# **RMP** Colloquia

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# Nucleation of the *AB* transition in superfluid <sup>3</sup>He: Surface effects and baked Alaska

P. Schiffer\*

AT&T Bell Laboratories, Murray Hill, New Jersey 07974

## D. D. Osheroff

#### Department of Physics, Stanford University, Stanford, California 94305

The first-order phase transition between the A and B phases of superfluid <sup>3</sup>He has remained an outstanding mystery in helium physics for nearly 20 years. The small difference in bulk free energies between the two phases, combined with the relatively large surface energy associated with the AB interface, leads to an anomalously large critical radius for nucleation, of order 1  $\mu$ m, suggesting a lifetime for the supercooled A phase against homogeneous nucleation far beyond the age of the universe. Yet anisotropy of the high-temperature phase minimizes the depairing effects of surfaces, thus making conventional heterogeneous nucleation unlikely. Recent experiments have been reported that lend support to one of the more exotic nucleation mechanisms ever proposed: Leggett's "baked Alaska" model, in which the B phase is nucleated by cosmic rays penetrating the supercooled A phase. The results of these experiments are discussed, along with the prospects for future work.

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# I. INTRODUCTION

While existing theory allows us to understand secondorder phase transitions with remarkable precision (Wilson, 1983), no comparable theory can explain the broad spectrum of first-order phase transitions observed in nature. The simplest theory, which often sets the scale over which a first-order phase transition will take place, is the homogeneous nucleation theory, in which the transition is nucleated by thermal fluctuations alone. These transitions are quite often inhomogeneous, however, resulting from impurities, effects of surfaces, or other extraneous factors. Furthermore, the length scale set by homogeneous nucleation theory is often only a few nanometers, over which experimentalists have little control of their samples. This is not the case for the more exotic systems in nature, superfluid <sup>3</sup>He, which provides the subject matter for this colloquium.

The <sup>3</sup>He superfluid phases (Lee and Richardson, 1978; Vollhardt and Wölfe, 1990) are BCS states, neutral analogs to conventional superconductors, except that their Cooper pairs possess one unit of angular momentum, l = 1, thus being *p*-wave states. This simple fact leads to a quite astounding richness of detail in the <sup>3</sup>He superfluids. For example, the total spin S = 1 of the Cooper pairs makes these superfluids magnetic. Their order parameter is described by a 3 by 3 matrix of complex coefficients, allowing an infinite range of variations in the ordered state. Only two states are known to be stable for bulk samples in low magnetic fields, corresponding to the A and the B phases. These phases, both involving p-wave states, share a common transition temperature,  $T_c$ , at which their free energies drop below that of the normal Fermi liquid as the temperature is lowered. As shown in the three-dimensional (3D) phase diagram in Fig. 1, the A phase has the lower free energy at temperatures near  $T_c$  and at pressures between about 22 and 34 bars (the melting pressure) in zero magnetic field, being thus the stable phase there. The B phase is stable at

<sup>\*</sup>Address after August 1995: Dept. of Physics, University of Notre Dame, Notre Dame, IN 46556.

lower temperatures at all pressures in zero magnetic field, and it is the only stable phase below 22 bars unless a magnetic field is applied.

Because the A and B phases have very different symmetries, the transition from A to B must be first order. Quite general principles indicate that this transition should never occur; rather, the supercooled A phase lifetime should exceed the age of the universe by many orders of magnitude. Yet the B phase is known to exist, enabling experimentalists to study the properties of not just one but two separate superfluid phases in the temperature region over which the A phase can be supercooled. This circumstance played an important role in the microscopic identification of the two phases in the mid-1970's.

## II. THERMODYNAMICS OF THE A AND B PHASES

The microscopic natures of the A and B phases appear to be well described by two *p*-wave BCS states studied theoretically in the early 1960s following the development of the BCS theory of superconductivity. Physicists looked for other degenerate Fermi fluids that might form BCS condensates, with <sup>3</sup>He being a prime candidate. In 1961 Anderson and Morel (1961) considered a rather peculiar, highly anisotropic *p*-wave state with the angular momenta of all Cooper pairs pointing in a single direction locally in space. As a result, the BCS energy gap for this state vanishes along the direction of the pair angular momenta, *l*. The authors termed this state an "orbital ferromagnet." Later, Brinkman and Anderson (1973) proposed this state as the *A* phase, making it known as the ABM (Anderson-Brinkman-Morel) state. The Coop-



FIG. 1. Schematic of the equilibrium phase diagram of superfluid <sup>3</sup>He (proportions are not scaled accurately). The top plane is the solid-liquid phase boundary. The *B* phase is preferred at lower temperatures than the *A* phase, but completely excluded at high magnetic fields (Greywall, 1986; Hahn, 1993).

er pairs in this state have both their spins pointing either up or down relative to the applied magnetic field, and thus this phase possesses almost the same magnetic susceptibility as the Pauli susceptibility of a normal Fermi liquid (Hensley *et al.*, 1993).

The B phase is believed to be in a state first studied by Balian and Werthamer (1963), who hoped to explain strong-coupling effects in lead. The authors showed this "BW" (Balian-Werthamer) state to possess in the weakcoupling BCS limit the lowest free energy of all p-wave BCS condensates. The BW state has an isotropic energy gap, with equal numbers of Cooper pairs with up-up and down-down spins in the symmetric combination updown+down-up. This last component reduces the BW state susceptibility below that of the Fermi liquid as the number of Cooper pairs increases with decreasing temperature. The nuclear magnetic susceptibility at T=0might therefore be expected to be two-thirds that of the Fermi liquid, but the actual value is closer to one-third because of strong ferromagnetic spin fluctuations in the liquid (Leggett, 1965).

