Nucleation of the Supercooled Normal to Superconducting Phase Transition in Small Indium Spheres Induced by γ Radiation

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(Received 21 February 1997)

We have observed that low energy γ rays nucleate the supercooled normal to superconducting phase transition in micron-sized indium spheres. The mechanism for the nucleation in the superconducting sphere may have similarities to some aspects of the "baked Alaska" model proposed by Leggett for liquid ^{3}He in which cosmic rays nucleate the transition from the supercooled superfluid *A* phase to the *B* phase. [S0031-9007(97)03535-7]

PACS numbers: 74.55.+h, 67.57.Bc, 74.25.Bt

Small, micron-sized, type I superconducting spheres ("granules") exhibit both superheated and supercooled behavior and the idea of exploiting the transition between the superheated superconducting and normal phases in the superheated superconducting granule (SSG) particle detector is due to the Orsay group [1–3]. Our group fabricated a new SSG detector based on a planar array of superheated superconductors (PASS) produced by photolithography followed by melting in the presence of a wetting agent [4], and we have been investigating its potential for detecting dark matter, i.e., weakly interacting massive particles (WIMPs), neutrons, and neutrinos [5,6]. By appropriately adjusting the temperature T and applied magnetic field B_0, the granules can be set in the superheated superconducting state very close to the line separating that phase with the normal one. Energy deposited in a granule by a particle or radiation then drives ("flips") it normal and the resulting change in the magnetic flux distribution due to the Meissner effect is sensed by a SQUID magnetometer.

It occurred to us that we could test whether the transition from the supercooled normal phase to the superconducting phase could also be induced by radiation. This might appear to be counterintuitive because the radiation would cause heating, but, in the case of liquid ^{3}He, Leggett has proposed that the nucleation of the transition from the supercooled superfluid *A* phase to the *B* phase is caused by cosmic rays [7]. In his "baked Alaska" model, the radiation produces a shower of secondary electrons in the superfluid which deposit energy of several hundred eV in the system. Quasiparticles then propagate out from a local "hot spot" in a hot shell that cools as it expands. The region of liquid inside of a shell is heated through the critical temperature for the normal to *A*-phase transition and, on the rapid recooling, it can go into the more stable *B* phase and form a nucleation center for the whole bulk of the liquid to undergo the transition. Schiffer *et al.* [8] have observed nucleation produced by ^{60}Co γ rays in smooth walled cells of ^{3}He consistent with this picture. Recently, O'Keefe *et al.* [9] have also observed effects in sample cells with rough surfaces. On the other hand, it has been proposed that the baked Alaska mechanism is not necessary for the analogous process of generation of vorticity by neutron irradiation and that diffusive cooling alone is sufficient to drive the nucleation [10,11]. A theoretical explanation of this nucleation process, which is of interest in both condensed matter physics and cosmology, has been recently proposed [12].

We speculate that a mechanism with some features of baked Alaska might also work in nucleating the supercooled normal to superconducting transition, although the length scales for the superconductor differ by orders of magnitude from superfluid ^{3}He.

The supercooling and superheating of very pure, micron-sized indium spheres, fabricated by ultrasonic dispersion, has been investigated by Feder and McLachlan by cycling the magnetic field at fixed temperature [13]. They described the supercooled and superheated transition fields, $B_{\rm sc}$ and $B_{\rm sh}$, respectively, in terms of parameters $\kappa_{\rm sc}(T)$ and $\kappa_{\rm sh}(T)$ which converge to a common value κ, the Ginzburg-Landau factor, as T approaches the critical temperature $T_{\rm C}$, and determined that $\kappa = 0.062$. The temperature dependences of $\kappa_{\rm sc}$ and $\kappa_{\rm sh}$ were seen to vary with different samples. They also discuss numerical calculations which show that as the sphere becomes small, i.e., the coherence length ξ becomes comparable to the sample dimensions, $B_{\rm sc}$ increases and $B_{\rm sh}$ decreases, with both converging to $B_{\rm c}$. Our indium spheres are produced by photolithography and melting of pure (99.99%) material. The measurements, made by cycling temperature at a fixed field, also show the full supercooling and superheating effects reported by Feder and McLaclan. Small differences in the temperature variation of $B_{\rm sc}$ and $B_{\rm sh}$ are presumably due to different sample preparation and size.

