Primary and Secondary Nucleation of the Transition between the A and B Phases of Superfluid ³He

M. Bartkowiak, S. N. Fisher, A. M. Guénault, R. P. Haley, G. R. Pickett, G. N. Plenderleith, and P. Skyba*

Department of Physics, Lancaster University, Lancaster, LA1 4YB, United Kingdom

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We have studied nucleation in superfluid 3 He across the A-B phase transition driven by a magnetic field, in a controllable environment at very low temperatures. Both $B \to A$ and $A \to B$ secondary nucleation appear to be governed by the survival of pockets of the new phase trapped at surfaces. We find that, at fields near B_{AB} , primary $A \to B$ nucleation *cannot* be triggered by ionizing or neutron irradiation even at very high intensities. In our cell primary $A \to B$ nucleation can only be triggered externally by mild mechanical shock.

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The several phases of superfluid ³He provide a system of unrivaled simplicity and purity for the study of phase transitions. One might think that elucidating the mechanisms of phase nucleation between two well-characterized coherent condensates should be straightforward. In fact, the nucleation of the first order transition between the A and B phases has been the subject of much controversy [1] and is not at all understood. The conventional picture of a nascent bubble of the nucleating phase reaching the critical radius by thermal fluctuation cannot be invoked since the critical radii are macroscopic and yield nucleation probabilities orders of magnitude lower than those observed. In consequence, alternative mechanisms have been postulated. Leggett has proposed nucleation in the warm volume locally heated by a particle event (from ambient radiation), the "baked Alaska" scenario [2]. Bunkov and Timofeevskaya [3] have considered nucleation from a similar hot spot via the Kibble mechanism [4,5]. Observations [6] have suggested that surface roughness plays a role, either via breakdown of superfluidity with superflow over excrescences or via surface traps of the wrong phase, the "lobster pots" of Leggett and Yip [7].

Previous measurements of nucleation have been made with varying temperature. In the present work we exploit instead the susceptibility difference between the phases by driving the *A-B* transition in either direction by a magnetic field. This method affords much greater control over the experimental parameters. Furthermore, since we work at low temperatures the transition occurs between almost pure condensates and excludes any influence of normal fluid-superfluid counterflow.

The experiment is made in a Lancaster nested cell stage. The sample is contained in a sapphire tube closed at the base, with an intrinsically smooth surface, which extends below the inner cell as shown in Fig. 1. The open top of the tube is closed by a thin epoxy-impregnated paper sheet pierced by a small orifice connecting the ³He inside with the bulk liquid in contact with the refrigerant. The sample is thus contained in a quasiparticle black body radiator with high sensitivity to deposited energy [8]. The sapphire

tube was cleaned by a solvent without abrasion but not assembled in the cell in a clean room.

All experiments reported here were made at zero bar. At this pressure the superfluid sample cools to $\sim 150~\mu \rm K$ at which point heat flowing out of the radiator orifice into the colder liquid outside balances the residual heat input to the sample. The sample temperature is inferred from the frequency width Δf_2 of a vibrating wire resonator, which in the *B* phase varies as $\exp(-\Delta/kT)$ [9], with Δ the energy gap.

Also shown in Fig. 1 are the superconducting solenoids which generate the field profile applied to the cell. The greater part of the field needed to reach the A-B transition is provided by a large three-coil solenoid. A smaller diameter solenoid assembly and a further set of gradient coils provide the fine-tuning. We observe the nucleation by ramping the field profile applied to the superfluid in the sapphire tube, such that the field on an isolated region of the liquid crosses the critical field B_{AB} , 340 mT.

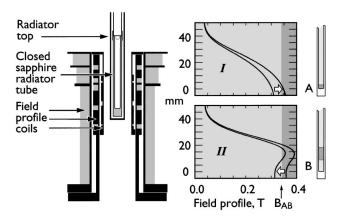


FIG. 1. Left: The sapphire quasiparticle black body radiator showing the closure at the top containing the quasiparticle radiator orifice and the heater and thermometer vibrating wire resonators, along with the solenoids for generating the applied field profiles. Right: Field profiles used for I: nucleating *A* phase at the base of the cell; II: nucleating *B* phase at the base of the cell.

