PHYSICAL REVIEW E **73**, 045301(R) (2006)

Oscillating droplets by decomposition on the spherical harmonics basis

S. Courty,* G. Lagubeau, and T. Tixier
Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, United Kingdom

(Received 17 September 2004; revised manuscript received 20 December 2005; published 6 April 2006)

In the framework of Rayleigh's description, we have investigated the eigenfrequencies of the capillary waves of a nonwetting droplet under forced oscillations (pointlike force). The theoretical model using the spherical harmonics $Y_{\ell,m}(\theta,\varphi)$ as a part of the solution of the Laplace equation, is in good agreement with the experimental results. This model can be generalized for all kinds of excitations with a sitting or a levitating droplet due to the decomposition of the excitation on the spherical harmonics basis. From this study, a different theoretical way of interpreting droplet bouncing is presented motivating a wide range of industrial applications.

DOI: 10.1103/PhysRevE.73.045301 PACS number(s): 47.55.D−, 62.10.+s, 68.08.−p

The beauty of water droplets has long been a source of inspiration for poets and they are still fascinating a large community of scientists even after a century of research [1]. The underlying physics of these objects of common appearance is extremely rich, from surface physics with nontrivial phenomena such as capillarity, wetting, or bouncing, to hydrodynamics. Recently, with the development of superhydrophobic surfaces [2,3] and the levitation of droplets by the Leidenfrost effect [4], the bouncing of the water droplet has been intensively studied by groups at Ecole Normale Supérieure [5] and at the Collège de France [6,7,9]. Quéré *et al.* have observed that a water droplet can bounce over 20 times with a restitution coefficient close to 0.9 throughout the trajectory [6]. In the limit of small deformations, it has been demonstrated that these "liquid balls" behave as a quasi-ideal spring, a "water spring." This elasticity is related to an efficient transfer between kinetic and surface energy during the drop deformation [7]. Finally, like Hertz [8], they have also measured an impact time τ between the drop and the surface of the order of 3 milliseconds for a millimeter-size droplet [9]. Besides the novelty and the fundamental aspects arising from this field of research, these experimental results have led to a wide range of practical applications from agricultural spray to ink-jet technology.

In this paper, we have developed a generalized theoretical model using the decomposition on the spherical harmonics basis to describe the eigenfrequencies of a spherical droplet (nonwetting) of an incompressible fluid. Droplets tend to assume their shape from a compromise between the action of surface tension (or capillary forces) and gravity. This compromise depends on the capillary length $\kappa^{-1}=\sqrt{\gamma/\rho g}$ which is obtained by equating the Laplace pressure γ/κ^{-1} and the hydrostatic pressure $\rho g\kappa^{-1}$ for a liquid of density ρ and with a surface tension γ. In the case of water (with a surface tension $\gamma\approx 72$ mN/m and a density $\rho\approx 1000$ kg/m^3), if the radius of the droplet R is larger than the capillary length ($\kappa^{-1}=2.7$ mm), gravity prevails over capillary forces [10]. The theoretical description for oscillations of a mass of liquid was treated first by Rayleigh [11]. In *Course of Theoretical Physics: Fluid Mechanics* [12], Landau gave a mathematical interpretation of the nature of free oscillations for a levitating spherical droplet under capillary forces by using spherical harmonics $Y_{\ell,m}(\theta,\varphi)$. In the framework of Rayleigh's description and Landau's formalism, we have developed a model for spherical droplet oscillations induced by a pointlike force. In the limit of small amplitudes, the capillary eigenmodes of a droplet, with a radius R, are obtained by deriving the Laplace equation for a surface slightly different from a sphere. This surface, defined by a function $r=r(\theta,\varphi)$, is given by $r=R+\zeta$, with ζ a small deviation, compared to $r\equiv R$ for a sphere (Fig. 1). The Laplace equation [13] is then obtained by determining the curvature Ω for a spherical surface $r=R+\zeta$, and gives the pressure difference $\Delta P=P-P_f=\gamma\Omega$:

$$\Delta P=\gamma\left\{\frac{2}{R}-\frac{2\zeta}{R^2}-\frac{1}{R^2}\left[\frac{1}{\sin^2\theta}\frac{\partial^2\zeta}{\partial\varphi^2}+\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\zeta}{\partial\theta}\right)\right]\right\}, \tag{1}$$

where P_f is the applied pressure and P is the internal pressure, related to the velocity potential ψ by $P=-\rho\partial\psi/\partial t$. The velocity potential must satisfy $\Delta\psi=0$ with a boundary condition at $r=R$. From a mathematical point of view, this is a typical boundary value problem where the solutions are nonelementary functions. In the case of a spherical droplet, where the pressure difference is due to the curvature, we consider the Dirichlet problem for the Laplace equation in a unit sphere. The velocity potential is $\Delta\psi=0$ for $R<1$ (inside