Our knowledge of the susceptibilities of the two superfluid phases serves to determine the difference in their free energies as a function of temperature. By definition,  $F_A - F_B$  vanishes in zero magnetic field at  $T_{AB}(H=0)$ , where F is the free energy of each phase and  $T_{AB}(H)$  is the field-dependent equilibrium transition temperature between the two phases. Applying a magnetic field, H, lowers the free energy of each phase by  $-\chi H^2/2$ , where  $\chi$  indicates the respective magnetic susceptibility. The excess of  $\chi_A$  over  $\chi_B$  lowers the free energy of the A phase below that of the B phase, thus stabilizing the A phase to a temperature  $T_{AB}(H) < T_{AB}(0)$ . At  $T_{AB}(H)$  the free energies of both phases are equal, therefore at this temperature in zero field the free energies of the two phases differ by exactly  $-\Delta \chi H^2/2$ , where  $\Delta \chi$  is simply the difference in the susceptibilities of the two phases at temperature T. The difference between the free energies of the two phases at any field is thus determined from our knowledge of their susceptibilities and of the phase line  $T_{AB}(H)$ . One finds from such a calculation that, near the melting pressure, the free-energy difference is almost linear in  $T - T_{AB}(H)$  down to  $0.5T_c$ , extrapolating to a value of about 1.3 ergs/cm<sup>3</sup> at H=0and T = 0.

The surface energy,  $\sigma_{AB}$ , at the interface between the A and B phases was measured by Osheroff and Cross (1977), who used a weak thermal gradient to stabilize the phase boundary in a vertical tube divided into an upper and a lower portion by a high-porosity screen with holes of uniform diameter. The authors slowly cooled the entire tube, with the lower portion colder, causing the AB interface to rise until becoming pinned at the screen. Further cooling would generate a pressure gradient across the interface, causing it to bow upward. When the radius of curvature equaled the radius of the holes in the screen, the interface would pop through the screen, with the B phase filling the upper region of the tube. The

maximum pressure difference supported by the interface was determined from the measured temperature interval over which the interface remained pinned at the screen. This pressure difference is easily found to be

$$\Delta P = \frac{2\sigma_{AB}}{R} ,$$

where R is the radius of the screen holes. The values of  $\sigma_{AB}$  measured by Osheroff and Cross for two different hole radii, shown in Fig. 2, are roughly a factor of  $10^4$ less than the <sup>3</sup>He liquid-to-vapor surface energy, which is itself three orders of magnitude smaller than typical surface energies between liquid metals and their vapors at their melting points. The solid line in the figure represents an estimate of the surface energy based on the assumption of proportionality to the free-energy difference between the superfluid and normal states over the volume determined by the surface area times the coherence length,  $\xi$ , which is correct in the Ginzburg-Landau regime. By choosing an appropriate path between the two complex order parameters, Cross was able to estimate the proportionality constant from a variational calculation. His value, 1.09, was somewhat larger than the experimental value of 0.7; however, Kaul and Kleinert (1980), and, more recently, Schopohl (1987), using different paths, were able to calculate lower energies, which are close to the experimental value.

## III. THE AB NUCLEATION PROBLEM

#### A. Homogeneous nucleation theory

In the simplest model for nucleation of the *B* phase, known as the homogeneous nucleation theory, the phase transition nucleates by a thermal fluctuation creating a microscopic droplet of *B* phase within the supercooled *A* phase. The droplet's surface energy,  $\sigma_{AB}$ , will cause it to shrink to zero unless its radius exceeds a critical value,



FIG. 2. Surface energy at the AB interface as a function of temperature; the inset shows raw data taken in the experiment (Osheroff and Cross, 1977).

 $R_c$ . To see this, consider the difference in the free energy of the system caused by the droplet's presence as a function of its radius (R):

$$\Delta F = 4\pi R^2 \sigma_{AB} + (4/3)\pi R^3 (F_B - F_A) \; .$$

The first term on the right is always positive, while the second term is negative at  $T < T_{AB}$ . For small R, the first term will always dominate due to its weaker radial dependence, as shown in Fig. 3. The critical radius, where  $\Delta F$  is greatest, is  $R_c = 2\sigma_{AB}/(F_A - F_B)$ , and the free-energy barrier to nucleation, represented by the maximum value of  $\Delta F$ , equals  $\Delta = \frac{2}{3}\pi R_c^3 (F_A - F_B)$ . Plugging in numbers for <sup>3</sup>He at melting pressure and at  $T=0.7T_c$ , as measured by Osheroff and Cross, one finds  $R_c = 1.45 \ \mu$ m, and  $\Delta = 8.2 \times 10^{-20}$  J. This critical radius is actually quite large, lying at a scale over which the experimentalist has both detailed knowledge and good control. By contrast, the critical radius for the nucleation of ice from supercooled water is about 4 nm, a much more difficult length scale to control. The critical volume for AB nucleation contains  $10^{10}$  atoms, as opposed to less than 10<sup>4</sup> molecules for ice.

The homogeneous nucleation theory predicts a lifetime of the supercooled phase against nucleation  $\tau = (1/f_0)e^{\Delta/k_BT}$ . The attempt frequency  $f_0$  is difficult to estimate, but we can safely assume it to be lower than the ratio of the number of atoms in the sample (typically less than a mole) to the time spent by light in crossing the nucleus of the <sup>3</sup>He atom, yielding  $f_0 < 10^{48}$  Hz. This value may seem overly conservative, but  $e^{\Delta/kT} = 10^{1470\,000}$ . Thus, regardless of the  $f_0$  estimate, the lifetime of the metastable supercooled A phase seems to exceed the lifetime of the universe by a comfortable factor. Moreover, such a large exponent makes it inconceivable that errors in measurement could explain away the difference. Even if  $\sigma_{AB}$  were smaller by an order of magnitude and  $F_A - F_B$  larger by an order of magnitude, the conclusion would not change. The experimental fact remains, however, that the AB transition does take place whenever the superfluid is cooled sufficiently below  $T_{AB}$ , although the amount of supercooling and the lifetime of the metastable A phase vary between experiments and even between runs within the same apparatus.



