The geyser effect in the vacuum expansion of solid $^3$He$_{0.54}$$^4$He$_{0.46}$ and the determination of the Poisson ratio

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Abstract. Periodic bursts, also called the geyser effect, previously observed in the vacuum expansion of pure and <1% $^3$He-doped solid $^4$He, are shown to occur also in solid $^3$He–$^4$He alloys. Results are reported for $^3$He$_{0.54}$$^4$He$_{0.46}$ for the temperature range of 1.73–2.13 K and pressures up to 92 bar, and then compared with the results for pure solid $^4$He. The geyser period is found to increase with pressure much faster in the mixed solid than in pure solid $^4$He. A direct measurement of the internal pressure exerted on the lateral walls of the source chamber provides the first information on the Poisson ratio of polycrystalline pure and mixed solids.

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1. Introduction

In recent years, much experimental and theoretical effort has been devoted to understanding the low-temperature physical nature of solid pure $^4$He, largely stimulated by the claimed observation of superfluidity [1]. The most recent consensus view indicates that the anomalies observed in the non-classical rotational inertia, shear modulus and the specific heat are not due to superfluidity of the ideal commensurate solid, but rather they must be related to vacancies and other intrinsic defects such as grain boundaries or dislocations, or extrinsic factors such as the $^3$He impurity level [2]. Defects of a similar nature have been invoked to explain a phenomenon called the geyser effect, observed in the expansion of solid $^4$He through a small orifice into vacuum under constant source pressure ($P_0$) and temperature ($T_0$). The flow, which in itself was unexpected, was dominated by periodic intensity bursts [3]. These were attributed to the injection of vacancies from the solid/liquid (s/l) interface into the solid, followed by a sudden collapse. At constant temperature above the upper lambda point ($T_0 > T_{\lambda} = 1.76$ K), the period was shown to increase with pressure as $[P_0 - P_m(T_0)]^{1/2}$, where $P_m(T_0)$ is the melting pressure at temperature $T_0$. Similarly, at constant pressure $P_0$ the period increases for decreasing temperature, with linear behaviour $\propto [T_m(P_0) - T_0]$ in proximity of the melting temperature $T_m(P_0)$.

For $T_0 < T_{\lambda}$ three striking anomalies were observed. In one anomaly, the pressure at which the periodic bursts disappear deviates from the melting curve, lying above by up to 4 bar at the lowest measured temperature (1.32 K). Another anomaly is the persistence of a pure Bernoulli liquid flow for $P_0$ above the melting point up to a pressure $P_0$, which is as much as 2 bar above the nominal melting pressure $P_m$. A third anomaly consists of a sharp drop in the period when the temperature falls below $T = 1.57$ K at all pressures up to 36 bar and possibly higher. As $P_0$ is decreased from 36 bar the remaining small period decreases until at $P_0 = 28.5$ bar (still above $P_m$) it drops to zero, signalling a uniform flow as for a liquid [3].

In a subsequent study [4], it was shown that small concentrations of $^3$He ($\lesssim 1\%$ down to 15 p.p.m.) are sufficient to completely remove the anomalies. It is generally accepted that $^3$He impurities in solid $^4$He, by virtue of their larger zero-point motion, exert a local compressional stress on the surrounding host lattice [5]. Thus $^3$He impurities can act as traps for vacancies at temperatures below the $^3$He-vacancy binding energy $\varepsilon_b$, which is of the order of 1 K [6] and should affect the geyser effect if it is due to the kinetics of excess vacancies. Another possible scenario for the observed anomalies is that the intergranular superfluidity of polycrystalline $^4$He [7–9] explains the frictionless flow, which is eventually inhibited by segregation of $^3$He atoms to the grain boundaries. At larger pressures and temperatures, i.e. away from the anomalous region, the dispersed $^3$He atoms will also act as scattering centres for vacancies. Thus a natural question is whether larger concentrations of $^3$He as studied here will also affect the normal geyser effect.
The present experiments show that even in a solid alloy of almost equal isotopic concentrations, such as $^{3}\text{He}_{0.54}^{4}\text{He}_{0.46}$, the geyser effect is observed. The period exhibits qualitatively the same behaviour with respect to pressure and temperature as in pure $^{4}\text{He}$ solid. However, the period for the alloy at a given temperature increases with pressure much faster than for pure solid $^{4}\text{He}$, passing from a low-pressure regime, where the alloy exhibits faster oscillations than pure $^{4}\text{He}$, to a high-pressure regime, where the oscillations of the alloy are considerably slower than for pure $^{4}\text{He}$.

