Oscillations in the Expansion of Solid ^{4}He into Vacuum

Giorgio Benedek,[†] Franco Dalfovo,[‡] Robert E. Grisenti,[§] Manuel Käsz,[§] and J. Peter Toennies*

Max-Planck-Institut für Dynamik und Selbstorganisation, Bunsenstrasse 10, 37073 Göttingen, Germany

(Received 9 March 2005; published 26 August 2005)

The particle flux through a micron-sized orifice into vacuum from a source chamber filled with solid ^{4}He exhibits a striking sequence of periodic bursts. The period increases (decreases) with pressure (temperature), vanishing at the melting point for temperatures above the upper λ point at 1.76 K. The oscillations are attributed to a periodic collapse of the solid induced by the accumulation of excess vacancies injected at the orifice and the period provides information on vacancy diffusivity. Dramatic deviations from this behavior below 1.76 K suggest important modifications in the flow properties of solid ^{4}He induced by the excess vacancies.

DOI: 10.1103/PhysRevLett.95.095301 PACS numbers: 67.80.Mg, 39.10.+j

In recent years the study of molecular beams of ^{4}He produced in cryogenic expansions of the gas or liquid have led to the discovery of a number of novel quantum phenomena. These include unexpected magic numbers in small ^{4}He clusters [1], microscopic manifestations of superfluidity in ^{4}He nanodroplets [2,3], orthotropic droplet beams [4], and evidence for superfluidity of para-hydrogen condensed around a molecule inside ^{4}He droplets [5]. Since helium is the only element which remains liquid down to 0 K and only solidifies at pressures above about 25 bar, it is tempting to consider an expansion from the solid phase driven by the large pressure gradient inside the source in the immediate vicinity of the orifice. The liquid formed as the pressure drops below the melting point will then be ejected as a beam. The experiments reported here confirm this expectation, but surprisingly exhibit in addition a striking oscillatory behavior of the beam intensity consisting of a periodic sequence of bursts with a period in the range of seconds or minutes. As discussed in this report these oscillations contain important new information about dynamical processes occurring in solid ^{4}He.

The apparatus is a simplified version of the one used to produce jets of liquid ^{4}He [4]. In these experiments the inlet pressure P_0 of the gas supply of the cryogenically cooled molecular beam source (temperature T_0) is raised above the melting pressure $P_m(T_0)$. The pinhole orifices have diameters ranging from $d = 1$ to 5 μm with a comparable length. The narrow beam is observed visually to be very similar to the beam of huge droplets and atoms produced in liquid jets [4]. The beam flux is determined by measuring the pressure $P_{\rm det}$ in a chamber at a distance of 10 mm from the source with a 3 mm diameter entrance aperture and pumped by two large turbo pumps (total pumping speed $S = 2760$ l/s). With the beam on, the detector pressure $P_{\rm det}$ is about 50 nbar, more than 3 orders of magnitude greater than the background pressure and the time constant of the detector response is less than 12 msec. The equation of continuity is used to calculate the exit velocity of the liquid jet from the measured flux $P_{\rm det}S$ through the relation $u = 4P_{\rm det}Sm/(kT_{\rm det}\rho\pi d^2)$, where $T_{\rm det}$ is the temperature of the detector chamber, k the Boltzmann constant, ρ the mass density of the liquid, and m the atomic mass. In these experiments u is of the order of 10^2 m/s and corresponds to a flow velocity of the solid $u_0 = (\rho/\rho_s)(\pi d^2/4A_0)u \sim 10^{-4}$ m/s, ρ_s being the solid mass density and $A_0 = 0.785$ mm^2 the inner section of the source cylindrical tube (Fig. 1).