FIG. 3. Schematic of the total energy of a bubble of *B* phase in supercooled *A* phase as a function of the radius of the bubble. The energy increases with increasing bubble size for  $R < R_c$ , due to the surface tension, and then decreases for  $R > R_c$ .

## B. Heterogeneous nucleation theory

Given the above analysis, the AB transition appears obviously to result from heterogeneous nucleation. Superfluid <sup>3</sup>He is, however, a very unusual system. There simply cannot be any dirt or dust suspended in a sample of <sup>3</sup>He that has existed near absolute zero inside a metal cryostat for several months, particularly with a particle size as large as the critical volume. The only thing which dissolves in <sup>3</sup>He is <sup>4</sup>He, which phase separates at low temperatures with an activation energy of about  $0.6k_{R}$ per atom. Thus at the temperatures of interest, we expect less than one atom of <sup>4</sup>He to be dissolved in a mole of <sup>3</sup>He sample. One might argue that the nucleation occurs on surfaces, necessary to cool and enclose the sample. Several experiments have shown, however, that the AB transition temperature actually decreases when superfluid <sup>3</sup>He is contained within narrow spaces between thin flat plates, thus indicating that surfaces actually tend to stabilize the A phase (Ahonen, Krusius, and Paalanen, 1976; Freeman and Richardson, 1990). This conclusion is understandable in view of the highly anisotropic nature of the A phase, since the orbital angular momenta of the Cooper pairs in the A phase align themselves normal to surfaces, which cannot happen in the B phase. The Aphase thus gains two advantages: First, the specular scattering of a Cooper pair conserves its angular momentum component normal to a surface, thus failing to cause pair breaking. Second, the gradient energies associated with variations in the amplitude or phase of the BCS wave function are lowest for variations along the angular momentum vector l. Thus the A phase order parameter recovers from pair breaking by the surfaces in a shorter distance than that of the B phase, minimizing the surface energy associated with the superfluid-solid interface.

One can worry about numerous more subtle effects that might at least lower the barrier to nucleation. For instance, quantized vorticity in superfluid <sup>3</sup>He can produce singularities in the liquid-crystal-like textures present in both the A and B phases, and other singularities can reside on surfaces. Experiments have shown that the nucleation rate is not increased by sample rotation (Hakonen et al., 1985), however, and careful calculations by Leggett and Yip (1989) found no singularities likely to suppress the nucleation barrier sufficiently to allow Bphase nucleation. Such exotic mechanisms as quantum tunneling have been proposed (Bailin and Love, 1980), but simple calculations show that they are even worse candidates to explain the transition than ordinary homogeneous nucleation (Leggett and Yip, 1989; Leggett, 1992).

## C. The baked Alaska model

As a possible solution to this mystery, Leggett (1984) proposed a diabolically clever nucleation mechanism, called the "baked Alaska" model for its topological similarity to the dessert of the same name. This model involves the interaction of cosmic radiation with <sup>3</sup>He at

mK temperatures, combined critically with the nature of thermal excitations in the liquid at these temperatures. In the model, cosmic ray muons traveling through the sample chamber produce secondary electrons [Fig. 4(a)] that stop somewhere within the superfluid. The rate of energy loss by an energetic electron to the surrounding medium generally varies inversely with its energy, thus a substantial amount of energy is deposited within a short distance of its stopping point [Fig. 4(b)]. A sufficient amount of energy might then be deposited with a radius  $R_c$  of the stopping point to warm a critical volume of the superfluid above  $T_c$ .

Normally, the heat from such an event would propagate away from its source diffusively, but this is not the case in liquid <sup>3</sup>He near  $T_c$ . The elementary thermal excitations are quasiparticles, namely, <sup>3</sup>He atoms with energies above the Fermi energy. The probability of such quasiparticles scattering off one another is proportional to their number, which is proportional to T, times the number of empty states available in which to scatter, which is also proportional to T. Thus the scattering rate varies as  $T^2$ ; and at  $T_c$ , roughly a factor of 400 below the Fermi temperature, the quasiparticle mean free path is  $\sim 2 \,\mu m$ , which exceeds  $R_c$ . In the absence of impurities and defects in the fluid, no other scattering processes occur, allowing quasiparticles near  $T_c$  to travel ballistically away from the heat source, as shown in Fig. 4(c). In time, these excitations evolve into a hot expanding shell as shown in Fig. 4(d), while the region near the heat source rapidly cools to below  $T_c$ .

Leggett argued that the time to cool below  $T_c$  inside the expanding quasiparticle shell was quite short, allowing the fluid to condense occasionally into the *B* phase rather than the *A* phase when its temperature first fell below  $T_c$ . If the superfluid inside the shell could not convert to the energetically preferred *A* phase before cooling below  $T_{AB}$ , this *B* fluid within the shell would become



FIG. 4. Schematic representation of the baked Alaska process as described in the text.

stable. The presence of the quasiparticle shell, separating the bubble of *B* superfluid from the surrounding *A* phase, would eliminate the surface energy which ordinarily causes such a bubble of *B* superfluid to collapse even at  $T < T_{AB}$ . If the shell did not dissipate before the dimensions of the *B* phase bubble exceeded  $R_c$ , the *B* phase would expand to fill the sample volume, just as it would in homogeneous nucleation.

