



Microgravity and Transfers/Interfaces and phase change, two-phase flow

## The Bjerknes instability during crystal nucleation by acoustic waves

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### Abstract

The instability of position of a growing spherical crystal in an acoustic field is studied. Due to the Bjerknes force, a spherical crystal, whose position is shifted from an antinode of pressure, moves in the acoustic field. This displacement, stable in the case of bubbles in a cavitation experiment, turns out to be unstable in the case of crystallization. This effect is studied for an arbitrary Atwood number. **To cite this article:** *M. Ben Amar, C. R. Mecanique 332 (2004).*

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### Résumé

**L'instabilité de Bjerknes au cours de la nucléation d'un cristal par onde acoustique.** On étudie ici l'instabilité de position d'un cristal sphérique dans un champ acoustique. Etant donné la force de Bjerknes, un cristal dont la position se trouve décalée par rapport à un ventre de pression se déplace dans le champ acoustique. Ce déplacement, stable dans le cas des bulles de cavitation, s'avère instable en cristallisation. Cet effet est étudié en fonction du nombre d'Atwood. **Pour citer cet article :** *M. Ben Amar, C. R. Mecanique 332 (2004).*

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### 1. Introduction

Recently, Chavanne et al. [1] have shown that noticeable overpressure in a liquid can trigger the production of a solid particle. The experiment was realized in a bath of Helium 4, at very low temperature (65 mK), at a pressure set to the solid–liquid melting pressure. A piezo-electric hemi-spherical generator produces an oscillating acoustic micro-wave which converges on an observation window. A sufficiently large over pressure allows the growth of a crystal, which eventually remelts when the over-pressure becomes negative. A model of the growth dynamics of the solid has been proposed in a previous publication [2]. The numerical results, although qualitatively in agreement with the experiment, do not recover the observation of Chavanne et al. of a solid which remelts about 100 ns

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before the pressure becomes lower than the equilibrium pressure. One possible explanation for this early collapse may be instabilities occurring during the growth of the nucleus, which may change its shape or its position in the acoustic field. A good candidate for shape deformation may be the Rayleigh–Taylor instability, which is expected to act during the growth phase, since the solid phase is more dense than the liquid [3]. In bubble cavitation, it is the contrary; the Rayleigh–Taylor instability occurs during the decrease of the bubble size, and especially at its collapse [3]. In this Note, our aim is to study an instability of position in the acoustic field. The question is the following: assuming that the crystal is not exactly at an antinode of pressure, does it try to move in the direction of this antinode? If this position is stable (resp. unstable), it will remain in (resp. leave) the vicinity of its initial position. Since the experimentalists detect the growth of the crystal, via density measurements at a fixed position, the answer has some importance. Anyway, the calculation can be made only for homogeneous nucleation where the base state is a spherical crystal. The transposition to an inhomogeneous nucleation process is not straightforward, since the crystal may remain stuck to the glass plate. However, it may be a lead into the understanding of the precocious collapse found experimentally. The goal of this Note is to focus on this instability, as a function of the Atwood number, or density contrast, which is small in the case of crystallization but of order one for bubble nucleation.

## 2. Interfacial laws for a spherical crystal

Crystal growth is governed by mass, momentum and energy conservation. These conditions have been listed explicitly in [2]. To simplify the analysis, we restrict our attention to the hard-sphere limit: we neglect waves in the solid, assuming the Young modulus infinite. In this case, the fluid velocity  $V_f$  is simply related to the growth rate of the crystal  $\dot{R}$ :  $V_f = -\beta \dot{R}$ , with the Atwood number given by  $\beta = (\rho_s - \rho_f)/\rho_f$ . In the Helium case, the density of the solid  $\rho_s$  is slightly larger than the density of the liquid  $\rho_f$  and the Atwood number is of order 0.1. The momentum conservation is then

$$P_s + \rho_s \dot{R}^2 = \rho_f (V_f - \dot{R})^2 + P_f + \frac{2\gamma}{R} \quad (1)$$

where  $P_s$ , and  $P_f$  mean the pressure in the solid and the fluid phases, respectively,  $\gamma$  being the surface tension. Since the exchange of heat can be neglected in the case of Helium (no measurable latent heat, infinite Peclet number, ...), it has been shown [2,4,5] that the Gibbs–Thomson relation [6,7] maintains the averaged pressure at the interface:

$$(P_s + P_f) - 2P_0 = \frac{2\gamma}{R} \frac{\rho_s + \rho_f}{\rho_s - \rho_f} \quad (2)$$

where  $P_0$  is the equilibrium pressure of the planar front, estimated to be around 25 bars. Hereafter, it will be chosen as our reference pressure. We do not consider here the kinetic effects which are also very weak for Helium [8].