Figure 2(a) displays a typical series of measurements of $P_{\rm det}(t)$ as a function of time at a constant source pressure $P_0 = 33$ bar for five decreasing source temperatures starting from the liquid He I phase and extending far into the solid phase as indicated in the phase diagram (Fig. 2, inset). Figure 2(b) shows a similar sequence of runs for increasing pressures at a constant temperature $T_0 = 1.88$ K. The periodic pulses appear only when the helium in the source is

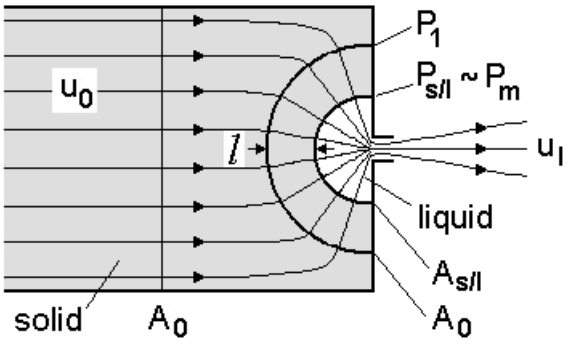

FIG. 1. Schematic diagram showing solid ^{4}He (gray area) flowing with a velocity u_0 (of the order of 10^{-4} m/s) inside the source. A micrometric diameter pinhole is centrally located at the end of a cylindrical copper tube of inner section $A_0 = 0.785$ mm^2. The solid fills the tube except near the orifice, where the pressure drops to almost zero at the orifice. The solid-liquid interface of area $A_{s/l}$ is located at a pressure $P_{s/l}$ approximately equal to the equilibrium melting pressure P_m. A large pressure gradient occurs in the constriction region of the solid flow (of thickness l), between the solid-liquid interface (at pressure $P_{s/l}$) and the section at pressure P_1. The liquid exits with a velocity u_l of the order of 10^2 m/s.

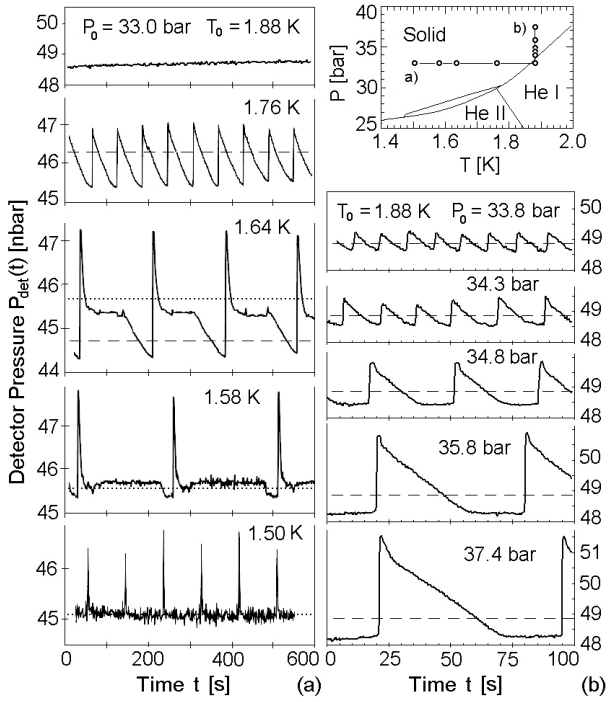

FIG. 2. Time dependence of the detector pressure signals for expansion through a 2 μm diameter orifice, for decreasing source temperatures at a constant pressure $P_0 = 33.0$ bar (a) and for increasing source pressures at a constant temperature $T_0 = 1.88$ K (b). The corresponding points (○) in the ^{4}He phase diagram are shown in the inset. The uniform signal from liquid ^{4}He at $P_0 = 33.0$ bar, $T_0 = 1.88$ K is shown in the top panel of (a). For comparison the signal at the melting pressure [$P_0 = P_m = 33.4$ bar in panel (b)] is indicated by a horizontal dashed line. In the anomalous region [$T_0 < 1.76$ K, panel (a)] the signal at the pressure $P_0 = P_u$ (see text and Fig. 4) is indicated by a horizontal dotted line.

solidified. They amount to only a few percent of the total detector signal, have reproducible characteristic shapes with maxima and minima which lie above and below the signal measured for the liquid at the melting pressure. The periods are remarkably constant to within less than 5% over measuring times of the order of hours.

