Strong Supercooling and Stimulation of the A -B Transition in Superfluid ³He

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We report the first studies of the $A-B$ transition in superfluid 3 He confined by microscopically smooth surfaces. The transition exhibited dramatic supercooling, down to $0.15T_c$. The lifetime τ of the metastable A phase has a temperature dependence consistent with Leggett's "baked Alaska" model based on nucleation by ionizing radiation. Radiation sources near the cryostat strongly reduce the lifetime, but do not alter its temperature dependence, and τ increases with increasing magnetic field in accord with Leggett's model.

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The A and B phases of superfluid 3 He correspond to two different $l = 1$ BCS states with different symmetries [I]. Upon cooling in low magnetic fields, the free energies of both superfluids drop below that of the normal Fermi liquid at a temperature T_c , where $T_c = 2.49$ mK at melting pressure [2]. Between T_c and a lower temperature T_{AB} , the A phase has a lower free energy than the B phase, and is stable. Below T_{AB} the B-phase free energy is lower, making that phase stable instead. The $A - B$ transition is supercooled by a variable amount below T_{AB} , but has never been observed below $0.5T_c$.

In 1977 Osheroff and Cross [3] measured the surface tension σ_{AB} between the A and B phases, and the difference in the bulk free energies, ΔF . Based on their data, the critical radius, $R_c = 2\sigma/|\Delta F|$, for growth of the B phase within the supercooled A phase is typically about 1μ m. For smaller radii, the surface energy would force a B-phase bubble to disappear. This R_c corresponds to an energy barrier for thermal nucleation of about $10^6 k_B T$, making homogeneous nucleation impossible in this system. Since the presence of smooth surfaces stabilizes the A phase, it was difficult to understand how the B phase was ever formed. In 1984, Leggett [4] proposed that the 8 phase nucleates through what he called the "baked Alaska" effect, initiated by cosmic rays passing through the metastable A phase. In his model, secondary electrons created by cosmic-ray muons would heat the superfluid above T_c in a volume of order R_c^3 . Since ³He quasiparticles at T_c have a mean free path comparable to R_c , the heated region would evolve into a shell of normal fluid with the center left relatively free of quasiparticles and thus cold. Leggett suggested that the B phase could form in this cold interior, and, protected by the shell of normal fluid, grow large enough that it would be stable when the shell dissipated. Swift and Buchanan [51 looked for coincidence between incident cosmic rays and 8-phase nucleation, but found no significant correlation. They did observe, however, that the B phase usually nucleated near the same positions in their cell.

Conjecturing that the barrier to nucleation might be depressed by textural effects associated with rough surfaces, we designed a sample cell to limit such surface contributions. We contained the 3 He in 1-mm-inner-diam fused silica tubes with one end melted shut. Scanning electron microscopy studies of such tubes showed their inner surfaces to be smooth to \sim 200 Å, much smaller than R_c . Each tube was flushed free of dust in a class-1000 clean room, and the open end was capped with 0.1- μ m Nuclepore [6] filter paper while in the clean room. As shown in Fig. 1, the open ends of the tubes were passed through holes in a NdFeB permanent magnet. The field inside these holes was ~ 0.6 T, enough [7] to stabilize the A phase to $T=0$ and thus prevent the B phase nucleated in other regions of the cell from propagating into the tubes.

Two tubes (labeled 1 and 2 in Fig. 1) were used to study the nucleation process. Tube 2 was almost twice as long as tube ¹ and had its bottom end inserted through a second permanent magnet. An NMR coil was placed around each of these tubes, and the coils were connected in parallel within a conventional cw NMR spectrometer.

FIG. 1. Sample cell used in these experiments. The diagram on the left has been distorted for labeling. The one on the right shows the proper aspect ratio.

The normal liquid 3 He signals had linewidths of about 200 Hz in a field of 28.2 mT applied normal to the tubes. We rotated the NMR magnet [8] so that field gradients caused by the NdFeB magnets separated the two Larmor frequencies by about 800 Hz, allowing the samples to be observed independently. A third tube, which did not pass through the top magnet, had its bottom 3 cm filled with 3 - μ m-diam Pt powder, used for pulsed NMR thermometry. The tubes were epoxied into a nylon body which was attached to a sintered silver heat exchanger and a copper wire nuclear demagnetization stage. In previous work with this heat exchanger and demagnetization stage, changing the magnetic field on the stage caused negligible heating, and T was almost exactly proportional to the applied field [8].