Figure 1 shows the hysteresis that results from superheating and supercooling when cycling temperature at a fixed magnetic field, $B_0 = 2.80$ mT. The granules are first all in the superconducting state at $T \sim 1.7$ K and, on increasing T, they sequentially flip from the superheated superconducting state until they are all in the normal state at $T \sim 3.3$ K. The total change of signal is the sum of all

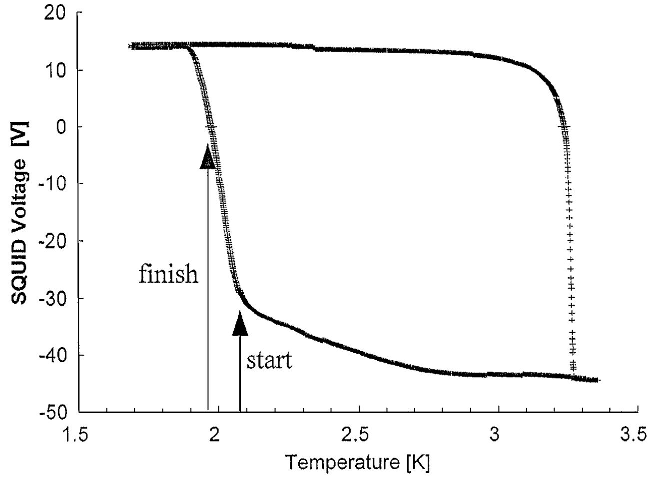

FIG. 1. The hysteresis curve for an indium PASS array in $B_0 = 3$ mT. Note that the spread of superheated superconducting to normal transitions (~20 mK) is narrower than the spread of the supercooled normal to superconducting transitions (~100 mK). This is due to the difference in the gradients of the phase boundary lines in the (B, T) phase diagram. The start and finish points of the experimental runs to investigate nucleation by γ rays are shown.

the flips of all the individual granules. The spread of superheated superconducting to normal transition temperatures is quite narrow, and this is one of the attributes of the PASS detector. On decreasing T, the granules are supercooled in the normal state until they sequentially undergo transitions into the superconducting state ("flops"). The spread of transition temperatures takes place over the interval $1.9 < T < 2.1$ K and is wider than the superheated case. This is due to the gradient dB_{sc}/dT in the (B, T) phase diagram being less than dB_{sh}/dT. The flips and flops are read out as steps on the SQUID read-out and the actual magnitude of the signals depends on the particular experimental condition pertaining in the experiment (B_0, geometry of pick-up coil, etc.). Normally, in the detecting mode [4–6], T and B_0 are set to be at the onset of the transition regime in which granules are beginning to flip (on increasing the temperature) from the superheated superconducting state to the normal state. For this experiment to test whether radiation can nucleate the superconductivity we set T and B_0 in the regime in which the granules flop (on decreasing T) from the supercooled normal state to the superconducting state, and Fig. 2 shows some of the flops actually observed in one of the runs.

The PASS sample consisted of a 100×100 square array of indium spheres on a Mylar substrate. The spheres had a diameter $d = 28$ μm and their centers were separated by 70 μm. The sample was glued on to a copper plate which was attached to a "cold finger" the temperature of which could be set with a precision better than 2 mK over the range 1.6–4.2 K. The array sat in one arm of a differential coil connected to the SQUID read-out system and a magnetic field could be applied using a

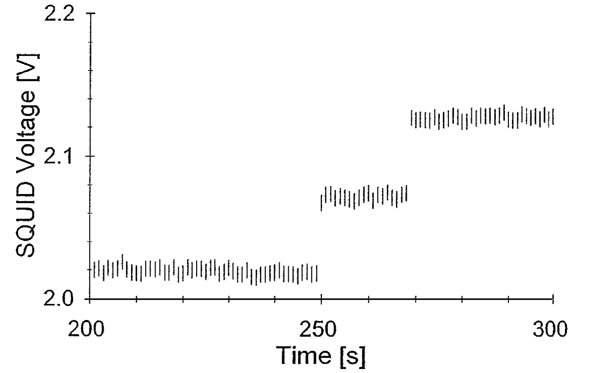

FIG. 2. Step signals from SQUID read-out due to individual granules undergoing flops from the supercooled normal to superconducting state.

superconducting solenoid. A 20 μC ^{119m}Sn source could be moved in the cryostat so that its 24 and 65 keV γ rays, with relative intensities 1:5, either impinged directly on to the PASS array or were shielded by a lead block.

