

Frost Heave in Argon

D-M. Zhu,^{1,*} O. E. Vilches,¹ J. G. Dash,¹ B. Sing,^{1,†} and J. S. Wettlaufer^{2,1}

¹*Department of Physics, University of Washington, Seattle, Washington 98105-1560*

²*Applied Physics Laboratory, University of Washington, Seattle, Washington 98105-55640*

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The freezing of argon in silica powder is observed to generate bands of pure solid argon in the same manner as in the phenomenon of ice lens formation in the freezing of moist ground. A first principles dynamical theory describes the mechanism of lens formation by the thermomolecular pressure-driven flow of interfacially melted films at the lens-solid boundary.

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Frost heave is an unwelcome but common phenomenon occurring during the freezing of moist ground. During heaving, water migrates through partially frozen soil along thermal gradients to lower temperature, typically upward toward the surface. The inflow produces an internal pressure in the soil which can become sufficiently large to force the ground apart, where the water freezes into segregated solid ice *lenses* [1]. Heaving and the subsequent melting of the lenses in warmer weather can be destructive of roads, pipelines, and other engineered structures. Studies of frost heave began in the early years of the twentieth century [2] and have continued to the present day. Measures to mitigate its effects have been developed from years of experience and research, primarily by soil scientists and geengineers [3], but the fundamental mechanisms have remained obscure [4]. Recent studies have explored the molecular basis for the phenomenon and have provided detailed explanations of features previously treated empirically [5]. One conclusion that can be drawn from the modern work is that frost heave and lens formation can occur in other substances and are not unique attributes of H₂O, its expansion on freezing, and soil chemistry. Here we report an experimental verification of the prediction of this phenomenon using an inert substance, argon, which contracts on freezing, and discuss it in light of a new quantitative theory [6,7].

There are several basic elements required for lens formation: (i) a fusible substance in a porous medium of limited cohesive strength, (ii) temperatures within the perturbed range of solid-liquid coexistence of the substance, where the freezing point is depressed below the normal phase boundary, such as can be caused by interfacial melting, capillarity, and dissolved impurities [5,8], (iii) a temperature gradient, causing a *thermomolecular pressure* which drives the liquid toward regions of lower chemical potential [9], and (iv) interfacial melting at the interface between the solid lens and the porous medium, to allow the rejection of particles of the medium from the growing solid [5–8].

The existence of thermomolecular pressure in a material other than H₂O has been demonstrated in a sample of ⁴He contained in a cell filled with porous Vycor glass [10]. In that study, the tensile strength and connectivity of the

Vycor prevented cracking, so that segregated solid lenses could not form. In the present work we demonstrate the segregation and growth of large lenses of pure solid argon in a powder of monosized silica particles.

The apparatus consists of a glass walled flat cell providing a thin sample space containing a powder of 2.2 μm diameter silica spheres, as shown schematically in Fig. 1(a). The cell is cooled by partial immersion of a bottom fin in liquid N₂ and filled by condensation from a supply of high purity argon gas. Temperatures at the top of the cell are determined from the vapor pressure of liquid argon and at the bottom of the cell by a platinum thermometer. An internal “standpipe,” a vertical glass tube (6.3 \times 0.23 \times 0.3 cm) free of powder, serves as an indicator of the location of the freezing level in the cell. Visual observations are made by videotelescope and recorded on tape. In a typical run we observed the beginning of lens formation within a few minutes after the bottom of the cell was cooled below the triple point of argon, 83.6 K. The start was seen by the sudden appearance of a narrow horizontal gap in the powder parallel to an isotherm. Depending on the temperature gradient, the gap would grow as a single lens for a relatively constant gradient or the single lens would soon cease growing and be followed by a succession of thin lenses higher in the cell following a sudden increase in the gradient caused by raising the liquid N₂ level. In a constant and relatively mild gradient the single lens would grow steadily, being fed by flow of liquid downward and lifting the mass of powder above. In Fig. 2 we display video frames of the cell during lens growth. Noteworthy features are the clarity of the solid and the clear demarcation of the powder limit at the top of the lens.