The present analysis of the geyser effect in the mixed solid is based on a previous extended study of the flow conditions in the whole apparatus characterizing the geyser effect in solid $^{4}\text{He}$ [10]. While the oscillatory part of the flow shaping the geyser signal was attributed to a counterflow of vacancies climbing the large pressure gradient in the constriction region near the orifice, the collapse was conjectured to occur where the pressure gradient drops to zero and vacancies accumulate up to a critical concentration. This region was originally thought to be just above the orifice constriction [3], but new experiments in which also the pressure pulses in the source chamber were measured [10] suggested that the collapse does not occur in the proximity of the orifice but in the feed line that connects the room temperature gas reservoir via the cryostat with the source chamber (figure 1). A study of the room-temperature gas reservoir pulses when the inlet valve was closed gave evidence that a plug is formed upstream in the feed line immediately after each geyser burst, and the next geyser burst occurs when the plug collapses under the increasing pressure gradient.

The pressure pulses in the source chamber were found to have time profiles very similar to those of the corresponding external detector pulses, and were accounted for by a Bernoulli flow of the liquid through the orifice [10, 11]. In that work the rise time of the geyser pulse and the following drop-off were also measured to be of the order of milliseconds. Weakly damped mechanical oscillations (ringing) of the source chamber pressure extending up to ten periods, with amplitudes $<0.2$ bar and periods of the order of 50 ms, were observed after each geyser burst and taken as evidence that the solid in the source chamber responds elastically to the sudden pressure rise. Moreover, no pressure difference could be detected in the source chamber with two pressure sensors 5 cm apart, indicating a very low average flow velocity in the $\mu$m s$^{-1}$ range. Thus, the scenario suggested by the simultaneous pressure measurements in different positions of the flow line is: (i) a plastic flow of the solid, sustained by the motion of defects, in the constriction regions near the orifice and in the feed line, with the former controlling the geyser signal shape, the latter the geyser collapse; (ii) a very slow flow of the solid inside the source cylinder, so as to keep the solid in a quasi-static condition [4].

As shown in the next section, this scenario also holds for the isotopic mixture. The quasi-static conditions of the solid inside the source and the simultaneous knowledge of the axial stress, determining the measured flow through the orifice, and the transverse stress measured by the sensor suggest a method to measure the Poisson ratio of the mixed solid from the ratio of their time derivatives. On the other hand, the shape of the geyser signals depends on the plastic flow of the solid towards the orifice and is well reproduced by the calculated vacancy upstream flow (section 3).

2. Experimental

The experimental apparatus is sketched in figure 1. Helium gas is introduced at a pressure $P_0$ into a 7.5 mm diameter copper source tube attached to a liquid-He cryostat at temperature $T_0$. 

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Figure 1. Schematic picture of the apparatus used for vacuum expansion of solid $^3\text{He}_{0.54}^4\text{He}_{0.46}$. The He isotopic mixture at pressure $P_0$ is introduced through an inlet tube into the source chamber (inset) in contact with a cryostat at temperature $T_0$. A jet of helium atoms exits through a 2 $\mu$m $\varnothing$ (inset) and streams through a skimmer into the detector chamber which is evacuated with a total pumping speed of 2760 litres s$^{-1}$. The cryostat chamber is pumped separately with 1000 litres s$^{-1}$. The change $P_{\text{det}}$ in the residual pressure of the detector chamber is directly proportional to the exit He atom flux. A pressure sensor measures the variation $\Delta P_{\perp}$ of the transverse stress exerted by the solid helium on the lateral wall of the source chamber.

As shown in the inset, a 2 $\mu$m diameter pinhole orifice is located at the end of a 1 mm diameter cylindrical protrusion. In the present experiments, a pressure sensor (high-pressure quartz sensor type 601A; Kistler Instruments, Winterthur, Switzerland) measures the changes of local pressure laterally exerted (transverse stress $\Delta P_{\perp}$) on the source tube wall 44 mm upstream of the orifice. After leaving the orifice the molten solid is converted into a visually observable directed beam consisting of a train of droplets as normally seen in liquid jets [11]. The flux of the ejected atoms was measured either from the change of residual gas pressure $P_{\text{det}}$ in the detector chamber, which is pumped by two large turbo pumps (total pumping speed $S = 2 \times 1380$ litres s$^{-1}$), or with a mass spectrometer in line with the droplet beam. The data presented in this paper have been
obtained with the first method. With the beam on, the detector pressure $P_{\text{det}}$ is about 50 nbar, more than three orders of magnitude greater than the background pressure, and the time constant of the detector response is less than 12 ms.