A plot of the pulse period τ_0 for a wider range of source pressures and temperatures is presented in Figs. 3(a) and 3(b). Above the upper λ point ($T_0 > 1.76$ K), which will be referred to as the region of regular behavior, the period invariably vanishes at the melting point [marked by T_m and P_m in Figs. 3(a) and 3(b), respectively]. For decreasing temperature at constant pressure the period increases with an incipient linear behavior at T_m (if $T_m > 1.76$ K). However, at all pressures P_0 the period reaches a maximum at $\sim$1.57 K [Fig. 3(a)] and then drops abruptly at lower temperatures. This sharp drop-off will be discussed later on. As P_0 approaches P_m the period vanishes according to $(P_0 - P_m)^\gamma$, where $\gamma = 0.50 \pm 0.07$ at $T_0 = 1.85$ K and 0.64 ± 0.07 at 2.17 K [Fig. 3(b)]. Very similar results were obtained with different diameter orifices. Moreover the period did not exhibit any noticeable hysteresis for increasing or decreasing pressures or temperatures over their entire range.

Whereas the signal for the liquid phase ($P_0 < P_m$) rises with pressure according to Bernoulli's law $P_{\rm det} \propto u = (2P_0/\rho)^{1/2}$ [6], above the melting pressure ($P_0 > P_m$) the signal and the corresponding exit velocity u averaged over the periodic oscillations remain constant with $u = (2P_m/\rho)^{1/2}$. This confirms the assumption that an interface exists inside the source between the solid and the liquid at the pressure $P_{s/l} = \frac{1}{2}u^2\rho$ which determines the instantaneous exit flow velocity of the liquid and oscillates periodically around P_m between a maximum $P_{s/l,\rm max}$ and a minimum $P_{s/l,\rm min}$ [Fig. 2(b)].

A stick-slip mechanism for the oscillations in the solid flow is immediately ruled out since, as generally expected and shown by Rozman *et al.* [7], the slip-and-stick frequency increases with the velocity and therefore with the applied pressure, whereas the oscillation frequencies observed in our experiments decrease for increasing pressure [Fig. 3(b)]. Experiments by Gordon *et al.* [8] show that solid He can flow inside a cylinder cell as a whole with negligible friction at the cell wall, even at temperatures as low as 1.4 K, and a very small pressure gradient. Also temperature changes are excluded since the melting temperature oscillations associated with the relatively small (1 to 5%) melting pressure oscillations would have to be as large as about 10% or more, which is not observed. An

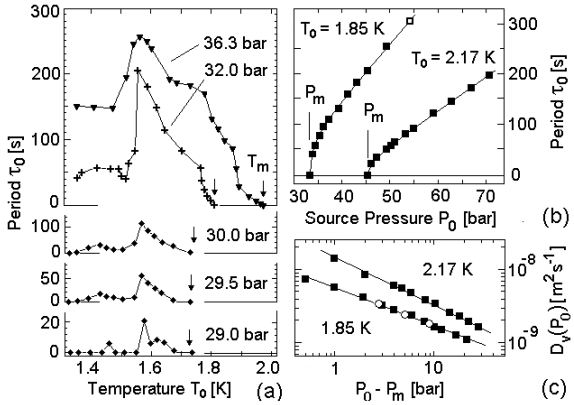

FIG. 3. (a) The oscillation period as a function of temperature at five different pressures measured for a 2 μm diameter orifice. The period increases for decreasing temperature below the melting temperature T_m (arrows) up to a maximum at $\sim$1.57 K, and then drops abruptly at lower temperatures. For $P_0 < 28.5$ bar the period vanishes completely everywhere. (b) The oscillation period as a function of pressure at two different temperatures above 1.76 K. The period vanishes at the melting pressure P_m. (c) The vacancy diffusion coefficient $D_\nu = L^2/\tau_0$ as a function of $P_0 - P_m$ with the length constant $L = 0.6$ mm fitted to the data of Zuev *et al.* [13] at 1.85 K (open circles). In all diagrams the data points have been connected by lines as guides for the eye.