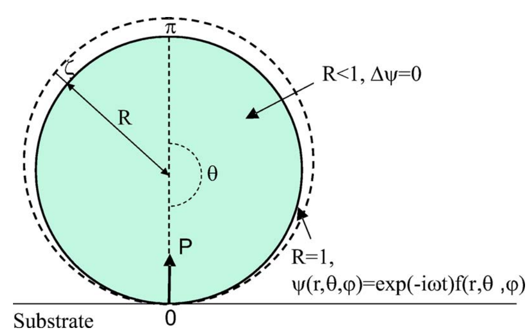

FIG. 1. (Color online) Problem of a Dirichlet for a droplet sitting on a substrate and deviating from its spherical shape.

*Author to whom correspondence should be addressed. Electronic address: sebastien.courty@lkb.ens.fr

the droplet, normalized by $1/R$) and $\psi(r,\theta,\varphi)=f(r,\theta,\varphi)$ for $R=1$ (Fig. 1). As usual, we first consider a solution of the Laplace equation in the form of a stationary wave $\psi(r,\theta,\varphi)=\exp(-i\omega t)f(r,\theta,\varphi)$. The function $f(r,\theta,\varphi)$ can be represented as a linear combination of volume spherical harmonics functions $r^{\ell}Y_{\ell,m}(\theta,\varphi)$, where $Y_{\ell,m}(\theta,\varphi)$ are the Legendre spherical harmonics, given by $Y_{\ell,m} = N_{\ell,m}P_{\ell}^{m}(\cos(\theta))\exp(im\varphi)$. The spherical harmonics, defined by the degree $\ell=k$ and the azimuthal order $m\epsilon[-k,k]$ with k an integer, are the restrictions to the sphere of a homogeneous harmonic polynomial, called the associated Legendre function $P_{\ell}^{m}(\cos\theta)\exp im\varphi$. The first factor in the spherical harmonics $Y_{\ell,m}(\theta,\varphi)$ expression, $N_{\ell,m}=\sqrt{(2\ell+1)(\ell-|m|)!/4\pi(\ell+|m|)!}$, is a normalization factor chosen to make the angular variables orthonormal. Then we seek a particular solution of the problem in the form $\psi=A\exp(-i\omega t)r^{\ell}Y_{\ell,m}(\theta,\varphi)$, where A is the oscillation amplitude. For a levitating sphere, the external pressure is constant. However, for a spherical droplet sitting on a substrate, the external pressure P_f is no longer constant. At the contact point ($\theta=0$) between the droplet and the substrate, the distribution of the applied pressure P_f must be a Dirac function that can be decomposed on the spherical harmonics basis,

$$P_f=\frac{F_c}{R^2}\sum_{\ell=2}^{\infty}\sum_{m=-\ell}^{m=\ell}N_{\ell,m}Y_{\ell,m}(\theta,\varphi), \tag{2}$$

where F_c is the force contact between the droplet and the substrate. As $P_f=-\rho\,\partial\psi/\partial t$ at $\theta=0$, we obtain $F_c/R^2 =[-\rho i\omega A\exp(-i\omega t)R^{\ell}]/N_{\ell,m}$. We substituted the pressure P_f [Eq. (2)] and ψ in the derivation of Eq. (1) with respect to time and with $\partial\zeta/\partial t=\partial\psi/\partial r$ (derivation described in *Course of Theoretical Physics: Fluid Mechanics* [12]) and used the fact that the spherical harmonics are eigenfunctions of the angular part of the Laplace operator L^2 that must satisfy

$$L^2[Y_{\ell,m}(\theta,\varphi)]+\ell(\ell+1)Y_{\ell,m}(\theta,\varphi)=0, \tag{3}$$

where $Y_{\ell,m}(\theta,\varphi)$ must be bounded for $0\leqslant\theta\leqslant\pi$, $0\leqslant\varphi\leqslant 2\pi$, and periodic in φ. Finally we obtain the dispersion relation that gives the eigenfrequencies of the forced capillary oscillations for a nonwetting droplet:

$$\omega_{\ell}^2=\frac{\gamma}{\rho R^3}\frac{\ell(\ell-1)(\ell+2)}{1+\sqrt{(2\ell+1)/4\pi}}. \tag{4}$$