Subsequent nucleation leads to the preferred phase immediately filling the available volume. Since the latent heat of the transition is large compared with our energy resolution [10], this process yields a sudden temperature transient which we detect.

Figure 1 also shows two field profiles representative of those used for the various nucleation regimes. To simplify the analysis of the thermal response all profiles are arranged to give a low field at the top of the sample cell to ensure that the 3 He surrounding the vibrating wire thermometers and at the radiator orifice is always in the B phase undistorted by magnetic field. The profiles illustrated induce nucleations at the base of the cell: I for $B \rightarrow A$ and II for $A \rightarrow B$. It became clear during the experiments that a small (~ 0.2 mm) particle of sintered silver had fallen into the bottom of the tube. Far from being a hindrance, this helps us to untangle the various nucleation mechanisms in different parts of the cell.

To make, say, an A-phase nucleation measurement, we begin with a field profile giving the B phase throughout the radiator. We then slowly ramp the current to the large solenoid, increasing the magnetic field on the superfluid until the A phase is nucleated, causing a sudden transient cooling of the radiator (the $A \rightarrow B$ signature being transient warming). We could analyze these measurements in terms of a notional nucleation rate, given some model of the process. However, for transparency we simply ramp the field uniformly (almost always at 4×10^{-5} T s⁻¹) and note the value at which nucleation occurs. These are necessarily secondary nucleations since the A phase had been present previously in the cell while we cooled into the superfluid state. Nevertheless, it is possible to observe primary nucleation of the B phase as described below.

We consider first the data on secondary nucleation. This is presented so as to illustrate the history dependence in the behavior since when we are nucleating say the A phase by increasing the field on the existing B phase, then the farther we have previously taken the field down $below\ B_{AB}$, the farther $above\ B_{AB}$ we have to take the field before the A phase is nucleated. We designate the interval we have to take the field beyond the B_{AB} value before the new phase nucleates as the "coercive" field interval, and the interval from B_{AB} which we previously took the field in the opposite direction before nucleating the new phase as the "retarding" field interval.

The principal features of our measurements can be seen in Fig. 2 where we plot the retarding versus coercive field intervals for both $A \rightarrow B$ and $B \rightarrow A$ nucleations. Both sets of data are taken with the maximum coercive fields at the base of the cell where the small sinter particle is situated. These measurements were made at 150 μ K where our temperature and energy sensitivity are greatest. However, other data show no temperature dependence up to quasiparticle densities an order of magnitude greater.

Consider the $A \rightarrow B$ data. At low field intervals there is a smooth dependence of the coercive interval on the

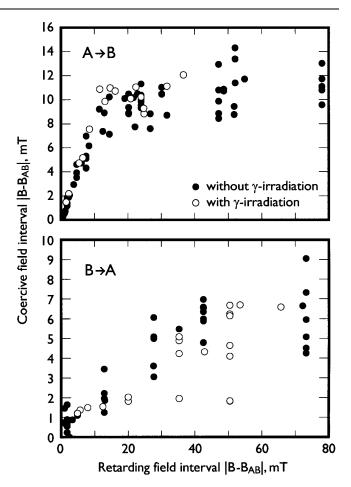


FIG. 2. $A \rightarrow B$ and $B \rightarrow A$ nucleations as functions of coercive and previous retarding field intervals (see text).

retarding interval which is quite reproducible for the same field profile. At higher retarding fields the slope falls and the data become less sensitive to the retarding field and the scatter increases.

The $B \to A$ data show qualitatively similar behavior with a more or less linear relation between coercive and retarding fields at low field intervals going over to similar retarding interval-insensitive behavior at higher fields (with no further change up to retarding fields of 330 mT). However, there are two significant differences: (a) the nucleation is always easier (that is, for the same retarding field excursion the coercive field is always lower than in the $A \to B$ case), and (b) the low field linear region now shows stochastic behavior with a wide band of scatter.

To interpret these results consider first the memory effect. Why should how far we have taken the magnetic field in the "wrong" direction have any influence when we return the field beyond B_{AB} to nucleate the preferred phase? Furthermore, why should this behavior be seen for both $A \rightarrow B$ and $B \rightarrow A$ nucleations?