oscillation of the melting pressure at fixed temperature necessarily implies the oscillation of some other coordinate of the solid and there seems to be no other disposable intrinsic parameter than the concentration of vacancies. Indeed Berent and Polturak [9] have observed that large plastic deformations in solid ^{4}He are driven by the motion of vacancies. In the present case the largest deformations occur in the region where the solid meets the highest resistance, i.e., in the shrinking region where a constriction of the solid flow cross section occurs from the upstream area A_0 to the area $A_{s/l}$ of the s/l interface (Fig. 1). It may then be assumed that the pressure gradient $\nabla P = (P_1 - P_{s/l})/l$ in the constriction region of thickness l (Fig. 1) is much larger than the average gradient $(P_0 - P_1)/l_0$ in the rest of the He feeding line of length l_0 ($\gg l$). Excess vacancies injected at the s/l interface, via the release of atoms from the solid into the liquid, are driven into the solid by the strong pressure gradient with a net force $F = (V_a - V^*)\nabla P$, where $V^* \cong 0.45 V_a$ is the isobaric vacancy formation volume [10,11] and $V_a \cong 34$ Å^3 is the solid ^{4}He atomic volume. This force pushing vacancies towards a higher stress has been introduced by Herring in the theory of diffusional creep in polycrystalline materials (Nabarro-Herring mechanism) [12]. Since vacancies can propagate further into the low-gradient region only by diffusion, they will concentrate with time in the shrinking region, until, at some critical vacancy concentration, a local collapse of the solid occurs, with an annihilation of vacancies and a resetting of the initial conditions. The ratio of the critical vacancy concentration near the s/l interface X_v^c to the corresponding equilibrium concentration at melting can be estimated from the pressure dependence of the vacancy concentration [9] as $X_v^c/X_v^m = \exp[V^*(P_m - P_{s/l,\min})/kT]$. Since $P_{s/l,\min}$ is smaller than P_m, X_v^c turns out to be larger than the equilibrium concentration at melting X_v^m. This explains the origin of the instability and of the periodic bursts in the beam flux, similar to those of a *geyser* effect. For example, for $P_0 = 54$ bar, $T_0 = 1.85$ K [open square in Fig. 3(b)], X_v^c is about 13% larger than X_v^m and 3.7 times larger than the equilibrium concentration X_v^0 at P_0, i.e., far away from the shrinking region. In the present range of pressures and temperatures X_v^0 is typically $\sim 10^{-2}$ [13], and therefore the flow velocity oscillations, also amounting to no more than a few percent of the average u_0, are compatible with a contribution of vacancies.

Since τ_0 is the time needed for excess vacancies to diffuse (drift) into the shrinking region and to accumulate up to the critical concentration, this time is inversely proportional to the vacancy diffusivity $\bar{D}_v$ (mobility $\bar{\mu}_v = \bar{D}_v/kT$) averaged over the pressure gradient. Averaging $\tau_0^{-1} \propto (P_0 - P_m)^{-\gamma}$ over the gradient leads to $D_v(P_0) = \bar{D}_v/(1+\gamma)$, and this is set equal to L^2/τ_0 with L a length constant. A fit to the vacancy diffusivity data by Zuev *et al.* [13] at 1.85 K [Fig. 3(c), open circles] gives $L = 0.6$ mm. Our new data [Fig. 3(c), full squares] confirm the temperature and pressure dependence of D_v measured by Zuev *et al.* [13] with an extension to higher pressures and temperatures. The present vacancy mechanism is also supported by a theory of the vacancy injection kinetics [14]. The solution of the transport equation, derived for a rather idealized case (constant, infinitely extending gradient and diffusivity independent of pressure), is found to account well for the shapes of the flux observed within a period in the regular region [14].

