Observation of Melting Instability in Highly Magnetized Solid ^{3}He

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A melting instability has been observed during rapid melting of highly magnetized solid ^{3}He. The instability occurred only if the solid is grown at low initial temperature and in high magnetic field, i.e., with high magnetization, and if the solid is melted sufficiently rapidly. After the instability of the interface occurred, the solid formed many cellular dendrites, directed parallel to the magnetic field. This is the first observation where a clear influence is seen of the magnetic field and the magnetization on a growth and melting process. The instability is attributed to a Mullins-Sekerka–type instability due to the magnetization gradient at the interface in the solid.

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Rapid growth of solids from the melt usually results in an instability of the interface, leading to dendritic growth, as described by Mullins and Sekerka [1]; however, the *melting* process is in general stable [2]. The growth instability is known to be caused by temperature gradients in the melt. Naively one might expect that the situation is reversed in ^{3}He at temperatures below the temperature of the melting pressure minimum, $T_{\min} = 316$ mK, since the latent heat is negative for $T < T_{\min}$. However, Rolley *et al.* [3] have shown that ^{3}He growth is also dendritic, while no evidence for dendritic melting was observed. So growth and melting processes in ^{3}He in zero magnetic field are similar to those investigated in more classical systems like water [4] or succinonitrile [5], with both the sign of the latent heat and of the temperature gradient reversed. Adding a magnetic field we could expect new effects on the growth and melting process of ^{3}He. The susceptibilities of the solid and the liquid are quite different and the magnetization of the solid and the liquid, for example, at 2 mK in 9 T is 90% and 4% of full magnetization, respectively. If one grows and/or melts solid faster than the spin lattice relaxation time, the magnetization should be conserved during the process. We thus have another parameter, magnetization, in addition to the mass and entropy. Castaing and Nozières [6] considered the process of a melting interface of magnetized solid phenomenologically and suggested that the magnetization in a thin layer of solid at the interface is enhanced compared to the magnetization in the bulk liquid and solid. Puech *et al.* [7] proposed that a Mullins-Sekerka–type dendritic instability could occur during the rapid melting, induced by the magnetization gradient in the solid.

Investigation of the melting process of magnetized solid ^{3}He is also interesting from another viewpoint: the study of highly magnetized liquid ^{3}He. Liquid ^{3}He forms a strongly interacting Fermi system, and while microscopic calculations do not reach the required accuracy to predict or explain the thermodynamic and kinetic properties of the Fermi liquid, various semiphenomenological models, like the paramagnon model [8] and the nearly solid model [9], have been proposed to explain the properties of liquid ^{3}He. Castaing and Nozières [6] suggested that the study of the magnetization dependence of liquid ^{3}He could decide upon the most realistic physical model. They also suggested a means to get highly magnetized liquid ^{3}He: by melting magnetized solid faster than the magnetic relaxation time. This so-called rapid melting technique was quite successful in producing magnetized liquid [10], reaching eventually magnetizations corresponding with effective fields of 200 T [11]. It became clear very soon, however, that knowledge of the melting process is indispensable in the analysis, because the measured magnetization is the overall magnetization of the liquid and solid mixture in the cell, while the physically relevant parameter like the melting pressure depends on the values of temperature and magnetization *at the interface.* Initially it was assumed [12] that the rapid pressure drop would cause the solid to break into very small pieces with characteristic size, d, of the order of the diffusion length, $L_s = D_s/v_I$; here D_s is the spin diffusion coefficient of solid ^{3}He and v_I the interface velocity. This hypothesis led to contradictory results for the susceptibility of the liquid at low magnetization. Subsequently Puech *et al.* [7] showed that a dendritic instability could occur and that the dendrites would be much larger than the diffusion length. This model gave better agreement with some of the rapid melting experiments [13]. However, no direct experimental confirmation was obtained yet.

In this Letter we present the first observation of the remarkable phenomenon, that highly magnetized solid ^{3}He breaks up in many pieces in several tens of seconds after the start of the melting process.