We do not believe that any memory of the field history can be retained in the bulk superfluid. Thus we must suppose that the memory is retained at the cell surfaces, presumably in the form of residual pockets of the wrong

phase retained in topologically favored sites held by surface tension. This idea is not new; Leggett and Yip [7] postulated lobster pots which would do precisely this. In our sapphire cell, with a flamed surface, such defects are absent but the equivalent role may be played by the geometrical traps formed by dirt particles on the surface and especially by the sinter particle at the cell base. As the retarding field is increased beyond B_{AB} these pockets are progressively emptied. When the field is returned through B_{AB} in the opposite direction the preferred phase can grow from these preexisting seed pockets when the geometry and field energy allow. The farther that the field was previously taken in the wrong direction the fewer pockets will have survived to seed the new phase, which immediately provides a mechanism for the dependence of ease of nucleation on the previous field excursion in the wrong direction. We believe this is the process operating in the case of the B-phase nucleation shown in Fig. 2, where there is a smooth dependence of the nucleation field on the excursion field.

Since the pockets argument relies on the surface tension to retain the seeds of the wrong phase, the mechanism should operate with $A \rightarrow B$ and $B \rightarrow A$ nucleations. However, the data of Fig. 2 show that there is a clear asymmetry between the two. $B \rightarrow A$ nucleations are not only more probable than $A \rightarrow B$ nucleations for similar retarding intervals, they also show a stochastic scatter at low fields not seen in the $A \rightarrow B$ data. Possibly there is an added random factor operating in the $B \rightarrow A$ case and we suggest that background ionizing radiation may play this role. An ionizing particle traversing the cell can locally heat the liquid above T_c and thus leave a trail of regions of normal liquid along its track. If the track pierces one of the surface pockets of the A phase then at that point normal liquid is placed in contact with the preferred phase. Both A and B phases then expand into the normal region along a front at T_c but the A phase in \sim 350 mT is very much more preferred. In fact, any local heating around the pocket will help the A phase expand into the bulk since the critical radius decreases with increasing temperature. Such a process would explain the stochastic scatter of the data and since the track must intersect a pocket, the pocket memory effect would also operate as observed. Conversely, since this mechanism relies on the energy difference of the two phases in fields close to 340 mT at T_c it offers no assistance to $A \rightarrow B$ nucleations.

We can partially confirm this picture in two ways. First, we are able to induce $B \rightarrow A$ nucleation higher up the cylinder to avoid the sinter particle at the base by using a profile similar to II such that the field at the cell base never exceeds B_{AB} . Higher up the sapphire cylinder the walls are inherently smooth with very little dirt. In this case we see similar behavior to that shown in Fig. 2. However, the memory effect is restricted to lower retarding fields, and coercive field intervals up to 3 times greater than those of Fig. 2 are needed to trigger nucleation at the higher retard-

ing fields. Second, we have also investigated the effect of ionizing radiation by exposing the cell to a gamma source. In this case we indeed see enhanced $B \to A$ nucleation but no effect whatsoever on $A \to B$ nucleation as shown in the figure.

A typical time for nucleation during a field sweep is 10 min. The total local radiation flux is $\sim 10 \text{ cm}^{-2} \text{ min}^{-1}$. Typical energy deposition rates for ionizing particles would suggest a particle track creates a track of liquid heated into the normal phase over a mean diameter of about $1 \mu \text{m}$. From this we estimate very crudely that, to yield the nucleation behavior observed, there are on the base of the cell and on the sinter particle about 10^7 "seeds" with an order of magnitude fewer on the vertical walls of the cell. This seems to tally well with the idea that the seeds are associated with dirt particles and interstices in the sinter particle.

Finally, we note that a slight mechanical shock to the cryostat can also induce nucleation in both directions within the band of scatter in Fig. 2, again not inconsistent with the above picture since mechanical shock might well disturb the interface of seed pockets.