Assuming a low probability of depositing sufficient energy for this process to work, Leggett showed the lifetime of the supercooled A phase to be proportional to  $\exp(\alpha R_C^N)$ , where  $\alpha$  depends upon the rate of energy dissipation by the electrons, and N depends upon details of the process. Requiring that sufficient energy be deposited to drive an entire critical volume above  $T_c$ , he found N=5, whereby the lifetime should vary as  $\exp(\alpha(R_c)^5)$ . However, if the process were to require energy sufficient to drive only a thin shell of radius  $R_c$  to its normal state, the lifetime should vary as  $\exp(\alpha(R_c)^3)$ . Both functions would provide for very rapid decreases in supercooled Aphase lifetime with decreasing temperature, but Leggett chose N=5 because all previous experiments had observed nucleation to occur rapidly upon reaching a certain critical temperature, which seemed to vary between different experiments.

In 1986–1987 a group at Los Alamos National Laboratory (Buchanan *et al.*, 1986; Swift and Buchanan, 1987) tested the Leggett hypothesis by searching for correlations between B phase nucleation in a sample cell and cosmic rays passing through two scintillation detectors placed above and below that cell. After several hundred nucleations, no statistically significant correlation between the two events emerged. Although this result was interpreted as discounting the baked Alaska theory, Leggett viewed the geometry of the experiment as far from ideal, since energetic particles could enter from the sides without triggering both (or either) scintillation detectors, and radioactive sources inside the cell could also induce the baked Alaska process without triggering the detectors.

## IV. THE RECENT EXPERIMENTS AT STANFORD

#### A. Experimental design

A new set of experiments on the *AB* transition has been conducted recently at Stanford University (Schiffer, O'Keefe, Hildreth, *et al.*, 1992; Schiffer, 1993) with a sample cell designed to test whether surface effects or textural singularities caused by rough surfaces are responsible for nucleating the *B* phase (Fig. 5). To create an environment free from surface roughness, the <sup>3</sup>He was contained in extruded cylindrical tubes of fused silica with surfaces smooth on the scale of 100 Å, smaller than the coherence length of the superfluid and 100-fold smaller than  $R_c$ . The open ends of the tubes were capped with 0.1-micron-pore filters in a clean room to keep dust from entering.



FIG. 5. Sample cell used in the AB nucleation experiments. The drawing on the left has been distorted for labeling purposes, while the drawing on the right shows the correct aspect ratio. Two empty tubes were used to study the transition, while a third contained Pt powder for Curie-law thermometry.

To study the AB transition in these tubes, it was necessary to prevent any AB interface produced by nucleation elsewhere in the sample cell from reaching the <sup>3</sup>He in the experimental tubes. This was especially important, since the superfluid makes thermal contact with the nuclear demagnetization refrigerator through a sintered silver heat exchanger with very rough surfaces. The tubes were thus passed through holes in a NdFeB permanent magnet. The high field within each hole would stabilize the A phase to arbitrarily low temperatures, forcing each tube to nucleate independently. The continuous body of superfluid passing through the magnet provided thermal contact with the <sup>3</sup>He in the tubes.

Nuclear magnetic resonance served to detect the AB phase transitions in the tubes remotely, taking advantage of the quite different NMR properties of the two phases. A NMR coil was wound on the outside of each tube, and a static magnetic field, 28.2 mT unless otherwise stated, was applied normal to the tube axes. The NMR signal of the A phase has nearly the same strength as that of the normal phase, but its resonant frequency is shifted above the Larmor frequency by a temperature-dependent amount. The temperature dependence of the A phase resonant frequency afforded an added advantage by indicating the temperature of the <sup>3</sup>He in the bottom of each tube whenever the NMR signal was measured. The Bphase susceptibility is  $\sim \frac{1}{3}$  that of the A phase at low temperatures, with a NMR absorption spread out over a relatively large range of frequencies by textural effects in the tubes' confined geometry. These two effects combine to reveal the AB transition as an effective disappearance of the resonance signal from its shifted A phase frequency.

sure as indicated in Fig. 6.

# B. Initial observations of B phase nucleation

Although the B phase did nucleate in both sample tubes whenever the superfluid was cooled sufficiently, the A phase could be supercooled over a broad range of pressures to significantly lower temperatures than had previously been possible. Near the melting pressure, where  $T_{AB}(H=0)$  equals 1.93 mK (i.e., 0.78 $T_c$ ), the A phase never nucleated much above  $0.5T_c$  [the lowest previously recorded temperature for the A phase (Fukuyama et al., 1987)]. At that pressure the A phase could typically be maintained at  $T \sim 0.4T_c$  for several hours before the B phase nucleated, and could occasionally be cooled to temperatures as low as 0.36 mK (or  $0.15T_c$ ), remaining stable as long as 30 minutes. Studies at 5, 12, 21, and 29.3 bars, as well as near the melting pressure, showed the supercooling of the A phase to decrease with decreasing pressure (Fig. 6). The solid lozenges in the figure indicate the lowest temperatures at which the A phase could be held sufficiently stable to measure the temperature with a Pt NMR thermometer (either in complete thermal equilibrium or in dynamic equilibrium during slow cooling). The solid squares indicate the minimum temperatures to which the A phase was cooled by rapidly decreasing the temperature (~30-50  $\mu$ K/min). The pressure dependence of the supercooling is consistent with the observations of earlier workers who studied the AB transition as a function of pressure, although they all reported nucleation at higher temperatures at each pres-



FIG. 6. Supercooling obtained in the Stanford experiments at 28.2 mT as a function of pressure, indicated by the solid squares and lozenges, as explained in the text. The minimum temperatures at which other workers observed nucleation of the *B* phase at various magnetic fields are also shown: open circles, Swift and Buchanan (H=0); open lozenges, Swift and Buchanan (H=20.0 mT); open square, Swift and Buchanan (H=20.0 mT); solid triangle, Fukuyama *et al.* (H=0);  $\times$ , Hakonen *et al.* (56.9 mT); open triangles, Hakonen *et al.* (28.4 mT); and +, Kleinberg *et al.* (4.9 mT).