The fluid velocity  $V_f$  satisfies the Euler equation (no viscosity for a superfluid) so the velocity potential (defined by  $V_f = \nabla \Phi_f$ ) satisfies the wave equation which has a general solution

$$\Phi_f = -\frac{F(t - r/c_f)}{r} + \Phi_0 \frac{\sin(kr)}{kr} \cos(kc_f t) \quad (3)$$

when the spherical symmetry is assumed.  $F$  is an arbitrary function,  $\Phi_0$  represents the amplitude of the imposed acoustic field  $\Phi_\infty(r, t)$ , assumed spherical, given the geometry of the ultrasonic generator. In the close vicinity of the crystal, since the velocities in the liquid are much smaller than the sound velocity, one can expand  $F(t - r/c_f)$  at small  $r$  near the crystal. The velocity in the liquid then becomes:

$$V_f = \nabla \Phi_f = \frac{F(t)}{R(t)^2} \quad (4)$$

This means that we reduce the wave equation to the Laplace equation, an approximation which is usually made in sonoluminescence and bubble cavitation. In the case of Helium, the maximum growth rate is estimated to be one

third of the sound speed, which gives for the fluid velocity  $V_f \sim \beta c_f/3$  and justifies the Laplace approximation since  $\beta \sim 0.1$ . A more precise treatment can be found in [9]. The background velocity is negligible as soon as the crystal radius is small.

One determines the pressure thanks to the Bernoulli equation:

$$P_f = \rho_f \left( \frac{F'(t)}{r} - \frac{1}{2} V_f^2 \right) + P_0 \frac{\sin(kr)}{kr} \sin(kc_f t) \tag{5}$$

with  $P_0 = \rho_f \Phi_0(kc_f)$ .

### 3. Instability of position during nucleation

The growth of a crystal can be perturbed by various instabilities, among them the Rayleigh–Taylor instability which may be responsible for a shape instability, which breaks the spherical symmetry. Here, we want to study the instability of position of the nucleus in the acoustic wave. According to cavitation theory, bubbles subject to an acoustic standing wave field move to gather at pressure nodes or antinodes. In the case where they are not exactly at the correct position, these bubbles can execute erratic motions, sometimes with shape oscillations. In any case, on average over a large number of cycles, the averaged position of these bubbles is fixed, as shown numerically by [10], but their position can vary periodically, the period covering many acoustic cycles. In our case, the problem is slightly different since we are concerned with a transient regime. Indeed, in the experiment of [1], the nucleation of the crystal occurs after a few periods of the acoustic wave and the crystal grows then melts during the same period. So our problem is not exactly an averaged effect after a long time but an instantaneous motion which may be of interest if the focusing of the wave on the plate of the window is not perfect. The nucleus may move from the window, and it then escapes possible density measurements. The calculation is made in the frame of the crystal and not in the laboratory frame. So in this frame, the center position is fixed. Assuming that the crystal displacement is  $d$  along the  $x$ -axis, the Laplace velocity potential centered at the crystal is then

$$\Phi_f = -\frac{F(t)}{r} - \frac{1}{2} \hat{v}_f(t) R(t)^3 \frac{\cos \theta}{r^2} + \Phi_\infty(r, t) \tag{6}$$

We define the monopole of each quantity, for example, for  $\Phi_f$ :  $\hat{\phi}_f = -\frac{1}{2} \hat{v}_f(t) \frac{R(t)^3}{r^2}$ . It denotes the first coefficient in the Fourier series. So we deduce from mass conservation:

$$\hat{v}_f = \dot{d} \tag{7}$$

The Bernoulli equation must be written in the laboratory frame where the velocity potential is denoted  $\tilde{\Phi}_f$ . In the laboratory frame, Eq. (5) is transformed into:

$$\partial_t \tilde{\Phi}_f = \partial_t \Phi_f - \dot{d} \vec{e}_x \cdot \vec{\nabla} \Phi_f \tag{8}$$

which gives:

$$\begin{aligned} \partial_t \tilde{\Phi}_f = & - \left[ \frac{\dot{F}(t)}{R(t)} + \partial_t (\hat{v}_f(t) R(t)^3) \frac{\cos \theta}{2R^2} \right] - \dot{d} \cos \theta \left( \frac{F}{R^2} + \hat{v}_f \cos \theta \right) \\ & + \frac{1}{2} \dot{d} \hat{v}_f \sin^2 \theta - (kc_f) \Phi_0 \frac{\sin(kr)}{kr} \sin(kc_f t) \end{aligned} \tag{9}$$