The $A - B$ transition was observed by the accompanying dramatic change in the cw NMR absorption signal. The A-phase NMR frequency is shifted above the Larmor value by a temperature-dependent amount (this shift was used to indicate thermal equilibrium within the tubes). The B-phase NMR signal, however, is smeared by textural effects in our geometry, and virtually disappears. We repeatedly swept over a frequency range several kHz above the Larmor frequency that included the A-phase signals from both tubes. The disappearance of either signal would indicate that the $A-B$ transition had taken place in the corresponding tube.

The absence of a significant heat leak into the tubes was indicated by the Pt temperature being nearly linear in the demagnetization field between 3 and 0.85 mK. The deviation from linearity corresponded to a 2-pW heat leak into the Pt powder, which would produce a negligible thermal gradient of 5 μ K across the ³He in the tubes. The frequency shifts in the two sample tubes were identical to within 30 Hz at all temperatures below T_c , and their temperature dependence closely matched previous measurements [9] between T_c and 1.5 mK. Thus we assume that, in equilibrium, all three tubes were at the same temperature indicated by the Pt thermometer, corrected slightly for the heat leak into the Pt. All of the measurements reported here were taken at a pressure of 34.1 \pm 0.2 bars.

We were able to cool the A phase in the sample tubes to much lower temperatures than had previously been attained in weak magnetic fields. Our samples could be held in the metastable A phase for many hours near 0.4 T_c (1 mK), and we could supercool the A phase to temperatures as low as $0.15T_c$ (0.39 mK) for nearly 30 min before the B phase nucleated. In previous work, the B phase usually nucleated at about $0.6T_c$ (1.5 mK) and always above $0.5T_c$ (1.25 mK) [10,11]. In contrast to previous observations [10], where nucleation only occurred while the sample was cooling, we also observed nucleation while the samples were in thermal equilibrium. We did not find nucleation to be affected by the cooling rate, although very rapid cooling allowed the A phase to

be supercooled much farther before nucleation occurred.

To simulate cosmic-ray muons, we obtained a 1.9-mCi Co source (emitting gamma rays at 1.17 and 1.33 MeV). Accounting for attenuation by the materials surrounding the sample tubes, we expected about 5000 gamma rays per second to be incident on tube 1. Through photoionization and Compton scattering within the 3 He and the fused silica, these gamma rays created highenergy electrons, some of which would travel through the ³He, simulating the secondary electrons created by cosmic-ray muons. We measured the lifetime τ of the metastable A phase by letting the samples come into thermal equilibrium and then placing the ${}^{60}Co$ source near the cryostat until the B phase had nucleated in both tubes. The presence of the source had a dramatic effect on the $A - B$ transitions, reducing τ by more than 3 orders of magnitude. At the lower temperatures, we used lead to attenuate the source, since nucleation would otherwise occur more quickly than we could accurately measure. The resulting data were then normalized to constant flux by assuming that the lifetime was inversely proportional to the incident gamma-ray flux, an assumption which we tested at 1.18 mK. The temperature of each sample was corrected for heating effects due to the gamma radiation, typically 30 μ K (as determined from the NMR shifts [12])when the irradiation time exceeded 10 min.

At a given temperature (T) , the data could be fitted by an exponential decay curve $N(t) = N_0 \exp(-t/\tau)$ where N_0 is the total number of samples and N is the number of samples still in the A phase after a time t . This indicates that nucleation results from a single stochastic process. Accordingly, the individual measured lifetimes were averaged to compute $\tau(T)$, with an error of $\pm \tau s N_0^{1/2}$. The lifetimes in tube 2 were shorter than those in tube ¹ by about a factor of 2. Thus, to improve our statistics, we doubled the lifetimes from tube 2 and combined the data

FIG. 2. Lifetime of the A phase in three different magnetic fields in the presence of the ${}^{60}Co$ source. The curves are fits to the data as explained in the text.

from both tubes. This factor of 2 is almost exactly the ratio of the tubes' volumes and surface areas, indicating that nucleation was probably not influenced by thermal history, e.g., failing to warm the longer tube completely into the normal phase (which was almost done for the shorter tube). We studied gamma nucleation in magnetic fields of 14, 28.2, and 100 mT, as is shown in Fig. 2.