In order to visualize the melting process of solid ^{3}He at a temperature of a few millikelvin, we used an optical system consisting of a charge-coupled device (CCD) operated at 65 K as a camera and a light emitting diode at 1 K. The original design of the optical system and the refrigerator [14] was modified to allow measurements in a magnetic field and at lower temperatures. The sample cell,

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schematically shown in Fig. 1, is a compressional cell with a cylindrical Kapton membrane. A pressure gauge, a carbon resistance thermometer, and a vibrating wire viscometer were installed in the upper part of the ^{3}He cell. The optical cell, located at the lower part of the sample cell, was a cylinder of 5 mm in diameter and 8 mm in length with fused silica glass windows. We obtained two-dimensional projection images through the cell. In Fig. 2, each image has a resolution of 300 $\times$ 300 pixels and the diameter of the light beam is roughly 4 mm. The time interval between images was 5 sec with an aperture time of 1.2 sec. The magnetic field was applied vertically. We measured the total magnetization of the sample by cw NMR absorption with the frequency sweep method. At the bottom of the optical cell, a nucleation heater was inserted and a carbon resistance thermometer was installed. The temperature of the sample was determined by the melting pressure of the sample and the vibrating wire viscometer [15] before rapid melting and by the carbon resistance thermometer during and after melting. Details of the experimental setup are presented elsewhere [16].

The solid was nucleated by a heat pulse or by continuous heating at the bottom heater and usually a few small pieces of solid were nucleated. Then the temperature was lowered and/or the pressure was decreased until one (or sometimes two or three) seed crystal was left, from which the solid was grown at constant temperature by compressing the ^{3}He. Usually the seed crystal was located near one of the windows and the grown solid touched the window as schematically shown in Fig. 1. During growth the solid was covered by (110) facets [17]; however, it was hard to get the orientation of the facet from our images. After growing, the solid was left to anneal from a few hours to 12 h to get equilibrium. To melt rapidly the pressure was then reduced from 3.4 to 3.1 MPa, in a time between 10 to 200 sec to overlap with earlier experiments [12,13,18]. Slow "equilibrium" melting takes at least 1 h.

In Fig. 2, we show a series of pictures of a rapid melting experiment in 8.9 T with an initial temperature of 2 mK. The first image is in equilibrium after the annealing, just before the melting starts. The solid is at the bottom and the thick black line in the middle indicates the place where the solid-liquid interface touches the window. It is caused by the slight bending of the interface due to adhesion combined with the small numerical aperture of the imaging system. The pressure of the ^{3}He in the cell was reduced from 3.4 to 3.1 MPa in 100 sec. It is clear that the interface remained smooth and stable during 40 sec. Then the interface became unstable and the liquid penetrated in the solid such that only vertical cellular dendrites were left.

In Fig. 3, we show the time evolution of the ^{3}He pressure, the total magnetization (i.e., the sum of the liquid and solid magnetization in the optical part of the cell), and the temperature during a rapid melting experiment starting from 9 mK in 8.9 T. Also the time when the instability occurred is indicated. When the melting starts, the temperature increases and the magnetization decays towards the equilibrium value. The relaxation rate of the magnetization changes from fast to slow at the moment when the instability covers the whole solid. The former

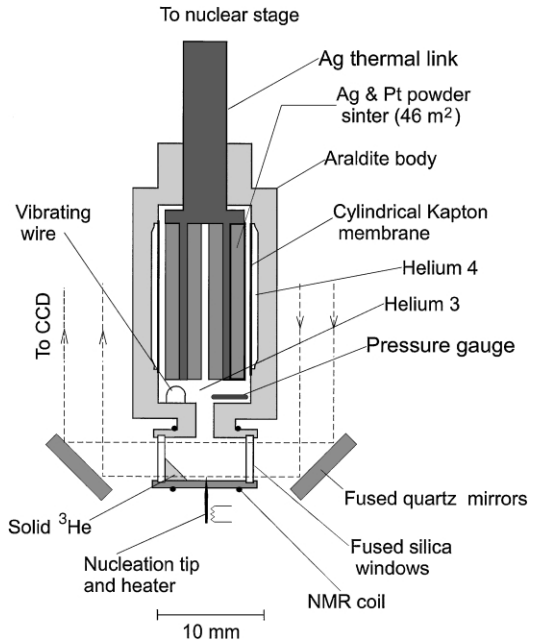

FIG. 1. The sample cell. The liquid-solid interface is usually not horizontal as shown in the optical cell. In most cases the solid touches one of the glass windows.