We mention here a few details of our observations on the nature of the nucleation process. Readers not concerned with such details may skip to the next section. While the B phase would nucleate at different temperatures each time the samples were cooled, the A phase was less stable in the longer than in the shorter tube. Nucleation occurred over a broad temperature range, between 0.36 and 1.3 mK near melting pressure. For samples cooled repeatedly to the same temperature, the time interval before nucleation would vary widely. The B phase nucleated not only while the samples were cooling, as was always seen in previous experiments, but also after having been in thermal equilibrium for several hours. The A phase could be supercooled farthest by letting the superfluid come into thermal equilibrium at a temperature where it was quasistable against the AB transition, and then cooled rapidly to low temperatures as demonstrated in Fig. 6.

Several unsuccessful attempts were made to stimulate the AB transition while the samples were deeply supercooled at about  $0.4T_c$ . High resonant rf levels in the detection coils served to saturate the A phase NMR absorption, without any evidence associating the B phase nucleation with such excitation. Other unsuccessful attempts to nucleate the B phase were made by creating acoustical noise and by hitting the cryostat (gently) while the A phase was strongly supercooled. On one occasion, a crystal of solid <sup>3</sup>He was grown in one of the tubes, but the A phase remained deeply supercooled (~1.15 mK) while the crystal was being grown and while the fluid/solid mixture remained in thermal equilibrium for about 30 minutes.

## C. Nucleation by irradiation

### 1. Data acquisition

Having failed to observe any relationship between ordinary external stimuli and B phase nucleation, the baked Alaska model was tested. Instead of correlating nucleation with the passage of cosmic-ray muons through the supercooled A phase, as tried with negative results by Swift and Buchanan (1987), the baked Alaska mechanism was simulated by bringing near the cryostat a <sup>60</sup>Co source of 1.9 milliCurie, thus producing far more highenergy electrons in the sample than would naturally occur. The decay of  ${}^{60}$ Co produces  $\gamma$  rays at 1.17 and 1.33 MeV, which easily penetrate the dewar and the various heat shields of the cryostat (producing  $\sim 5000 \gamma$ rays/second incident on the shorter sample tube). While most of the  $\gamma$  rays would pass straight through the apparatus, a few interacted with the electrons in the fused silica and in the <sup>3</sup>He samples through Compton scattering and photoionization, producing a stream of secondary electrons in the <sup>3</sup>He.

Placing the unshielded  $^{60}$ Co source near the cryostat dramatically reduced the lifetime  $\tau$  of the metastable su-

percooled A phase, but the A phase still displayed a wide range in lifetimes even when measured at constant temperature. At any given temperature, the number of trials for which the A phase remained after a time t followed an exponential decay, characteristic of nucleation by a single stochastic process. Starting with  $N_0$  samples of supercooled A phase at time t = 0, the number of samples in which the AB transition had not occurred at time  $t = t_0$  is given by  $N = N_0 \exp[-t_0/\tau]$ , where  $\tau$  is the average of the individual measurements of the A phase lifetime in the limit of  $N_0 \rightarrow \infty$ . With the  $\gamma$  source in place, the value of  $\tau$  measured in the shorter tube was consistently about twice that measured in the longer tube at the same temperature. This enhancement is consistent with the relative size (both volume and surface area) of the two tubes.

#### 2. Dependence on radiation type

The lifetime of the supercooled A phase measured in the presence of the <sup>60</sup>Co source between 0.91 and 1.33 mK is shown in Fig. 7. These data are fit well by a strong exponential function of temperature, as suggested by the baked Alaska model (discussed below and shown as a solid curve in the figure). The increase in the nucleation rate due to the presence of the <sup>60</sup>Co source was also quantified by measuring  $\tau$  in the presence of background radiation alone at a field of 28.2 mT, with results also shown in Fig. 7, for a temperature range between 1.2 and 0.87 mK. The curve drawn through these data is the same as was used to fit the <sup>60</sup>Co data, but multiplied by a factor of 1650. That both data sets are fit well by the same functional form suggests that a single mechanism is



FIG. 7. Lifetime of the metastable A phase as a function of temperature in a magnetic field of 28.2 mT with different ionizing radiations incident on the samples. All three data sets are fit to the same functional form as discussed in the text and shown by the solid lines. These were obtained by fitting the functional form to the gamma-ray data and then multiplying by constant factors of 7 and 1650 to fit the neutron and background data, respectively.

responsible for nucleation of the *B* phase in both cases, and that radiation was responsible for nucleation even in the absence of the <sup>60</sup>Co source. The relatively weak temperature dependence of  $\tau$  observed at low temperatures explains why the lowest *A* phase temperatures were attained with rapid cooling.

The lifetime was also measured as a function of temperature in the presence of a thermal neutron source from which  $\sim 2$  neutrons/second were incident on the samples. Thermal neutrons have a large cross section for capture by <sup>3</sup>He through the reaction

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

with an absorption length for the neutrons of about 100 microns for the density of <sup>3</sup>He at melting pressure. While high-energy electrons (such as those created by  $\gamma$ rays or muons passing through the samples) deposit their energy sparsely until the last few keV, which are deposited within a few microns, the 0.764 MeV kinetic energy is dissipated over a path length of about 40 microns by the resultant proton and triton which travel in opposite directions. The microscopic nature of the heating by neutron capture should therefore be very different from that generated by cosmic ray muons or  $^{60}$ Co  $\gamma$  rays, and one might expect the B phase to be nucleated differently, if at all, by the neutrons. As displayed in Fig. 7, however, the measured values of  $\tau$  in the presence of the thermal neutron source were again much shorter than in the absence of a radioactive source.