From the measured flux $P_{\text{det}}S$, conservation of mass leads to the following equation for the exit velocity of the liquid jet:

$$u_1 = \frac{4P_{\text{det}}S}{(kT_{\text{det}}m\rho_s\pi d^2)},$$

where $T_{\text{det}}$ is the temperature of the detector chamber, $\rho_s$ the mass density of the liquid, $m$ the atomic mass and $d$ the orifice diameter. In the region immediately upstream of the orifice between the s/l interface and the orifice, the liquid flow was found to obey Bernoulli’s law [11] so that the exit velocity $u_1$ is directly related to the instantaneous interface pressure $P_{s/l}$ by

$$u_1 \approx \left(\frac{2P_{s/l}}{\rho_s}\right)^{1/2}.$$  

Thus, in effect, the detector monitors the changes in the s/l pressure at a given temperature $T_0$. In these experiments $u_1$ is of the order of $10^2$ m s$^{-1}$ and would correspond, in the absence of an upstream plug, to an average flow speed of the solid inside the 7.5 mm diameter source cylinder.

$$u_0 = \left(\frac{\rho_s}{\rho_i}\right) \frac{\pi d^2}{4A_0} u_1 \sim 7 \mu\text{m s}^{-1},$$

where $\rho_i$ is the mass density of the solid and $A_0 = 44.18$ mm$^2$ is the cross section of the source cylinder (figure 1 (inset)). In the presence of an upstream plug, the motion of atoms inside the source chamber towards the orifice, averaged over a cross section, is even slower. The flow in the source chamber is likely to be non-uniform over the cross-section and nevertheless slow enough everywhere to ensure quasistatic conditions at the pressure sensor positions.

In the original experiments [3], only the axial components of the applied and interface stress ($P_0$ and $P_{s/l}$, respectively) were measured. In the present experiments it is now possible to monitor the transverse component $P_\perp$ of the stress with the differential pressure sensor mounted on the inner wall of the source chamber (figure 1). In a series of preliminary experiments with solid $^4$He, a second pressure sensor was located in the rear of the source chamber at the entrance of the feed line in order to monitor the pressure gradient in the source chamber during the flow. Since no measurable pressure difference was observed, the present study was carried out with a single sensor as shown in figure 1.

Figure 2 shows a comparison of the geyser signals of the detector pressure $P_{\text{det}}$ (figure 2(a)) with the transverse stress $\Delta P_\perp$ measured with the pressure sensor (figure 2(b)) for the mixed $^3$He$_{0.54}^4$He$_{0.46}$ solid at $P_0 = 58.8$ bar and $T_0 = 1.88$ K. The corresponding absolute values of the interface pressure $P_{s/l}$, as obtained from equations (1) and (2), and of $P_\perp$ are plotted in the right-hand ordinate scales of figures 2(a) and (b), respectively. The profiles of $P_{\text{det}}$ and $P_\perp$ oscillations shown in figures 2(a) and (b) are practically identical, and the relative amplitudes $\Delta P_{s/l}/P_{s/l} = 0.261$ and $\Delta P_{\text{det}}/P_{\text{det}} = 0.127$ are approximately in the ratio of 2 : 1 since, according to equations (1) and (2), $\Delta P_{s/l}/P_{s/l} = 2\Delta P_{\text{det}}/P_{\text{det}}$. The proportionality between $P_\perp$ and $P_{s/l}$ has been thoroughly discussed in [10]. Figure 2(c) shows for comparison the geyser signal for the pure $^4$He solid measured under similar pressure and temperature conditions. Although the shape of the signal is very different from that of the mixed solid, the period is almost the same.
Figure 2. Comparison of the geyser signal in the vacuum expansion of a solid $^{3}\text{He}_{0.54}^{4}\text{He}_{0.46}$ alloy as measured through the oscillation of the detector pressure (a) and of the transverse stress by the sensor inside the source chamber (b). The respective absolute values of the s/l interface pressure (a) and internal transverse stress (b) are given by the right-hand ordinate scales. Note that the scale of $P_{s/l}$ is not linear due to the relationship $P_{s/l} \propto \sqrt{P_{\text{det}}}$ [10]. For comparison, the geyser signal for pure solid $^{4}\text{He}$ at a similar pressure and temperature is shown in (c): while the geyser period is only slightly longer than that of the mixed solid, the signal profile is very different.

As seen in figures 2(a) and (b), at each geyser burst both $P_{s/l}$ and $P_{\perp}$ quickly rise to the same applied pressure $P_0$, thus resetting an initial hydrostatic condition. The minimum value of $P_{\perp}$ just before the collapse ($P_{\perp,\text{min}}$) is close to the pressure at which the geyser period, for the given temperature $T_0$, vanishes. For pure $^{4}\text{He}$, the vanishing-period pressure is just the melting pressure $P_m$ at $T_0$ [10], whereas for the mixed solid this is expected to be the solidus pressure $P_s$ [12, 13]. Similarly to solid $^{4}\text{He}$ [3], where the minimum of $P_{s/l}$ is generally below the melting pressure.