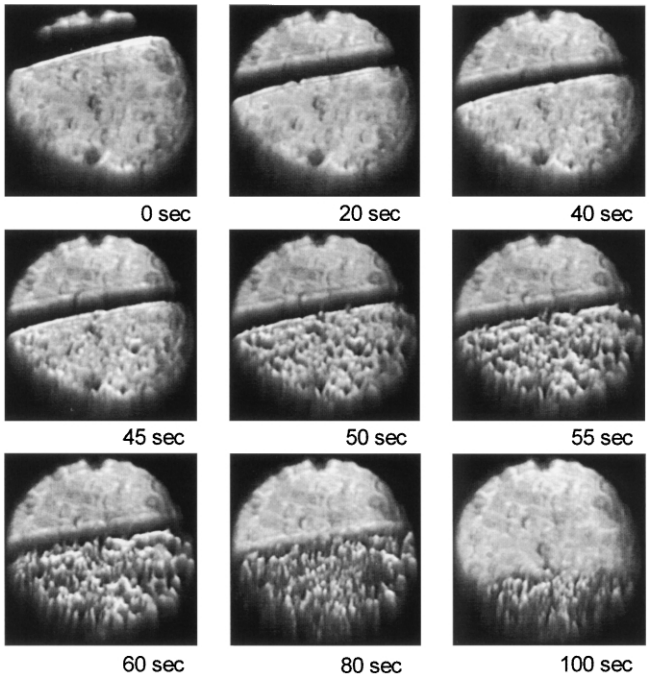

FIG. 2. CCD images of melting ^{3}He in a magnetic field of 8.9 T and initial temperature of 2 mK. The black line indicates where the solid-liquid interface touches the window.

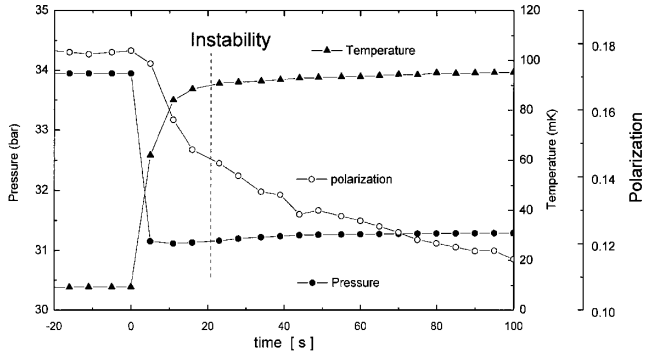

FIG. 3. The time evolution of the pressure, the temperature, and the total magnetization of the ^{3}He during a rapid melting experiment in 8.9 T and with the initial temperature of 9 mK.

decay rate is of the order of the typical relaxation rate of the solid [19] and the latter is that of liquid ^{3}He [20]. The change in the relaxation rate is related to the instability and does not indicate the disappearance of the solid. The solid disappeared at 150 sec without change in the relaxation rate. At that moment the temperature was already high and the magnetization of the solid had mostly decayed; thus the contribution of the solid to the total magnetization was small.

In Fig. 4 we show the images during a rapid melting experiment of solid ^{3}He in zero magnetic field. The initial temperature was 0.5 mK and the decompression time from 3.4 to 3.1 MPa was 30 sec. The solid is located at the left side of the image and shows two facets. It melted fast and smoothly without showing any instability.

We performed about 50 rapid melting experiments in low magnetic field ($\leq$50 mT) with initial temperature between 0.5 and 100 mK, and in 8.9 T between 2 and 100 mK. The common features are as follows: (1) The instability occurred only if the solid was grown in high magnetic field and at low initial temperature (roughly below 20 mK), i.e., highly magnetized solid, and if the

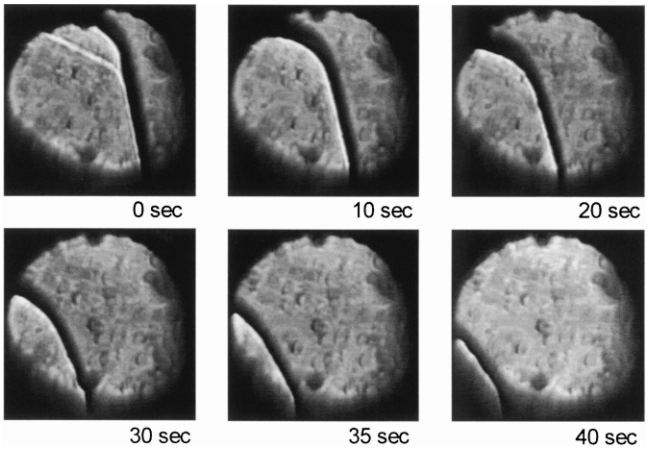

FIG. 4. Images during rapid melting of a ^{3}He crystal in zero magnetic field from 0.5 mK.