The functional form fit to the neutron data in Fig. 7 is, again, the same as for the  $^{60}$ Co data, multiplied by a constant factor of 7. That the nucleation should display the same temperature dependence for these very different energy depositions is quite surprising and not easily explained. That neutrons reduced the lifetime at all provides rather conclusive evidence that radiation alone can cause nucleation of the *B* phase, excluding the possibility that the  $\gamma$  rays were causing nucleation only through heating of the fused-silica tube walls, in which many of the  $\gamma$ 's scatter and in which the resultant electrons deposit energy more densely than in the <sup>3</sup>He.

#### 3. Dependence on temperature and magnetic field

Although the lifetime of the A phase was clearly reduced by the presence of a radioactive source, the question remained as to whether the observed B phase nucleation was, in fact, due to the baked Alaska effect predicted by Leggett. The strong observed temperature dependence of the A phase lifetime  $\tau$  lends itself to comparison with the model's predictions. We compare the data to the relatively simple early functional form discussed above (Leggett and Yip, 1989) rather than with the more precise form derived later, which does not substantially improve the agreement between theory and experiment (Schiffer *et al.*, 1995). Leggett and Yip found  $\tau \sim C_0 \exp[\alpha (R_c / R_0)^N]$ , where  $C_0$  is expressed in seconds,  $\alpha$  is dimensionless,  $R_0$  is the value of  $R_c$  in the limit of  $T \rightarrow 0$  and  $H \rightarrow 0$ , and N was estimated to be between 3 and 5. Given that  $R_c = 2\sigma_{AB} / \Delta F$  in the homogeneous nucleation theory, the known temperature dependences of  $\sigma_{AB}$  and  $\Delta F$  can be used to fit the above form to the observed temperature dependence of  $\tau$ . We assume  $\sigma_{AB} = \sigma_{AB} (T=0)(1-T/T_c)^{1/2}$ , a dependence consistent with the lowest-temperature data of Osheroff and Cross (1977), and  $\Delta F(H=0)$  is proportional to  $(1-T/T_{AB})$ , consistent with the quadratic depression of  $T_{AB}$  by a magnetic field (Osheroff, 1972; Scholz, 1981). The field dependence of  $\Delta F$  arises mainly from the different susceptibilities of the A and B phases, being represented by the term  $(H_{eff}/H_c)^2$  in the final expression

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

We define  $H_{\rm eff} = (\chi_A/\chi_B)^{1/2}H$ , assuming the nucleation to occur at constant magnetization (Schiffer *et al.*, 1995), and  $H_c = 0.63$  T in accordance with the measured dependence of  $T_{AB}$  on a magnetic field (Scholz, 1981). We take  $R_0 = 0.45 \ \mu\text{m}$  by extrapolating from the measured values of the surface energy (Osheroff and Cross, 1977) and from the critical field needed to reduce  $T_{AB}$  to 0 (Gould, 1991). This expression for  $R_c$  is probably good to 30% over our temperature range. The A phase lifetimes in the presence of the <sup>60</sup>Co source at 28.2 mT were then fit well by

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

as shown by the solid line in Fig. 7. Although the exponent 3/2 of  $R_c/R_0$  departs significantly from Leggett and Yip's estimate of 3-5, the present exponential form of the data is otherwise consistent with the baked Alaska model, and the data would certainly allow N to be as high as 2 without seriously impairing the fit.

Unfortunately, the curvature in the  $log(\tau)$  plots is sufficiently small that any number of functions of the form  $\exp[f(T)]$  fit the data adequately. For example, the data would fit an expression as simple as  $\tau_0 \exp[aT^n]$ just as well as the above function. However, if  $\tau$  is, in fact, only a function of  $R_c$ , its field dependence should also be predicted by that of  $R_c$ , through the above equation. To test this hypothesis, we measured  $\tau$  as a function of temperature in fields of 14 and 100 mT as well as 28.2 mT. The upper curve in Fig. 8 was determined by fitting the 28.2 mT data and applying our expression for  $R_c$  with  $H_{\text{eff}} = 0.173$  T, consistent with a field of 100 mT and with the relative susceptibilities, using no free parameters. The curve in the figure clearly agrees well with the 100 mT data, supporting the model and the conjecture that nucleation occurs at constant magnetization. A similar analysis for a magnetic field of 14 mT predicts little deviation from the 28.2 mT fit in our temperature range as shown by the dashed curve in Fig. 8, in disagreement with our 14 mT results at the higher temperatures, where the measured values for  $\tau$  lie significantly below the 28.2 mT data. We suspect a parallel nucleation mechanism may become important in low fields, or, alternatively, this simple functional form may not properly



FIG. 8. Temperature dependence of the lifetime of the metastable A phase in the presence of the <sup>60</sup>Co source at three different magnetic fields. The fits are described in the text. The function does not fit the high-temperature data at 14 mT well, suggesting that a parallel nucleation mechanism may be active.

account for the relevant physics [the more exact form given by Schiffer *et al.* (1995) does not, however, improve this situation greatly]. Regardless of this shortfall, the good agreement between data and theoretical expectations at higher fields strongly supports the baked Alaska model.