The location of the liquid and solid levels in the standpipe and the powder cell were digitized as a function of time. From these data the rates of growth of argon lenses under different temperature gradients were extracted. The maximum observed growth rate for this cell configuration for the first lens was 1.6 cm h⁻¹. Attempts at obtaining higher rates by increasing the temperature gradient always produced a rapid advance of the freezing front engulfing the powder particles and the eventual formation of another argon lens at a higher level in the cell. We interpret

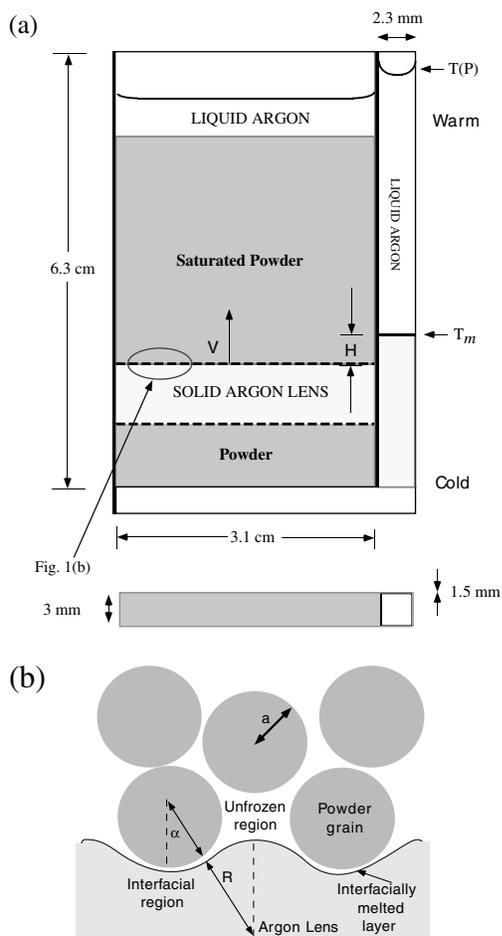


FIG. 1. (a) Schematic of the experimental cell. (b) A blowup of the area circled in (a). The liquid film at the particle-lens interface near two powder particles, where the solid/liquid interface has radius of curvature R . The inflowing premelted liquid applies the pressure, created by intermolecular forces, to lift the mass of powder [6].

observations of the lens growth within the framework of the theory outlined below.

A quantitative theory of lens growth was recently developed by Worster and Wettlaufer [6], aspects of which have been elaborated by Rempel and Worster [7] in the context of the rejection of particles by an advancing solidification front. We summarize the lens growth theory in the following paragraphs. Consider a pure substance filling the pores of a powder or porous matrix. The melting point in the pores is lowered by a combination of surface energy and curvature of the solid-liquid interface, a shift known as the Gibbs-Thomson effect. For an isotropic solid, the shift in the temperature T is $T_m - T = \frac{T_m \gamma}{\rho_s q} \mathcal{K}$, where T_m is the melting temperature of the bulk substance, γ is the solid-liquid interfacial energy, ρ_s is the solid density, q is the latent heat of fusion, and \mathcal{K} is the surface curvature, which is positive for convex interfaces seen from the liquid side. The geometry of the pores imposes a spectrum of freezing temperatures on the substance; hence there may be only partial freezing at temperatures and pressures within a range of the normal domain of bulk solid. As the tempera-

ture decreases the solid fraction grows within the pores. If the intermolecular interactions are such that the liquid phase wets the solid-particle boundary, a *premelted* film can exist at the particle-argon interface at temperatures below the normal bulk coexistence range [5,8]. For van der Waals interactions the thickness of an equilibrium film, d , on a planar surface is given by

$$d = \lambda \left(\frac{T_m - T}{T_m} \right)^{-1/3}, \quad (1)$$

where $\lambda^3 = A/6\pi\rho_s q$. The Hamaker constant A is an important microscopic parameter that describes the strength of the intermolecular adhesion forces between the grain, the film, and the lens.