The schematic phase diagram reported in [13] shows that the region between the solidus and liquidus lines is very narrow, which justifies the terms ‘melting temperature’ and ‘melting pressure’ for the values reported in table I of [13]. Actually these values appear to coincide with the solidus values.
Figure 3. The geyser signals for the mixed $^{3}\text{He}_{0.54}^{4}\text{He}_{0.46}$ solid at $P_0 = 77.5$ bar and $T_0 = 2.05$ K shown on expanded time scales. Just after the geyser burst, some characteristic elastic oscillations in the ms time scale (ringing) are observed (a). The initial slopes of $\Delta P_{\perp}$ (b) and $P_{\text{det}}$ (c), shown on the same pressure scale and on the time scale of seconds, allow for the derivation of the Poisson ratio $\nu$ at the given pressure and temperature. The minimum of $P_{s/l}$ ($P_{s/l,\min}$ in (c), right-hand ordinate), at which the geyser burst starts, occurs about 3 bars below $P_{\perp,\min}$ (panel (b), right-hand scale). The portion of the $\Delta P_{\perp}$ signal in (b) where the ringing oscillations (a) are observed is enclosed in the small grey rectangle.

The minimum transverse stress $P_{\perp,\min}$ is plotted as a function of temperature in figure 4 for both $^{3}\text{He}_{0.54}^{4}\text{He}_{0.46}$ and $^{4}\text{He}$ samples. The $^{4}\text{He}$ data points are seen to follow as expected the corresponding melting line, whereas those of $^{3}\text{He}_{0.54}^{4}\text{He}_{0.46}$ are slightly below the line.
The minimum transverse stress measured just before the geyser collapse for $^{3}$He$^{0.54}$He$^{0.46}$ (black dots) and $^{4}$He (blue dots) solids as a function of temperature. While $P_{\perp, \text{min}}$ for $^{4}$He follows the corresponding melting line, the data points for $^{3}$He$^{0.54}$He$^{0.46}$ are slightly below the line interpolating the melting (actually the solidus) data reported by Fraass and Simmons [12] (grey dots) and the solidus data (open red circles) from Simmons [13] for the slightly different mixture $^{3}$He$^{0.51}$He$^{0.49}$. Interpolating the melting pressure data reported by Fraass and Simmons [12] for the mixture of similar composition $^{3}$He$^{0.51}$He$^{0.49}$. Since their melting data appear to correspond to the solidus line, it is tempting to associate $P_{s}$ with the pressure at which (at a given $T_{0}$) the geyser period vanishes in the mixed phases. The small deviation of $P_{\perp, \text{min}}$ from $P_{s}$, if not instrumental, may mean that in mixed systems the collapse does not occur at $P_{s}$ but slightly below in the coexistence region between solidus and liquidus.

It was previously observed in solid $^{4}$He [10] that the pressure pulse produced by the geyser burst yields mechanical oscillations (ringing) in the source chamber solid with periods in the range of a few tens of millisecond. As seen in figure 3(a), also the $^{3}$He$^{0.54}$He$^{0.46}$ solid shows some ringing oscillations after the geyser burst. The theory of ringing [10] provides for $^{4}$He a good description of the signal oscillations in terms of the elastic constants derived from the known speed of longitudinal sound. The period of 43 ms observed here for the mixed solid at 77.5 bar and 2.05 K is comparable to the 41 ms ringing of solid $^{4}$He at a similar temperature (1.94 K) but considerably smaller pressure (43.5 bar). The occurrence of such weakly damped periodic oscillations clearly indicates an elastic response of the solid. This is consistent with the fact that the oscillation amplitude, after the initial fast drop-off of $P_{s}$ (figure 3(a)) of about 0.7 bar (probably due to a rapid recrystallization), is of the order of 0.1 bar, thus well below the expected yield stress for the mixed solid. The latter must also be $\sim$0.6 bar as found in both hcp-$^{4}$He at 1.8 K [14] and bcc-$^{3}$He at 1 K [15].