To interpret the temperature and field dependence, we assume, as did Leggett and Yip [13], that τ depends exponentially upon $R_c = 2\delta/\Delta F$ to some power, and only upon R_c . We take the temperature dependence of σ as $(T-T/T_c)^{1/2}$ from the data of Osheroff and Cross [3]. We further assume $\Delta F(H=0)$ is proportional to 1 $-T/T_{AB}$, to be consistent with the quadratic depression of T_{AB} with magnetic field [14]. The field dependence of ΔF comes mainly from the term $-\frac{1}{2}(\chi_A - \chi_B)H^2$, which is accounted for by the term $-(H_{\text{eff}}/H_c)^2$ in the final expression: \overline{a}

$$
R_c = R_0 \frac{(1 - T/T_c)^{1/2}}{1 - T/T_{AB} - (H_{\text{eff}}/H_c)^2}.
$$
 (1)

Here $H_{\text{eff}} = (\chi_A/\chi_B)^{1/2} H$, as we assume that nucleation occurs at constant magnetization [15], and $H_c = 0.63$ T to be consistent with the measured dependence of T_{AB} on magnetic field [14]. This is not the $T=0$ critical field, due to the T^4 dependence of ΔF at low T. We find $R_0 = 0.45$ µm from the critical fields determined by Gould [7]. This expression for R_c is probably good to 30%. The 28.2-mT data are then fitted well by the expression

$$
\tau = 0.000211 \exp[5.25(R_c/R_0)^{3/2}] \tag{2}
$$

as is shown by the solid line in Fig. 2. Although the exponent of 3/2 is far from Leggett and Yip's estimate of 5, the exponential form of the data is consistent with the "baked Alaska" model.

A good test of the above analysis is how well we can predict the values of τ in other magnetic fields. The dashed line in Fig. 2 is determined using (1) and (2) with $H_{\text{eff}} = 0.173$ T, consistent with a field of 100 mT and the relative susceptibilities. Using no free parameters, this fits the data very well, supporting our model and the conjecture that nucleation occurs at constant magnetization. Similar analysis for a magnetic field of 14 mT, however, predicts very little deviation from the 28.2-mT fit in our temperature range. This does not agree with our 14-mT results at the higher temperatures, and we suspect that in low fields a parallel nucleation mechanism may become important.

We also measured τ as a function of temperature in the presence of a moderated PuBe neutron source, estimated to provide about 2.5 ± 1 thermal neutrons per second incident on tube 1, and a negligible flux of 2.2- Mev gamma rays. Thermal neutrons have a large cross section for capture by 3 He through the reaction

$$
{}^{3}\text{He} + n \rightarrow {}^{3}\text{H} + \text{H} + 0.764 \text{ MeV}
$$

with an absorption length of 100 μ m. The 0.764-MeV kinetic energy is dissipated over a path length of about 40 μ m. Finally, we measured τ in the presence of background radiation alone, as discussed below. Figure 3 shows values of τ as a function of temperature for the three radiation sources in a field of 28.2 mT. All three sets of data are fitted by the same functional form given by (2), varying only the leading constant (values of 0.00148 and 0.347 are used for the neutron and background curves shown in Fig. 3). The neutron result is surprising since the neutrons should have caused much more intense local heating than the electrons, and one might expect a different temperature dependence for the nucleation probability.

Monte Carlo calculations [16] were performed to compare the nature of heating caused by the gamma rays and muons, since the heat distribution from individual electrons produced by the two sources should be similar. Based on the fluxes incident on the cell, we calculated the ratio of the rates at which enough energy (> 500 eV) was dumped into a sufficiently small volume (-10^{-19}) $m³$) to heat that volume well above T_c . Below about 1500 eV, this ratio is fairly independent of the cutoff energy. These "baked Alaska" events should be equivalent to opportunities for a "baked Alaska" nucleation of the B phase. From the gamma flux we found 170 such events per second, while for the muon flux we expected 0.017 events per second. This suggests that the ratio of the background to gamma induced nucleation rates should be 10000, rather than the observed ratio of 1600. We suspect that the difference is from additional sources of background radiation, possibly tritium impurities [17] which in even the cleanest 3 He could easily account for the difference.