A temperature gradient induces a gradient in the premelted film thickness, and the unfrozen liquid tends to flow toward regions of lower chemical potential, i.e., toward lower temperature. This tendency is expressible as a thermomolecular pressure difference, an excess effective pressure on the liquid over that on the solid, which is a function of the phase properties of the substance [5]. The most general form of the thermomolecular pressure gradient driving the flow, to first order in the temperature gradient ∇T , is given by

$$\nabla P_\ell = \rho_\ell q (\nabla T / T_m) + (\rho_\ell / \rho_s) \nabla P_s, \quad (2)$$

where P_ℓ (P_s) is the pressure in the liquid (solid).

When the inflow to a region raises the local pressure to a critical level to open a gap in the powder, the liquid therein begins to freeze, since its Gibbs-Thomson shift is reduced. In order for a pure solid lens to continue to grow it must lift the mass of powder by forces exerted on each grain at the lens boundary. These forces are applied through the liquid films interfacially melted at the solid-grain interface. Figure 1(b) portrays the geometry and conditions at the boundary. The profile of the solid-liquid interface is composed of two regions: an interfacially melted film between the lower portion of a grain and a solid-free region between the grains. The two regions have oppositely directed curvatures, which cause different temperature shifts, positive in the interfacial region and negative between the grains. The thickness d of the film in the interfacial region depends on the local conditions and the interactions between the liquid, the solid, and the grain. In the interfacial region the film thickness can be approximated by the planar relation when $d\mathcal{K} \ll 1$. In this case, relating the local temperature to the temperature gradient,

$$\left(\frac{\lambda}{d} \right)^3 = \frac{\nabla T H}{T_m} + \frac{2\gamma}{\rho_s q a}, \quad (3)$$

where H is defined as the distance of the center of a grain of radius a , adjacent to the lens, behind the T_m isotherm.

The flow of the film—the thermomolecular pressure opposed by the viscous resistance—balances the repulsive interaction between the grain and the lens and the overburden pressure due to the weight of powder above. Relating the thermomolecular pressure to the temperature gradient,