Under these conditions the initial variations with time of $P_{s/l}$ and $P_{\perp}$ with respect to the hydrostatic pressure $P_{0}$ are not equal, the axial stress $P_{s/l}$ decreasing faster than the transverse stress $P_{\perp}$. Their initial time derivatives allow for the determination of the Poisson ratio $\nu$ of the solid at pressure $P_{0}$ and temperature $T_{0}$ from the equation [16]

$$\frac{(dP_{\perp}/dt)_{0}}{(dP_{s/l}/dt)_{0}} = \frac{\nu}{1 - \nu},$$

where $(dP_{s/l}/dt)_{0} \simeq 2(P_{s/l, \text{max}}/P_{\text{det}})(dP_{\text{det}}/dt)_{0}$, and the slopes of $P_{\perp}$ and $P_{\text{det}}$ are derived from the respective geyser plots at short times like those of figures 3(b) and (c). In table 1, some
values of the Poisson ratio for the mixed $^3$He$_{0.54}$$^4$He$_{0.46}$ solid are compared to values for the pure $^4$He solid obtained in the same way. The isotopically mixed solid has an average Poisson ratio which is smaller than that of solid $^4$He by only a few per cent. Within the experimental uncertainties of ±0.02 an average value of 0.38 may be taken for both samples in the considered range of pressure and temperature. This value is larger than the typical values (∼1/3) of classical polycrystalline metals [16] and also of high-pressure room-temperature $^4$He crystals [17, 18].

A more instructive comparison between the values from the geyser effect ($v_{ge}$) and those calculated as functions of the crystallographic direction [24, 25] comes from the experimental elastic constants of hcp-$^4$He and bcc-$^3$He in the quantum regime.

Figure 5 shows the Poisson ratio components calculated from the experimental elastic constants of bcc-$^3$He at a molar volume $V_m = 21.6\ \text{cm}^3\ \text{mole}^{-1}$ [19, 20] and of hcp-$^4$He at 19.0 cm$^3$ mole$^{-1}$ [20–23] (both corresponding to $P_0 = 65.5\ \text{bar}$ and $T_0 = 1.2\ \text{K}$). The basal ($v_b$) and sagittal ($v_s$) components of the Poisson ratio for hcp-$^4$He calculated as functions of the zenith angle $\theta$ with respect to the $c$-axis (and any arbitrary azimuth angle $\varphi$) [25] exhibit indeed a large angular dependence. The axial stress has the largest anisotropy in the basal direction ($\theta = 90^\circ$) and a value averaged over all axial and transverse directions $\bar{v} = 0.282$, valid for a polycrystalline sample.

For bcc-$^3$He, only the average of the two components is plotted for simplicity, using the analytical expression given by Date and Andrews [26], as a function of the angle $\theta$ from the direction [001] for both $\varphi = 0^\circ$ and $45^\circ$. In bcc-$^3$He the Poisson ratio (actually the average over the two components) shows a deep minimum ($v = 0.27$) in the $\langle 111 \rangle$ direction and an average value $\bar{v} = 0.473$ for the polycrystalline material. Since in cubic materials the Poisson ratio is equal to $c_{12}/(c_{11} + c_{12})$ in the $\langle 100 \rangle$ direction, a value of $v$ close to 0.5 implies $c_{12} \cong c_{11}$ and a very large anisotropy $A = 2c_{44}/(c_{11} - c_{12})$. Indeed, bcc-$^3$He with the elastic constants of [20] at the molar volume of 21.6 cm$^3$ mole$^{-1}$ has the largest anisotropy among all cubic elemental solids ($A = 23.7$). Also hcp-$^4$He in the quantum regime [23] strongly deviates from the quasi-isotropic condition occurring at high pressures and temperatures [17]. The most intriguing feature of figure 5 is the large difference between the average Poisson ratios of the two isotopes: bcc-$^3$He

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Table 1. Poisson ratios $v$ for the mixed $^3$He$_{0.54}$$^4$He$_{0.46}$ and pure $^4$He solids at different pressures and temperatures. The last three columns list the period $\tau_0$ and the fitted collision $\tau_c$ and recombination times $\tau_r$ from measurements with a 2 $\mu$m orifice.

| Solid           | $P_0$ (bar) | $T_0$ (K) | $\nu$  | $\tau_0$ (s) | $\tau_c$ (s) | $\tau_r$ (s) |
|-----------------|-------------|-----------|-------|-------------|-------------|-------------|
| $^3$He$_{0.54}$$^4$He$_{0.46}$ | 58.8        | 1.87      | 0.39  | 343         | 227         | 25.2        |
| $^3$He$_{0.54}$$^4$He$_{0.46}$ | 77.5        | 2.05      | 0.36  | 400         | 250         | 41.7        |
| $^3$He$_{0.54}$$^4$He$_{0.46}$ | 92.0        | 1.91      | 0.37  | 924         | 404         | 23.8        |
| $^4$He          | 48.6        | 2.04      | 0.41  | 130         | –           | –           |
| $^4$He          | 56.7        | 1.92      | 0.38  | 356         | 343         | 24.8        |
| $^4$He          | 92.0        | 2.92      | 0.38  | 130         | –           | –           |

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8 These authors find $v$ varying between 0.34 at 13 GPa kbar and 0.31 at 32.3 GPa. Although in this pressure and temperature range solid $^4$He is still hcp [19], they show that, within the experimental uncertainties, $c_{33} \cong c_{11}$, $c_{13} \cong c_{12}$ and $c_{11} - c_{12} \cong 2c_{44}$, which indicates an almost isotropic solid.