The first run to investigate nucleation by irradiation was performed with the PASS sample shielded from the γ-ray source with $B_0 = 2.80$ mT and an initial setting of $T = 2.082$ K (see Fig. 1). The temperature was decreased by about 4 mK and then held at the new value for 900 s. The change in temperature caused a number of flops (~300). Ideally on stopping at the new temperature there should be no further events. However, there is always some "overshoot" in temperature so that flops continued to occur particularly over the first 100 s of the 900 s period (as shown in Fig. 3). The number of these overshoot flop events then dropped with time, but was never negligible, and counts were recorded for the time intervals t to $t + 100$ s for $t = 100$, 200, 300, 400, 500, 600, 700, and 800 s (the counts in the first interval 0–100 s being ignored). The temperature was then stepped down another

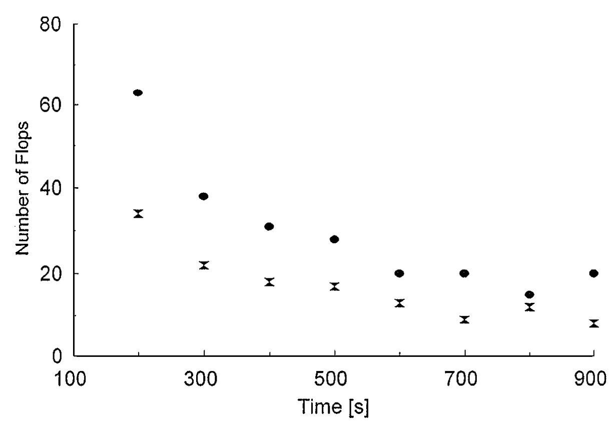

FIG. 3. Counts of supercooled normal to superconducting transitions (flops) accumulated in successive 100 s intervals after adjusting to a new temperature in the supercooled normal to superconducting transition regime. Data are for granules exposed to γ rays (●) or shielded from them (⧗).

4 mK and counts of flop events again taken over the next 900 s. This process was repeated 30 times, i.e., over a temperature interval of 120 mK so that all the grains were accessed, until the final temperature of 1.962 K was reached.

In the second run the same procedure was followed except that, in this case, the PASS array was exposed to the γ rays from the ^{119m}Sn source and the results from this run are also shown in Fig. 3. The data are the total counts for the time intervals t to $t + 100$ s in all 30 counting periods.

We conclude that the difference in the recorded number of events for the two runs is due to the flops caused by γ rays. At a given temperature there are a certain fraction of the granules whose transition from the supercooled normal state to the superconducting state can be nucleated by γ radiation. The rate at which these flops occur depends on the γ-ray intensity. As time progresses through the 900 s counting period the number of granules ready to flop diminishes so that the number of events recorded in a given 100 s period decreases with time as observed.

A control experiment was also performed. The temperature and applied field were set so that the granules were ready to undergo flips from the superheated superconducting state to the normal state and then the temperature was advanced upwards in steps. Runs were conducted with the PASS shielded and open to the ^{119m}Sn source. The number of flips in 100 s time intervals was recorded in each case. As expected, a similar analysis showed that there was an excess of flips observed in the unshielded run (by approximately the same amount as in the supercooled normal to superconducting case).

The supercooled normal to superconducting experiment was repeated, but with a 7 nCi ^{226}Ra source, which produced 4.8 MeV α particles, replacing the ^{119m}Sn γ source. In this case there was no observable difference in the two runs with the PASS sample shielded and exposed to the source. However, another control experiment performed in the same way as that described above with the ^{119m}Sn source demonstrated that α particles did nucleate the superheated superconducting to normal transition in the granules. In fact, the α source flips granules an order of magnitude more efficiently than the γ source. Note also that flips due to α particles have been observed in a previous experiment [6]. Thus one concludes that the α particles did *not* induce nucleation of the supercooled normal to superconducting transition, presumably because too much energy is deposited into a granule.