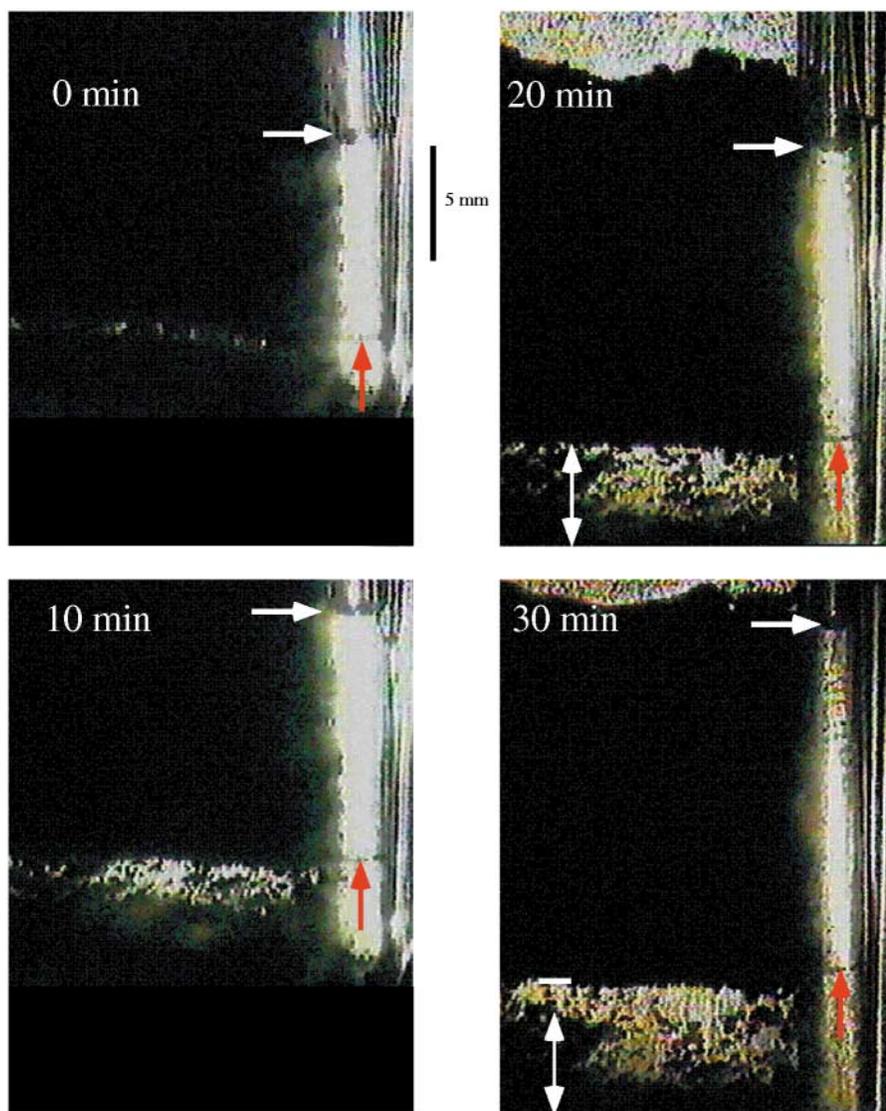


FIG. 2 (color). Video sequence of cell and lens growth over a period of 30 min viewed using transmitted light. The red (white) arrow distinguishes the solid-liquid (liquid-vapor) interface in the standpipe. The speed V of a lens is measured from its first appearance when the powder ruptures and the transparency of the solid argon allows light transmission. We use the observation of an approximately horizontal line in the cell parallel to the solid/liquid meniscus in the standpipe, and hence the T_m isotherm, to define the time origin. As time evolves the solid region grows by the transport, under the influence of a thermomolecular pressure gradient, of unfrozen liquid around the powder particles and into the warm side of the lens [see Fig. 1(b)]. Some powder is trapped between the lens and the cell wall which is seen as dark patches. After 20 min the powder surface in the cell becomes visible. The magnitude of the advance of the lens in the subsequent 10 min is the distance between the white double headed arrow and the white horizontal bar.

$$P_T = \frac{\rho_s q \nabla T}{T_m} (H + \Gamma), \quad (4)$$

where $\Gamma = (2\gamma T_m)/(\rho_s q a \nabla T)$. The total thermomolecular force is calculated by integrating along the contact profile. In the limit that the angle α is small the net viscous force is calculated to be

$$F_\mu = \frac{3\pi\mu a^4 V \nabla T}{2\lambda^3 T_m} (H + \Gamma) \alpha^4, \quad (5)$$

where μ is the dynamic viscosity and V is the speed of the lens [6].

In close packed powders and the conditions of a single growing lens the Gibbs-Thomson shift is greater than the undercooling $\Delta = H \nabla T$ at the lens interface; hence there is no solid within the powder, and the radius of curvature of the lens in the unfrozen region is much greater than the radius of a grain. The critical undercooling that is necessary for the thermomolecular pressure to overcome the overburden is then

$$\Delta_c = P_o^{1/3} \left(\frac{2\gamma}{a} \right)^{2/3} \frac{T_m}{\rho_s q}, \quad (6)$$

where P_o is the overburden pressure. The balance of forces leads to the equation for the lens speed

$$V = \frac{A}{9\pi\mu a^2} \frac{2\gamma}{a} \left(\frac{T_m}{\rho_s q} \right) \frac{\Delta^3 - \Delta_c^3}{\Delta^4}. \quad (7)$$