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Figure 5. The Poisson ratio for an axial stress forming an angle $\theta$ with the $c$-axis (and any azimuthal angle $\varphi$) for hcp-$^4$He and with a (001) axis, with $\varphi = 0^\circ$ and $45^\circ$, for bcc-$^3$He. For hcp-$^4$He both basal ($v_b$) and sagittal ($v_s$) components of the Poisson ratio [25] are plotted, whereas for bcc-$^3$He only the average $(v_b + v_s)/2$ is shown for both azimuths [26]. The averages $\bar{v}$ listed on the right margin are calculated over all possible axial and transverse orientations and apply to polycrystalline materials. For hcp-$^4$He the component averages are $\bar{v}_s = 0.184$, $\bar{v}_b = 0.379$, the latter being very close to the values $v_{ge}$ obtained from the geyser effect for hcp-$^4$He and $^3$He-$^4$He.

has a $\bar{v}$ close to 0.5, which confers a quasi-liquid behaviour, versus $\bar{v} \sim 0.3$ for hcp-$^4$He, a value close to that of most polycrystalline metals.

The Poisson ratio $v_{ge}(^4$He$)$ derived from the geyser effect for pure $^4$He (figure 5) is considerably larger than $\bar{v}(^4$He$)$, which would exclude a polycrystalline phase for the solid filling the source chamber. It has been suggested above that the sharp (millisecond) pressure drop of the geyser signal immediately after the collapse (figure 3(a)) could be associated with recrystallization and growth of crystallites to the size of the source chamber (Balibar et al [27]; see also Sasaki et al [27]). The fact that $v_{ge}(^4$He$) \approx \bar{v}_b(^4$He$)$ might even suggest a prevalent orientation of crystallites with the $c$-axis in the feed line direction. On the other hand, $v_{ge}(^3$He-$^4$He$)$ for the mixed solid is about the average of $\bar{v}(^3$He$)$ and $\bar{v}(^4$He$)$ and nothing can be inferred about the crystallinity of this phase, whether hcp or bcc or a mixture. The geyser effect and the signal shape apparently do not depend on the crystalline structure of the solid inside the source chamber. There are, however, relevant differences in the plastic flow dynamics in the constriction regions.

Figure 6 shows the period of the mixed $^3$He-$^4$He as compared to those of pure $^4$He, all measured with an orifice diameter of 2 $\mu$m as functions of temperature at different pressures. For $P_0$ around 45 bar the period of $^3$He-$^4$He is well below that of pure $^4$He, whereas at $P_0 = 92$ bar the period for $^3$He-$^4$He is substantially longer than that for $^4$He at the same temperature. At intermediate pressures the periods are nearly the same. Since the geyser period vanishes at the melting (solidus) line, the higher solidus pressure of the isotopic mixture readily explains why periods are shorter than those in pure $^4$He at lower pressures. More interesting is their faster increase with pressure.
Figure 6. Geyser signals for the mixed $^{3}\text{He}_{0.54}^{4}\text{He}_{0.46}$ solid for increasing pressures at nearly equal temperatures. The period increases with pressure much faster than for the pure $^{4}\text{He}$ solid.

According to the analysis of the geyser phenomenon of solid $^{4}\text{He}$ presented in a previous paper [10], the geyser burst is caused by a sudden collapse of an upstream plug due to an accumulation of vacancies drifting upstream under the pressure gradient (the plug model). According to this view the geyser rate (reciprocal of the period) depends on vacancy mobility under the given pressure and temperature conditions, so that the rate itself can be taken as a measure of vacancy mobility (or diffusivity, via Einstein relations) up to a constant depending on the system geometry [3]. Although the massive plastic flow of the solid in the constriction regions can be associated with the motion of other defects, first of all dislocations, in [3], we argued that the periodic geyser bursts cannot be attributed to a slip-and-stick phenomenon at the source walls, since the period should decrease instead of increasing with increasing $P_0$. On this basis, a model based on vacancy transport and accumulation was suggested. In the following discussion, it is shown that the vacancy kinetics can account for the actual shape of the observed geyser signals.