## D. Monte Carlo simulations

To provide a better understanding of how the radiation interacts with the samples, the Stanford experiment was simulated with the EGS4 Monte Carlo program (Nelson et al., 1985). Specifically, the simulations were intended to determine the rate at which a given flux of radiation creates "baked Alaska events," defined as the deposition of a minimum energy of a few hundred eV  $(E_{\min})$  in a small volume ( $\sim R_c$ ) of the supercooled A phase, thereby raising its temperature well above  $T_c$  and becoming a candidate for the baked Alaska nucleation process (Schiffer et al., 1995). In addition, the results of these simulations could be combined with the experimental data to calculate the efficiency,  $\varepsilon$ , of the baked Alaska process, defined as the fraction of baked Alaska events that result in nucleation of the B phase. The details of how EGS4 treats the electrons are discussed in Schiffer (1993) and more completely in Nelson et al. (1985).

In order for the *B* phase to nucleate by the baked Alaska mechanism, the model requires a baked Alaska event as described above. The rate (*R*) of such events was taken as the number of times (*N*) an amount of energy greater than  $E_{\min}$  was deposited into a 0.5- $\mu$ m-diameter volume of the simulated sample for a given radiation flux. Setting  $E_{\min} = 500$  eV (Schiffer *et al.*, 1995) and radiation fluxes appropriate to this experiment, the resulting value of *R* was about 170 events per second from the  $\gamma$  rays and about 0.025 events per second from the muons. Since the values of R estimated by this method depend strongly on  $E_{\rm min}$ , one cannot determine the exact efficiency of the baked Alaska process in nucleating the Bphase. The results do indicate, however, that at least ~100 baked Alaska events are necessary to nucleate the B phase ( $\varepsilon \sim 10^{-2}$ ), since the A phase lifetime was at least 1-2 seconds in the presence of the <sup>60</sup>Co source, even at the lowest temperatures.

Another method for estimating R counts the number of electrons stopped in the helium for a given amount of incident radiation, since each electron is likely to deposit energy densely enough to create a baked Alaska event only when stopped. This method predicts values for R of 45 and 0.012 baked Alaska events per second for the  $\gamma$ rays and for the cosmic ray muons, respectively. These values are somewhat smaller than the other estimates based on  $E_{\min} = 500$  eV, but they do not significantly change the estimate of  $\varepsilon$ . There is no simple way to predict the uncertainty in these values of R, but they are reasonably close to the values obtained through the other method, suggesting internal consistency of the results.

For  $E_{\min}$  below ~2000 eV, the Monte Carlo simulations show that the ratio of baked Alaska events produced by the known fluxes of muons and  $\gamma$  rays will be independent of  $E_{\min}$ , approximately 4000-7000 depending on the assumptions made in the calculations. Experimentally, as discussed above, the ratio between the Aphase lifetimes in the presence and absence of the <sup>60</sup>Co source was temperature independent at about 1650. This is a factor of 2-4 below the Monte Carlo value, thus in fairly good agreement given the approximate nature of the Monte Carlo calculations. Since the temperature dependences of the nucleation rates seemed identical in the absence and presence of the <sup>60</sup>Co source, the discrepancy with experiment is probably not due to any different nucleation mechanism not involving ionizing radiation. Other possible sources of discrepancy include the presence of hadron showers and the creation of baked Alaska events from secondary electrons produced outside the fused-silica tubes containing the helium.

A similar comparison of the Monte Carlo results for the  $\gamma$  rays to the experimental data can be made for the neutron data. Since the *B* phase nucleation rate with neutrons exceeds the background rate by a factor of ~230, the background effects should be minimal for them, as are those of the small flux of  $\gamma$  rays from the neutron source. The value of *R* for the neutron irradiation is estimated as

 $R_{\text{neutron}} = (\text{neutrons/second})$ 

# $\times$ (baked Alaska events/neutron).

Because the protons and tritons resulting from neutron absorption in the <sup>3</sup>He are more massive and have higher kinetic energy than electrons, they will deposit energy much more densely and over a longer path than the electrons created by the <sup>60</sup>Co  $\gamma$  rays and cosmic rays. In ad-

dition, the resultant secondary electrons have low energies ( $\sim 1$  keV) and should be stopped within a few microns. Since the resulting heat distribution in the <sup>3</sup>He will resemble a "hot sausage" rather than the point source of the idealized baked Alaska model, it is difficult to predict how many baked Alaska events should result from each neutron. Because the deposited heat is not pointlike, it is perhaps surprising that any baked Alaska events would result from such a decay, it being harder to produce a cold region surrounded by hot quasiparticles. One approach would be to assume that each charged decay byproduct is equivalent to a single baked Alaska event (as in the above analysis for electrons), a reasonable assumption since the proton and triton travel in opposite directions. With this approach, each neutron absorbed would produce two events, a minimum estimate since the high density of energy deposition should produce multiple baked Alaska events. With the estimated flux of  $2.5\pm1$ neutrons per second incident on the <sup>3</sup>He and the assumption (in view of their large cross section) that all incident neutrons are absorbed, 5±2 baked Alaska events per second would be expected under neutron irradiation. This rate is a factor of  $\sim 7-15$  lower than the low estimate of R for the  $\gamma$  rays and is in reasonable agreement with the factor of 7 difference between the experimental nucleation rates in the presence of the <sup>60</sup>Co and neutron sources, especially considering our value for  $R_{neutron}$  is a minimum estimate.

## V. CONCLUSIONS

While the problem of AB nucleation is certainly not resolved, the Stanford experiments demonstrate that ionizing radiation can lead to nucleation of the *B* phase in superfluid <sup>3</sup>He. That the nucleation rate increases by orders of magnitude when radiation sources are placed nearby is incontrovertible evidence that radiation has this effect. The question does arise whether this increase, in fact, demonstrates the validity of the baked Alaska model. While no alternative mechanism has been proposed, any reasonable possibility would undoubtedly involve much of the physics underlying the baked Alaska model. Furthermore, the model successfully predicts the observed magnetic-field dependence of the nucleation rate, and the discrepancies are not in conflict with the essential correctness of the baked Alaska model.