We now turn to the more interesting problem of primary nucleation. We cannot investigate primary A-phase nucleation since we cannot avoid crossing T_c in finite field. To allow primary B-phase nucleation, we must ensure that the base of the cell is held above B_{AB} from the normal state down to the lowest temperatures to avoid any B-phase precursors. We then reduce the field at the base of the cell using profile II. The greatest undermagnetization of the A phase which we can attain is ~ 30 mT, more than double the interval needed for secondary nucleation. This field deficit is just at the limit for achieving primary nucleation of the B phase and yields a spontaneous nucleation probability of the order of 1 per h; see Fig. 3A. This allows us to hold the field at this value and observe at leisure what external factors trigger nucleation. We can apply heat pulses, mechanical shock, and both gamma and neutron irradiation. We can immediately dismiss thermal pulses which have no effect. To our surprise, exposure to neutron and gamma radiation has never been seen to trigger primary nucleation in our experiments. This appears to rule out completely any of the baked Alaska and Kibble mechanism scenarios in this situation. (However, one should remember that in our case any bulk nucleation via the Kibble/"baked Alaska" mechanism with gamma radiation should be highly suppressed since we are working close to the A-B transition line where the critical bubble radius, r_c , is quite large; at our largest coercive field we estimate a lower limit for r_c of $\sim 5~\mu\mathrm{m}$ from the standard result $r_c = 2\sigma/\Delta G$ with a minimum value for the surface energy, σ , and a maximum value for the magnetic free energy difference, ΔG . B-phase nucleations should be further suppressed since the nascent B-phase domain initially generated by the fireball must cool through the transition line where the critical bubble radius diverges.)

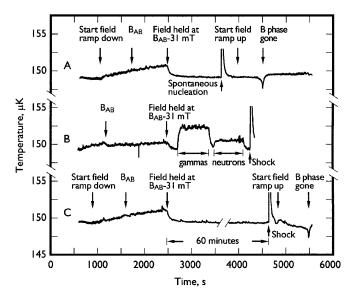


FIG. 3. Three temperature traces showing primary nucleation. Variation in the baseline arises from the thermal effects on the liquid as the field is ramped. A: spontaneous nucleation 20 min. after the coercive field reaches -31 mT. B: no nucleation on exposing the cell to \sim 700 s of gamma irradiation and a similar period of neutron irradiation. The heating of the sample is apparent. During the neutron irradiation a total of \sim 100 pJ is deposited in the liquid from capture processes which liberate \sim 700 keV of energy per neutron [11], i.e., over the 650 s of neutron exposure, around 860 fireballs above T_c were produced with no nucleation being triggered. C: primary nucleation triggered by a gentle mechanical shock after a 60 min wait at the same coercive field.

Nevertheless, it is worth emphasizing that we can apply both types of radiation up to levels so high that we observe direct heating of the liquid but no nucleation; see Fig. 3B.

The only effective external means of triggering primary nucleation is by the application of a mild mechanical shock to the cryostat, as seen in Fig. 3C. Since the specimen is contained in a radiator which is energy sensitive we can calibrate any shock in terms of the energy deposited in the liquid. We cannot detect the heat deposited at the level of shock needed to trigger nucleation. It would seem to be that a mechanical shock generates a relative velocity between the liquid in the radiator and dirt particles in the base which exceeds some local critical velocity for the breakdown of superfluidity. While our field configuration is very versatile, given the conditions under which it is used, we cannot generate such a reentrant profile as to allow us to observe primary nucleation higher in the radiator away from the base. This is a pity since in this case it would seem that primary nucleation would be virtually impossible.

O'Keefe *et al.* [6] have seen primary $A \rightarrow B$ nucleation induced by radiation, at high pressures and low fields

(but with rapidly decreasing probability up to fields of 100 mT). At our low pressures and much higher fields we see no influence of radiation whatsoever. However, the B phase is still spontaneously nucleated in our case. Given our cell configuration it is hard not to believe that all primary $A \rightarrow B$ nucleations seen under our conditions are induced by mechanical disturbance of one form or another.

To conclude, at zero bar, low temperatures and in fields near B_{AB} in secondary nucleation we see both $A \rightarrow B$ and $B \rightarrow A$ nucleation dependent on the field history suggesting a memory effect from seeds of the previous phase trapped on surfaces. Such $B \rightarrow A$ nucleation appears to be assisted by background radiation and is much enhanced by externally applied radiation. External radiation has *no* effect on $A \rightarrow B$ nucleation. Surprisingly we have never been able to induce primary $A \rightarrow B$ nucleation by radiation even at very high levels. To take the story a step farther one would need to devise a more compact radiator where a field minimum could be set up in the bulk to avoid any influence of surfaces.

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- *Permanent address: Institute of Experimental Physics, Slovak Academy of Sciences, Watsonova 43, 04353 Košice, Slovakia.
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