3. Discussion

According to the plug model described in [10], a plug is formed somewhere in the feed line due to a jamming of crystallites after solidification, and the collapse is a sort of unjamming transition.
Figure 7. Geyser periods for the mixed $^3\text{He}_{0.54}^4\text{He}_{0.46}$ solid as functions of temperature for different pressures (●), compared with the periods for the pure $^4\text{He}$ solid measured under similar conditions (□). The full and broken curves, ending at the temperature where the period vanishes (solidus $T_s(P_0)$ and melting temperature $T_m(P_0)$, respectively), are a guide to the eyes. The 77.5 bar curve for $^3\text{He}_{0.54}^4\text{He}_{0.46}$ interpolating two data points is obtained from rescaling the 58.8 bar curve as $(\tau_0, T_0 - 2.14 \text{ K}) \rightarrow (s\tau_0, sT_0 - 2.33 \text{ K})$ with $s = 2.33/2.14 = 1.088$.

triggered by the increasing pressure difference across the plug (from zero to about $P_0 - P_m$) and the consequent invasion of vacancies either diffusing or drifting against the pressure gradient or generated by the decrease of pressure. The unjamming occurs at some critical average vacancy concentration $\nu_c$. The time $\tau_0$ required to reach $\nu_c$ is inversely proportional to the vacancy mobility $\mu_v$ (or diffusivity $D_v = kT \mu_v$). The fact that both vacancy mobility and concentration rapidly decrease with increasing pressure $P_0$ (or decreasing temperature) explains why the geyser period $\tau_0$ becomes longer at higher $P_0$ (or smaller $T_0$). Once it is accepted that the geyser effect depends on defect kinetics, it appears that only thermodynamic defects such as vacancies can account for the unusual fact that the process slows down at higher pressures or lower temperatures. The role of vacancies has been underlined also by Day and Beamish [28, 29] in a study of the flow of solid helium under a pressure gradient through either Vycor or a glass capillary array at temperatures and pressures similar to those of the present experiments.

On this basis, the data shown in figures 6 and 7 suggest that at low pressures vacancies are more abundant and (or) more mobile in the mixed than in the pure solid, whereas at higher pressures the vacancy concentration and (or) mobility are (is) much reduced in the mixed solid with respect to the pure one. This is consistent with the observation of Fraass and Simmons that the vacancy concentration in $^3\text{He}_{0.51}^4\text{He}_{0.49}$ at the solidus line and 1.33 K is as large as 4% or more [13, 30] and that the vacancy formation energy rapidly increases for decreasing molar volume, from 8 K at 20.9 cm$^3$ mole$^{-1}$ to 21 K at 18.8 cm$^3$ mole$^{-1}$ [13, 31]. In this respect, the presence of $^3\text{He}$ in the $^4\text{He}$ solid lattice has two opposite effects: one is enlargement of the
lattice spacing, which favours vacancy diffusivity; the other is introduction of inelastic scattering centres for vacancies, which hinders their diffusivity. Apparently a pressure increase impairs the former mechanism and aids the latter.

This interpretation is confirmed by a comparative analysis of the geyser signals shown in figure 2(b) and (c) for $^3$He$_{0.54}$$^4$He$_{0.46}$ and $^4$He. Use is made of a previous kinetic theory describing the injection of vacancies into a solid from the s/l interface as an effect of a pressure gradient [3, 32]. According to this theory the solid flow is accompanied by a corresponding opposite flow of vacancies from the s/l interface into the solid bulk, driven by a strong pressure gradient. The time-dependent vacancy flow is held responsible for the oscillatory part of the liquid jet, i.e. for the shape of the geyser signal (except perhaps for the very first drop-off of the signal a few ms after the collapse, which is attributed to recrystallization (figure 3(a)). In the plug picture, vacancies are also climbing the pressure gradient and accumulating in the plug volume, thus provoking its collapse under the external pressure $P_0$. The kinetic equation for vacancies is similar to that for excess carriers in doped semiconductors, with a drift term (drift velocity $u_\beta$), a diffusive term (diffusivity $D_v = \tau_v u_\beta^2/4$, where $\tau_v$ is a thermalization time, i.e. the time required by a vacancy to dissipate in the drift motion the thermal energy $kT_0$), a vacancy lifetime $\tau_\ell$ against sticking and segregation at defects or recombination with interstitials, and a generation term at the s/l interface described by an interface generation velocity $u_\gamma$. The model also applies to the vacancy motion into the feed line plug with the s/l interface replaced by the plug/source chamber interface, the source chamber acting as a large vacancy reservoir. Solving the kinetic equation in its simplest form, where a constant pressure gradient is assumed, the oscillating part of the flow has the form [32]

$$j_{\text{osc}}(t) = \frac{1}{2} v_0 u_\gamma \left[ \frac{e^{-\beta y}}{\sqrt{\pi y}} + 2e^{-\gamma y} \sqrt{y} + \frac{\alpha}{\sqrt{\beta}} \text{erf}{\sqrt{\beta y}} \right],$$