melting was sufficiently fast. No instability was observed during melting in low magnetic field nor high initial temperature ($\sim$100 mK) even in 8.9 T. The features did not depend on initial liquid phase (superfluid or normal fluid) and initial solid phase (high field phase, U2D2 phase, paramagnetic phase). We did not see the instability during equilibrium melting even in high magnetic field and at low temperature. (2) The instability originated mostly from the interface, although some images showed that nucleation of liquid droplets in the bulk solid might occur almost at the same time. (3) The melting sequence followed the order: smooth melting, instability at the interface, cellular melting, and melting of the remaining cellular dendrites. If the initial amount of solid was too small, the solid melted completely during the smooth melting process. The smooth melting persisted from a few to a few tens of seconds depending on the depressurizing rate. This is rather different from usual dendritic growth, which starts immediately when growth starts. (4) The diameter of the cellular dendrites was roughly 100 μm with the tendency that faster melting results in a smaller diameter. (5) The direction of the cellular dendrites was always the same, vertical, which was parallel to gravity and to the magnetic field. Even when we forced the flow of mass, heat, and magnetization to be horizontal with a different cell by putting a horizontal inlet tube to the optical part, the solid did melt in the same way. The direction was not related to crystal axes, either, since the facets of the initially nucleated solid seeds are randomly orientated. Since gravitational energies are negligible, we think that the magnetic field direction determines the direction of the cellular dendrites. (6) The change of the relaxation rate of the magnetization occurred almost simultaneously with the instability. The vividness of the change depended on the melting; sometimes it was clearly like a kink and sometimes it was a smooth changeover.

Next we compare our observations with the melting scenario as proposed by Puech *et al.* [7]. They considered the situation of a moving (melting) interface with $v_I > (D_s/T_{1,s})^{1/2}$, with $T_{1,s}$ the spin relaxation time of the solid, where the magnetization is enhanced at the interface in a thin layer (L_s) of solid compared to the magnetization in the bulk liquid and solid. They predicted that a dendritic instability would occur due to the gradient of the magnetization. The fact that the instability occurs only with highly magnetized solid and with sufficiently high melting speed, and that it starts from the interface, features (1) and (2), strongly supports the basic idea of Puech *et al.* [7].

They also calculated some physical parameters for the simple case of uniform temperature and no spin relaxation. They derive that a planar interface is stable only when the melting velocity is larger than a critical velocity $v_c = v_0(m_s - m_l)^2$, where m_s and m_l are the magnetization of the solid and liquid at the interface, respectively. For

reasonable estimates of the properties of ^{3}He, the prefactor is given by $v_0 \approx 20$ mm/s, and the melting velocity in our experiments is in the predicted unstable regime. The inverse of the most unstable perturbation wave vector, being more or less the radius of curvature of the dendrites, is calculated to be of the order of 1 μm for our melting speed, $v_I \sim 0.02$ mm/s, with the tendency that faster melting increases the diameter. The diameter observed in the experiment was $\sim$0.1 mm, and the tendency is opposite to the prediction. The calculated effective growth time of the instability was of the order of 50 ms for the most unstable mode and the model does not expect the sustained smooth melting we saw before the instability. Related to the vertical direction of the cellular dendrites, feature (5), they did not include any anisotropy on magnetic field direction.

The change of the decay rate of the magnetization, feature (6), can be understood by realizing that at the moment of the instability a significant amount of the solid melts into liquid in a short period, after which the main contribution to the total magnetization comes from the liquid, so the relaxation rate changes to a typical value for liquid ^{3}He. The change of the decay rate of the magnetization observed in several earlier rapid melting experiments [21] is considered to be related to the instability. The observed dendrites' diameter supports the basic assumption in the analysis of Bonfait *et al.* [13] since during the melting the radius of curvature of the interface is much larger than L_s, which is estimated to be of the order of 1 μm for our v_I.

In conclusion, we observed an interface instability during rapid melting of highly magnetized solid ^{3}He. The melting sequence followed the order of planar interface melting, the interface instability, the formation of cellular dendrites, and melting of the remaining solid. All the dendrites were parallel to the magnetic field. The instability is attributed to a Mullins-Sekerka–type instability due to the magnetization gradient at the interface in the solid proposed by Puech *et al.*

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