Figure 3 shows a family of curves of the behavior of lens growth rate versus the undercooling Δ , calculated for values of the Hamaker constant spanning the result of experiments on argon-graphite films [11]. The characteristic shape of each curve is a rapid rise above the threshold Δ_c to a maximum, and then a more gradual decline of rate at higher undercooling. The initial rise is caused by the increasing temperature gradient and consequently greater thermomolecular pressure. But as the temperature decreases the interfacial film thickness falls, which increases the flow resistance. This competes with and

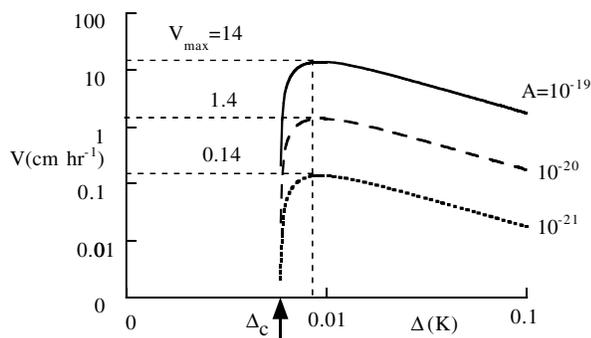


FIG. 3. Theoretical growth speed versus undercooling at the lens front, for three values of the Hamaker constant shown in the figure. For a given Hamaker constant, a curve represents the possible steady growth speed V for a single argon lens as a function of the undercooling $\Delta = H\nabla T$. A minimum undercooling Δ_c is required in order for the thermomolecular pressure to overcome the overburden as described by Eq. (7). There are no steady solutions with a growth rate greater than V_{\max} (see text).

eventually dominates the trend due to thermomolecular pressure, so that the growth rate reaches a maximum,

$$V_{\max} = \frac{A}{3\pi a^2 \mu 4^{4/3}} \left(\frac{2\gamma_{sl}}{P_O a} \right)^{1/3}, \quad (8)$$

and then falls. Above the maximum, as the undercooling exceeds the Gibbs-Thomson shift, the solid begins to invade the powder region. When this begins the flow is choked off, and growth stops. Then if the gradient is sufficiently large, a new lens will begin some distance away at a warmer location, and the process continues. In this way under natural conditions a succession of thin lenses is sometimes produced in freezing soils [1].

Precise mapping of the stable branch of the curve is hindered by difficulties owing to the cell dimensions and in maintaining a steady state temperature gradient in the liquid N_2 cooled system. Although the quasi-two-dimensional design enables continued visual observation of the freezing process, it also produces considerable frictional resistance to the heave. Thus the effective overburden, which in field and in large scale laboratory studies of ice frost heave is simply the weight of the soil or powder above the lens, is presently a composite of the weight and the wall friction. Evidence for the frictional resistance is the fact that at the end of an experiment the powder is found to be strongly compressed. The frictional term appears to be strongly dominant, for we estimate that the weight of powder alone would produce a critical threshold $\Delta_c = 0.01$ K with a typical thickness of powder 2 cm above the lens, whereas the experimental thresholds were more than an order of magnitude greater.

However, the experiment provides qualitative confirmation of theory in several aspects. It demonstrates solid lens formation in a material other than H_2O . It shows that a lens appears when the supercooling exceeds a certain threshold and that the lens will continue to grow under a mild tem-

perature gradient. When the gradient is larger the growth is halted and a second lens begins to form. There is a maximum growth rate of the primary lens (observed to be 1.6 cm h^{-1} , within the range of Fig. 3) which should determine the ratio of the Hamaker constant to the effective overburden pressure [see Eq. (8)]. We conclude that the experiment follows the qualitative pattern described by the theory and exhibited in freezing soils.

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*Permanent address: Department of Physics, University of Missouri, Kansas City, MO 64110.

†Visiting student from Institute of Physics, University of Tübingen, 72076 Tübingen, Germany.

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