The possibility has been suggested (Buchanan *et al.*, 1986; Leggett and Yip, 1989) that the baked Alaska mechanism would require the coincidence of ionizing radiation with some sort of textural singularity in the A phase order parameter. While this concept cannot be ruled out completely as the source of nucleation in the Stanford experiments, it seems quite unlikely, in view of the relatively low average number of ionizing events necessary for nucleation to occur and of the low expected density of textural singularities. Both the Monte Carlo simulations and the neutron data suggest that only a few hundred baked Alaska events are needed for nucleation

at the lowest temperatures. Combined with the low expected number of textural singularities and their microscopic nature, this consideration seems to rule out the necessity of coincidence. The Stanford results certainly do not, however, rule out the possibility of textureassisted nucleation by radiation in other experimental geometries as discussed below.

The ability to supercool the A phase deeply in a smooth-walled sample cell was the second major result of these experiments. In every other reported result, experimenters saw the B phase nucleate at temperatures well above those observed in the Stanford experiments. Even taking into account that other sample volumes were up to 100 times larger, radiation alone could not have been responsible for previous nucleation at higher temperatures, given the steeply increasing values of  $\tau$  observed with increasing temperature. The most significant difference between the Stanford cell and others lies in its isolation of the samples from rough surfaces, suggesting again that nucleation in other experiments could have been associated with textural singularities created by rough surfaces.

The hypothesis that textural singularities induced by surface or geometry aid nucleation is consistent with Swift and Buchanan's (1987) finding that the *B* phase nucleated preferentially at certain positions in their sample cell. This result could also be attributed to a localized high concentration of some radioactive isotope, such as  $^{14}C$  in the epoxy of their cell, producing nucleation predominantly at those positions; but the texture-based explanation seems much more probable, since one would expect such isotopes to be rather uniformly distributed.

Yet another possible high-temperature nucleation mechanism, absent from the Stanford experiments, is somewhat related to the baked Alaska scenario. Other cells were liable to contain pockets in their surfaces connected to the bulk liquid by narrow channels ("lobster pots" in the language of Leggett and Yip, 1989) which, given the various rates of cooling, could independently condense into the B phase on passing through  $T_c$  as in the baked Alaska scenario. If such pockets were sufficiently small, the probability of nucleation of the Aphase would be low, while the AB interface would remain stuck at the mouth of the narrow channel, isolating the B liquid from the bulk A phase. Once  $R_c$  fell below the channel diameter, the interface could pop in the other direction, causing nucleation in the main cell. This mechanism would be consistent with the coolingrate dependence of the degree of supercooling observed by the Los Alamos group (Boyd and Swift, 1993). It is, however, somewhat difficult to imagine that all of the sample cells used in previous experiments had a sufficient number of such "lobster pots" to insure that at least one of them would always be guaranteed to nucleate the Bphase-especially taking into consideration the B phase's depression in the presence of walls.

In the most recent Los Alamos experiments (Boyd and Swift, 1993), and in the experiments continuing at Stan-

ford (O'Keefe *et al.*, 1995), it has been observed that in the presence of rough surfaces the A phase can be more deeply supercooled when the rate of cooling is slowest. This observation suggests that superfluid flow driven by thermal gradients (hydrodynamic heat flow) near sharp surface features may promote nucleation either with or without the assistance of ionizing radiation.

The ability to supercool the A phase deeply in smooth-walled sample cells opens up the lowtemperature and low-field portion of the phase diagram to experimental study of the A phase. Low-temperature A phase NMR measurements, conducted in parallel with the Stanford experiments described in this paper, led to the first confirmation of the theoretically predicted, lowtemperature, limiting behavior of an ABM state and to the first experimental evaluation of the zero-temperature A phase energy gap (Schiffer, O'Keefe, Fukuyama, and Osheroff, 1992). Other experiments taking advantage of the suppression of the AB transition in superfluid contained by smooth walls have also been recently conducted (Parts *et al.*, 1995).

Future experiments based on the Stanford results might include new studies of the propagation of the ABinterface at lower temperatures, where its velocity would approach the Fermi velocity. The possibility may exist of studying macroscopic quantum tunneling as a nucleation mechanism at lower sample pressures. It is also possible that this system at lower pressures might act as an efficient detector of WIMPs (weakly interacting massive particles) and other weakly interacting particles. One would have to worry seriously about background rejection, but magnetic stabilization of the A phase near virtually all sample surfaces using multipole magnets might fairly easily make the system insensitive to both  $\beta$  and  $\alpha$ decays in the walls of the sample chamber.

Although great progress has been made toward understanding how the B phase nucleates, several questions remain. Primary among these, as indicated above, are the questions of what role rough surfaces play in the nucleation process and what mechanism led to the hightemperature nucleation observed by other groups. Experiments are under way at Stanford to study how surfaces affect the nucleation rate by inserting either arbitrary rough surfaces or well-defined surface irregularities into a smooth-walled chamber (O'Keefe *et al.*, 1995).

Ideally, future experiments might take place deep underground and with all tritium carefully removed from the <sup>3</sup>He to reduce the background radiation as much as possible. Future experiments could also be conducted at lower pressures where supercooling of the transition is quite limited and nucleation occurs close to  $T_{AB}$ , possibly testing the pressure dependence of the nucleation rate suggested by Schiffer *et al.* (1995). Lower-pressure experiments would have the advantage of shorter thermal relaxation times due to the higher thermal conductivity and lower heat capacity at lower pressures and to the smaller temperature ranges through which the samples would need to be cycled to "reset" the phase between

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runs. Clearly, much study of this unique phase transition remains to be done.

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