The probability of nucleation per "baked Alaska" event is always low, so that even at our lowest temperatures several hundred events took place per nucleation.

FIG. 3. Lifetime of the A phase in the presence of different radiation sources at $H = 28.2$ mT. The curves are fits to the data as explained in the text.

This number is small enough, however, to discount the possibility that nucleation required the coincidence of a "baked Alaska" event with a textural singularity. Assuming that nucleation could occur only if the energy deposition were within 1 μ m of a singularity, the probability of such a coincidence would be $\sim 10^{-11}N$ where N is the number of point singularities in our sample, expected to be less than several thousand. Additionally, based on the equilibrium vorticity in He II [18], coincidence of a "baked Alaska" event with a vortex line is still of negligible importance, even with the much larger probability per vortex.

Given the strong temperature dependence of τ , we doubt that radiation alone was responsible for the nucleation previously observed at higher temperatures. Nucleation in those cases was possibly induced by radiation in concert with the many textural defects caused by rough surfaces. In more open geometries, hydrodynamic heat flow could have created vortex tangles while cooling, with which radiation might have interacted. The Los Alamos group [19], however, saw nucleation in their 3 mm-diam field-isolated sample tubes (with rough walls) at cooling rates of only 25 nK/s, much too slow to create vortex tangles.

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[ll D. M. Lee and R. C. Richardson, in The Physics of Liquid and Solid Helium, edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, 1978), Pt. II; or

in The Superfluid Phases of Helium 3, edited by D. Vollhardt and P. Wolfle (Taylor and Francis, London, 1990).

- [2] D. S. Greywall, Phys. Rev. B 33, 7520 (1986).
- [31 D. D. Osheroff and M. C. Cross, Phys. Rev. Lett. 3\$, 905 (1977).
- [4] A. J. Leggett, Phys. Rev. Lett. 53, 1096 (1984). See also A. J. Leggett, Phys. Rev. Lett. 54, 246 (1985).
- [5] G. W. Swift and D. S. Buchanan, in Proceedings of the Eighteenth International Conference on Low Tempera ture Physics [Jpn J. Appl. Phys. 26-3, 1828 (1987)].
- [6] Nuclepore Corporation, 7035 Commerce Circle, Pleasanton, CA 94566.
- [7] C. M. Gould (private communication).
- [8] Y. P. Feng, P. Schiffer, and D. D. Osheroff, Phys. Rev. Lett. 67, 691 (1991); Y. P. Feng, Ph.D. thesis, Stanford University, 1991 (unpublished).
- [91 D. D. Osheroff and W. F. Brinkman, Phys. Rev. Lett. 32, 584 (1974); D. D. Osheroff, Ph. D. thesis, Cornell University, 1973 (unpublished).
- [10]P. J. Hakonen, M. Krusius, M. M. Salomaa, and J. T. Simola, Phys. Rev. Lett. 54, 245 (1985).
- [11]Hiroshi Fukuyama, Hidehiko Ishimoto, Tetsurou Tazaki, and Shinji Ogawa, Phys. Rev. B 36, 8921 (1987).
- [12] The frequency shift as a function of temperature was calibrated at low temperatures while the samples were in equilibrium. P. Schiffer, M. T. O'Keefe, Hiroshi Fukuyama, and D. D. Osheroff (to be published).
- [13] A. J. Leggett and S. K. Yip, in Superfluid ${}^{3}He$, edited by L. P. Pitaevskii and W. P. Halperin (North-Holland, Amsterdam, 1989).
- [14] H. R. Scholz, Ph.D. thesis, The Ohio State University, 1981 (unpublished).
- [151 A. J. Leggett (private communication).
- [16] Simulations were based on the EGS4 code system. See W. R. Nelson, Hideo Hirayama, and D. W. O. Rodgers, The EGS4 Code System, Stanford Linear Accelerator Center Report No. 265, Stanford, CA, 1985 (unpublished).
- [17] C. Buchal, M. Kubota, R. M. Mueller, and F. Pobell (unpublished).
- [18] D. D. Awschalom and K. W. Schwarz, Phys. Rev. Lett. 52, 49 (1984).
- [19] S. T. P. Boyd and G. W. Swift (to be published).

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