He mixtures [1]. Numerous experimental attempts however failed to yield unambiguous results; rather, they detected heterogeneous nucleation which might be connected with vortex formation. In the case of solid helium, this problem should be resolved through the creation of high-quality impurity-free samples. The quantum character of the diffusion processes in helium ensures fairly high diffusion coefficients, permitting the peformance of experiments within reasonable times.

2. Experimental details

We report measurements on a 1% solution of ^{3}He in ^{4}He, in crystals grown at a pressure of 36 bar. Specimens of crystalographic quality were grown at constant pressure as described in [2]and [3]. This system separates into droplets of fairly pure solid ^{3}He, starting at a temperature of approximately 106 mK.We have performed simultaneous measurements of NMR and pressure during the phase separation process.

Observation of the pressure change following a cooling step allows study of the kinetics of the process by which the new phase droplets form; in particular we can obtain the growth rate of the droplets.

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By contrast, NMR is an equilibrium (really a quasi-equilibrium) measurement. Standard relaxation time data exhibit a double-exponential decay from which we can determine the number of ^{3}He atoms in the background matrix and the number in the growing droplets[2]. However spin-echo measurements in a magnetic field gradient, conventionally used to study spin diffusion, in this system also permit the determination of the size of the droplets[3]. Knowing the size of the droplets and the number of atoms in the droplet phase then allows determination of the number of droplets present - corresponding to the number of nucleation sites produced during the nucleation stage.

Thus NMR permits determination of the nuclei concentration, while pressure measurements permit determination of the droplet growth rate. Within the framework of the homogeneous nucleation model these different quantities are both related to the surface tension coefficient at the droplet boundary. And within this framework both give, independently, the same surface tension value.

3. Theory

Homogeneous nucleation in a uniform supersaturated mixture proceeds through the formation of clusters of the new phase at random sites. If the number of particles n in a cluster is smaller than a certain critical value $n_{\rm c}$ determined by the competition between the bulk and surface contributions to the thermodynamic potential, the cluster is unstable and it vanishes. When $n > n_{\rm c}$, however, the cluster grows. For a spherical cluster in a dilute binary mixture, $n_{\rm c}$ is given by

$$n_{\rm c} = \left(\frac{\beta}{\ln\left(c_0/c_{\rm f}\right)} \right)^3$$

where c_0 is the initial ^{3}He concentration of the mixture, $c_{\rm f}$ is the equilibrium ^{3}He concentration of the matrix at the cluster boundary at the temperature $T_{\rm f}$ - after the temperature step and

$$\beta = \frac{8\pi}{3} \frac{\sigma a^2}{k_{\rm B} T_{\rm f}}.$$

Here σ is the surface tension at the boundary while a is the atomic distance.

The nucleation rate is a very rapid function of the degree of supersaturation. As a consequence the formation of new nuclei occurs only in the early part of the phase separation process. This results in a maximal cluster concentration (per site) given by [4]

$$N_{\rm m} = \sqrt{2} c_0^{7/4} \left(\frac{3}{2\pi\beta} \right)^{3/8} \exp\left(-\frac{3\beta^3}{8 \ln^2\left(c_0/c_{\rm f}\right)} \right).$$

This quantity may be compared with the cluster concentration as measured by NMR. Typically, $N_{\rm m}$ is deduced to be $\sim 8.5 \times 10^{-15}$.

Subsequent growth of new-phase droplets occurs through diffusion of ^{3}He atoms, through the background matrix, to the nucleation sites. The characteristic time for this growth process is thus

$$\tau_D = \frac{a^2}{3D} c_0^{-1/3} N_{\rm m}^{-2/3}$$

where D is the diffusion coefficient of the ^{3}He in the matrix[5]. This quantity τ_D may be compared with the droplet growth rate as determined from pressure measurements. Typically τ_D is found to be $\sim$5000 s.

All quantities in the expressions for $N_{\rm m}$ and τ_D are known except for the surface tension σ. This means that σ may be determined independently from the NMR measurements and from the pressure measurements. Further details of the determination of σ are given in [6]. The final result is that the surface tension of the droplets is found to be 4.9×10^{-13} J m^{-2} in both cases.

4. Conclusion

Phase separation has been observed in dilute solid solutions of ^{3}He in ^{4}He using both NMR and pressure measurements. Within the framework of the homogeneous nucleation model the surface tension of the phase-separated droplets was found to be the same for the two types of measurements. This gives convincing support for the interpretation that the phase separation process in this system occurs through the mechanism of homogeneous nucleation.

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