We assume negligible the spatial derivative of the background pressure. Finally, we get:

$$\left[ \frac{\dot{F}(t)}{R(t)} + \partial_t (\hat{v}_f(t) R(t)^3) \frac{\cos \theta}{2R^2} + \dot{d} \cos \theta \frac{F(t)}{R(t)^2} + \frac{1}{2} \dot{d} \hat{v}_f (3 \cos^2 \theta - 1) \right]$$

$$\begin{aligned}
& -\frac{1}{2} \left[ \left( \frac{F}{R^2} \right)^2 + \hat{v}_f^2 \left( \cos^2 \theta + \frac{1}{4} \sin^2 \theta \right) + 2 \frac{F}{R^2} \hat{v}_f \cos \theta \right] \\
& + \frac{P_\infty(r, t) - \vec{x} \vec{f}_{b,x}}{\rho_f} + \frac{\beta}{2} \left( \frac{F}{R^2} - \dot{R} \right)^2 + \beta \left( \frac{F}{R^2} - \dot{R} \right) (\hat{v}_f - \dot{d}) \cos \theta = 2 \frac{\gamma}{R \beta \rho_f}
\end{aligned} \quad (10)$$

$\vec{x} \vec{f}_b$  is a local potential due to a force per unit volume acting at the crystal surface whose origin comes from the Bjerknes force. By definition, the Bjerknes force is  $\vec{F}_b = - \int P_\infty \vec{n} dS$ , the surface  $S$  being the crystal surface. We assume a spherical standing wave field:

$$P_\infty(r, t) = P_0 \sin(kr)/(kr) \sin(kc_f t)$$

with  $r \sim (R + d \cos \theta)$  at the surface of the crystal if the mismatch is small.

The force along the  $x$ -axis, to linear order in  $d$  is then:

$$F_{b,x} = - \int P_\infty \vec{e}_x \cdot \vec{n} dS = P_0 j_1(kR)(kd)(2\pi R^2) \sin(kc_f t) \int_0^\pi \cos^2 \theta \sin \theta d\theta \quad (11)$$

The quantity  $j_1(u)$  is the spherical Bessel function of first order, related to the spherical Bessel function of zero order  $j_0(u) = \sin(u)/u$  by the relation  $j_1(u) = -j_0'(u)$ . It gives a force per unit volume along the  $x$ -axis:

$$f_{b,x} = \frac{3F_{b,x}}{4\pi R^3} = P_0(k^2 d) \sin(kc_f t)/3$$

This calculation assumes that  $R \gg d$ , an approximation not valid at the nucleation threshold. An exact calculation gives:

$$f_{b,x} = -\frac{P_0}{(k^2 d R)^2} \frac{3}{8R} \sin(kc_f t) [M(U_+) - M(U_-)] \quad (12)$$

$M(U)$  being given by:

$$M(U) = k^2(R^2 + d^2) \cos U - U^2 \cos U + 2U \sin U + 2 \cos U \quad (13)$$

with  $U_+ = k(R + d)$  and  $U_- = k(R - d)$ .

As expected the Bjerknes force is unstable when the crystal is close to an antinode of pressure: the side of the crystal near the focus of the acoustic wave is submitted to a higher pressure than the other side, so the crystal is pushed and moves off the focus. Due to the principle of action and reaction, the crystal exerts a force on the flow. Since Bernoulli equation governs this flow, when the crystal is at rest, we assume that the density of force for the flow is  $-f_{bx}$  which gives a potential contribution  $\vec{x} \vec{f}_b$ .

We average the Bernoulli equation:  $\langle \sin^2 \theta \rangle = \frac{1}{2} \int_0^\pi \sin^2 \theta d\theta = 2 \langle \cos^2 \theta \rangle = 2/3$ . So we find that the Bernoulli relation is modified only by one contribution, which comes from the square of the fluid velocity at the crystal:  $V_f^2 = F^2/R^4 = (\beta \dot{R})^2$  is transformed into  $V_f^2 + \frac{1}{2} \hat{v}_f^2$ .

Isolating the contribution in  $\cos \theta$  and replacing  $F$  and  $\hat{v}_f$  by their values:  $F = -\beta R^2 \dot{R}$  and  $\hat{v}_f = \dot{d}$ , we derive an equation for  $d$ , the distance between the focus and the crystal:

$$\ddot{d}R + 3\dot{R}\dot{d} = 2RP_0 k^2 d \sin(kc_f t)/(3\rho_f) \quad (14)$$

#### 4. Discussion of the results and conclusion

We recover quite the same equation as for bubble cavitation. It is independent of the Atwood number. The difference comes only from realistic experimental considerations. During crystallization, the microwave is focussed on the glass plate and the crystal grows near the focus. Any mismatch of position of the center with the focus

increases during the crystallization process and can explain a displacement of the crystal in the microwave field. It can explain also the experimental observation of a density decrease when the crystal is always under pressure: perhaps the crystal moves in the acoustic field and so does not feel the available over-pressure. Moreover, this displacement decreases the kinetic energy of the flow. Anyway, it turns out that this effect is weak and is perhaps relevant for noticeable pressure values. Remember that the correct dimensionless parameter is  $P_0/(\rho_f c_f^2)$  which remains small in the Helium experiment: about 0.01. The effect is weak if we assume negligible the initial value of the crystal velocity  $\dot{d}$ , but may also be important depending on this value. It may thus be an explanation for discrepancies between experimental and theoretical results.

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