(5)

where $y \equiv t/\tau_v$, $\beta \equiv 1 + \tau_\gamma/\tau_\ell$, $\alpha \equiv u_\gamma/u_\ell$ and $v_0$ is the initial collapse-induced depletion of the vacancy concentration. In the limit of a large pressure gradient, where vacancies generated at the s/l interface are swept away into the solid, and diffusion and recombination terms may be neglected, continuity requires $u_\ell = u_\gamma$ ($\alpha = 1$). With these approximations the geyser signal shape depends on the single parameter $\beta - 1 = \tau_\ell/\tau_\gamma$. However, in fitting the experimental signals with equation (5) and a suitable $\tau_\ell/\tau_\gamma$, the fitting interval $0 < y < y_0 \equiv \tau_0/\tau_v$ also has to be chosen, which allows us to obtain the thermalization and recombination times, $\tau_v$ and $\tau_\ell$. Despite the model approximations, mostly affecting the short-time part of the signal, the variety of geyser shapes displayed in figures 2 and 6 can be fairly well reproduced by equation (5) as illustrated in figure 8. The fitted times $\tau_v$ and $\tau_\ell$, which are listed in table 1, range between half a minute to several minutes, $\tau_v$ being comparable to $\tau_0$ and $\tau_\ell$ an order of magnitude smaller.

The large change in the pulse shape seen in figure 2 from pure (long flat tail) to mixed solid (triangular tail) with no substantial change in the period and a modest change of the parameters indicates a delicate balance between diffusive and drift dynamics. A similar change in shape was also found for solid $^4$He at $P_0 = 32$ bar and $T_0 = 1.64$ K with the addition of just 1% $^3$He [4]. Perhaps even more surprising is the large change of the pulse shape in $^4$He + 1% $^3$He at $P_0 = 32$ bar with just a slight decrease of $T_0$ from 1.64 to 1.52 K, which is accompanied by almost a doubling of the period [4]. This observation was attributed to the sudden onset of trapping of vacancies near the $^3$He sites with a binding energy of about 1.6 K.

9 A systematic two-parameter fitting of pure $^4$He geyser signals at different pressures and temperatures has been done by Käs [33].

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Figure 8. Calculated geyser signals over one period according to equation (5) with the parameters $\tau_v/\tau_s$ and $\tau_0/\tau_v$ which best reproduce the salient features of the experimental geyser pulse shapes shown in figures 2 and 6 for $^4\text{He}$ (a) and $^3\text{He}_{0.54}$-$^4\text{He}_{0.46}$ (b). The corresponding periods, vacancy collision time $\tau_v$ and recombination time $\tau_s$ are listed in table 1.

The comparison (table 1) between pure and mixed solids under similar conditions ($P_0 \sim 58$ bar, $T_0 \sim 1.9$ K) shows almost equal recombination times $\tau_r$, whereas the thermalization time for the mixed solid $\tau_v = 227$ s is substantially shorter than $\tau_v = 343$ s for pure $^4\text{He}$. This result is consistent with the intrinsic disorder of the mixed $^3\text{He}_{0.54}$-$^4\text{He}_{0.46}$ solid and with respect to pure $^4\text{He}$. It should be considered, however, that the recombination is the dominant effect, with a corresponding mean free path $u_v \tau_r$ of the order of $10^2$ $\mu$m. In case this corresponds to the grain size of the polycrystalline material, the vacancy recombination is actually a grain-boundary segregation leading to the growth of intergranular voids. This suggests that the structural collapse (unjamming) may be attributed to this mechanism.

In conclusion, the geyser effect known to occur in the vacuum expansion of solid $^4\text{He}$, either pure or lightly doped with $^3\text{He}$, is found to occur also for isotopic solid mixtures such as $^3\text{He}_{0.54}$-$^4\text{He}_{0.46}$. While the observed Poisson ratio is about the same in the mixed and pure solids, important differences occur in the pressure dependence of the geyser period and in the signal shape, which are here attributed to a change in vacancy kinetics. The absence in the solid alloy of flow anomalies of the kind observed in pure $^4\text{He}$ solid in the explored pressure and temperature range is entirely consistent with this interpretation.

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