If the nucleation by the γ rays is by a mechanism similar to baked Alaska, a number of conditions have to be met:

(1) A local hot spot should be created which heats a region with dimensions greater than the coherence length ($\xi \sim 0.2$ μm for indium [13]) from about 2 K to above the thermodynamic superconducting-normal transition point at 3.2 K (in $B_0 = 2.8$ mT).

(2) The heated region should be an expanding shell so that the interior can cool to the ambient temperature, but also be protected by the shell from the exterior normal phase. The small interior sphere can then undergo a spontaneous transition into the superconducting state. It is here that there is an important difference between the situation in the indium sphere and that in the ^{3}He liquid: In the latter case, there is not just the first-order A-B transition but also the second-order transition at which the normal phase becomes simultaneously unstable to both the A phase and the B phase. However, after the small sphere has heated up and cooled down, it may go into the superconducting state because, as has been shown [13], B_{sc} is higher for a small volume with linear dimensions $\sim\xi$.

(3) Finally, in order for the small sphere to nucleate the transition of the bulk, the temperature of the whole granule must remain close to the transition point, i.e., the temperature rise of the granule must be very small.

A 24 or 65 keV γ-ray incident on an indium granule loses energy primarily by the photoelectric effect in which the photon energy is almost completely transferred to the ejected electron. The photoelectron produced by this effect itself loses the great part of its energy by ionization of atoms and electron-electron collisions along its track in the granule. There will also be some electron-nucleus collisions, but the energy loss by nuclear recoil will be at most $\sim$1 eV per collision [14]. It can be assumed that the energy loss per unit length is constant and the mean range for the photoelectron is estimated to be 1 and 10 μm for the 24 and 65 keV γ rays, respectively [15], so that the energies of the 24 and the 65 keV electrons are completely absorbed with high probabilities. If we consider the deposition of energy at a point which then heats an expanding sphere of material, the energy needed to produce a sphere of diameter ξ at temperature 3.2 K is approximately 30 eV. Since the photoelectrons lose on average about 10 eV in ionization or electron-electron collisions per atomic event there will certainly be enough energy deposited in a microscopic region to more than satisfy condition (1).

When the hot spot is very small and at a high temperature the heat spreads out by thermal diffusion: There is strong electron-phonon coupling so that entropy goes into the electronic system. As the temperature falls into the Kelvin regime, the mean free paths of the electrons and phonons become relatively long, and they decouple and propagate freely and separately. The electron mean free path λ_e in the bulk material at 4.2 K is $\sim$100 μm [16], so that it is limited by the sample size in our experiment. Consequently, $\lambda_e \gg \xi$ in the Kelvin regime and condition (2) could be satisfied.

The final temperature increase of the whole superconducting granule due to absorption of energy E from the photoelectron is simply

$$\delta T = 6E/\pi d^3 \rho c_V \,, \quad (1)$$

where ρ is the molar density of indium ($0.063\ \text{mole cm}^{-3}$) and c_V is the specific heat. The latter quantity is made up of electronic and phonon contributions, i.e.,

$$c_V = c_e + c_{\text{ph}} \,, \quad (2)$$

where $c_e = 1.69\ T\ \text{mJ mole}^{-1}\ \text{K}^{-2}$ and $c_{\text{ph}} = 1.54\ T^3\ \text{mJ mole}^{-1}\ \text{K}^{-4}$. For a granule initially at 2 K the temperature increase due to deposition of 24 keV is merely 0.3 mK, and even for the complete 65 keV deposition it would still be less than 1 mK. Whereas the ambient temperature is raised very little in the photon absorption, the situation for the absorption of α particles is drastically different. The mean range of 4.8 MeV α particles in indium is $\sim$10 μm so that if one hits a granule most of its energy is absorbed. Even if only 1 MeV is absorbed, Eq. (1) gives a global temperature increase of $\sim$15 mK. Thus condition (3) is satisfied by the γ rays, but not for the α particles because, in the latter case, the ambient temperature of the granule is raised too far above the transition region.

We conclude that a low energy γ ray, which does not deposit a large amount of energy in a granule, can nucleate the transition from the supercooled normal state to the superconducting state. A potential application for the nucleation effect is in detection of low energy radiation and particles that do not deposit too much energy into the system, e.g., WIMPs.

This research was supported by the Natural Sciences and Engineering Research Council of Canada. Discussions with P. C. E. Stamp and comments by A. J. Leggett are much appreciated.

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