This analysis, however, is unable to explain the anomalies below 1.76 K. Figure 4(b) shows the typical behavior of the period as a function of pressure in the anomalous region (here $T_0 = 1.39$ K) and the corresponding beam flux. In this case the pressure above which the flux is constant, denoted by P_u, no longer coincides with the pressure at which the period vanishes (P_τ) and both are larger than the melting pressure P_m. Thus, although the source is filled with solid it continues to flow as if liquid up to P_u. In this regime P_u, and no longer P_m, is the threshold for signal oscillations [Fig. 2(a)] below which the instability occurs. Above P_u there is a region of pressures in which the flow is constant, but oscillations are not observed. The perfect reversibility of $P_u(T)$ when measured

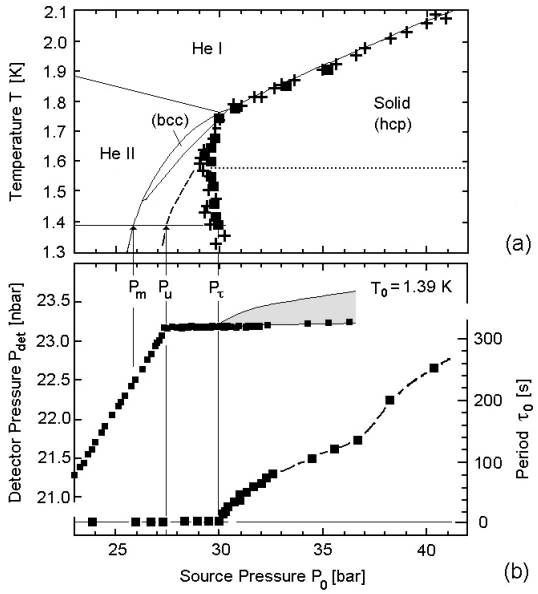

FIG. 4. (a) Phase diagram of ^{4}He with the points at which the oscillation period τ_0 vanishes for $d = 1$ μm (■) and 2 μm (+). (b) The oscillation period (right hand scale) and amplitude (gray area, left hand scale) as a function of pressure at $T_0 = 1.39$ K vanish at a pressure P_τ which is 4.2 bar above P_m. The corresponding mean flux intensity (expressed by $P_{\rm det}$, left hand scale) shows the sudden switch from the Bernoulli regime to a constant value, not at P_m as in the normal case, but at a pressure P_u which is almost 2 bar above the melting pressure. The locus of P_u is shown in (a) as a dashed line. It intersects the locus of P_τ at $T = 1.57$ K (dotted line) where the period drop [see Fig. 3(a)] is observed.

at increasing or decreasing pressures and constant temperature T_0 indicates that some change in the properties of solid ^{4}He occurs at $P_u(T)$, presumably induced by the injected excess vacancies. The dramatic changes observed at P_u are also reflected in the sudden sharp drop of the period at $T_0 = 1.57$ K [Fig. 3(a)] which coincides with the temperature at which $P_u = P_\tau$. An important observation is that the addition of 1% of ^{3}He is sufficient to reestablish the regular behavior (vanishing τ_0 at P_m and no drop of τ_0 below 1.57 K), which suggests that the described phenomena are related to vacancies. It is also interesting to note that the region where the anomalies are observed corresponds to three of the four scenarios which have been theoretically considered by Galli and Reatto [6] for a vacancy induced supersolid phase [15,16]. Recent theoretical studies [17] on the measurements of nonclassical rotational inertia in solid ^{4}He reported by Kim and Chan [16] argue that equilibrium vacancies may not be suffi>cient to trigger a transition to a supersolid phase. Burovski *et al.* [18] suggest an alternative explanation of Kim and Chan experiments in terms of interface superfluidity at the grain boundaries. This interpretation appears to be consistent also with the present surprising observation of a Bernoulli-like flow of the solid below P_u.

We thank Luciano Reatto and Davide Galli (University of Milano) for valuable discussions, Oleg Kornilov for a critical reading of the manuscript and Jens Pick and Pablo Nieto for technical assistance.

*Corresponding author.
Email address: jtoenni@gwdg.de
†Permanent address: Dipartimento di Scienza dei Materiali and Istituto Nazionale per la Fisica della Materia, Università di Milano-Bicocca, Via Cozzi 53, 20125 Milano, Italy.
‡Permanent address: INFM-BEC and Dipartimento di Fisica, Università di Trento, Via Sommarive, 38050 Povo, Italy.
§Permanent address: Institut für Kernphysik, University of Frankfurt, Max-von-Laue-Str. 1, 60438 Frankfurt/Main, Germany.

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