Spherical harmonics $Y_{\ell,m}(\theta,\varphi)$ are very useful to physically understand the droplet oscillations. On the sphere, $\ell-|m|$ and $|m|$ correspond to the number of parallels (isolatitudes) and local meridians (isolongitudes) for which the harmonic function is null, respectively. Between these nodal circles, the function is alternatively positive or negative. In the case of a sitting spherical droplet, azimuthal orders $|m|$ are forbidden since the perturbation point coincides with a nodal line (a meridian which passes through the two poles) for all values of $|m|$. Then the dispersion relation (4) depends only on the degree ℓ, from which a wavelength $\lambda=2\pi R/\ell$ can be defined as ℓ corresponds to the total number of nodes on the sphere. If we compare the dispersion relation [Eq. (4)] with free droplet oscillations, a difference in oscillation times τ is observed. In the case of forced oscillations, the force, resulting from the applied pressure P_f, leads to a gain of the surface area due to the spherical harmonics and behaves consequently as a negative forced surface tension γ_f. This forced surface tension is given by defining an effective surface tension $\gamma_{eff}=\gamma/(1+N_{\ell,|m|})$ and we obtain $\gamma_f=\gamma_{eff}-\gamma =-\gamma N_{\ell,|m|}/(1+N_{\ell,|m|})$. As $\tau\approx\sqrt{\rho R^3/\gamma}$ and $\gamma_{eff}<\gamma$ the forced oscillations time is greater than the free oscillation time. By an analogy with a harmonic spring, this is similar to the case where the applied force on the spring is proportional to its displacement.

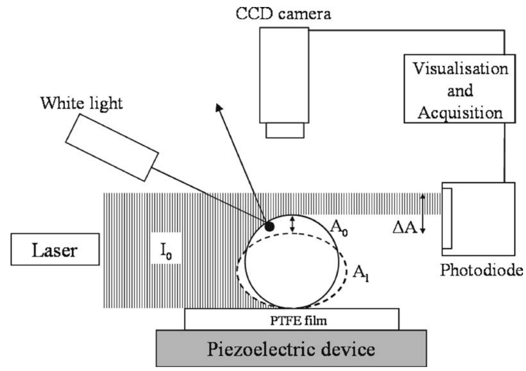

FIG. 2. Scheme of the optical apparatus for the detection of droplet eigenmodes. Forced capillary oscillations of mercury droplets are obtained with a piezoelectric substrate covered by a PTFE film to assure a contact angle θ_m close to 180°. Even eigenmodes $\ell=2k$ are determined from the frequency spectra. Odd eigenmodes $\ell=2k+1$ are detected by visualizing the trace of a reflection dot (black dot) on the surface of the mercury droplet.

The dispersion relation (4) has been tested by analyzing experimentally the dependence of ω with the degree ℓ of the spherical harmonics function. Since the degree ℓ corresponds to the droplet eigenmodes, they have been determined from the frequency spectrum obtained with a simple apparatus. The experiment, presented in Fig. 2, consists in inducing forced capillary oscillations of a spherical (nonwetting) droplet placed on a piezoelectric device covered by a PTFE film. To assure a nonwettability on the PTFE substrate, mercury droplets ($\rho=13450\ \mathrm{kg\,m^{-3}}$) of known radius R were used since they possess a high surface tension. With a dynamic contact angle apparatus (FTA200, First Ten Angstroms) both the contact angle θ_m and surface tension γ have been checked and we found $\theta_m=(175\pm5)°$ and $\gamma\approx 465\ \mathrm{mN/m}$. The droplet eigenmode oscillations were detected with an optical apparatus, as shown in Fig. 2. A laser diode mounted with a cylindrical lens was generating a vertical line larger than the diameter of the mercury droplet, and the incident light was collected into a high speed photodiode (from Radiospares). Since mercury droplets do not transmit the incident beam light, the intensity variation on the photodiode is directly related to the calibrated oscillation amplitudes ΔA of the droplet (the calibration of oscillation amplitudes has been realized by displacing vertically the droplet with a precision stage). The piezoelectric device (from Ceramitone, Radiospares) was supplied by a numerical function generator. The frequency spectra are obtained by applying a frequency sweep ranging from 1 Hz up to 2 kHz with a rate of

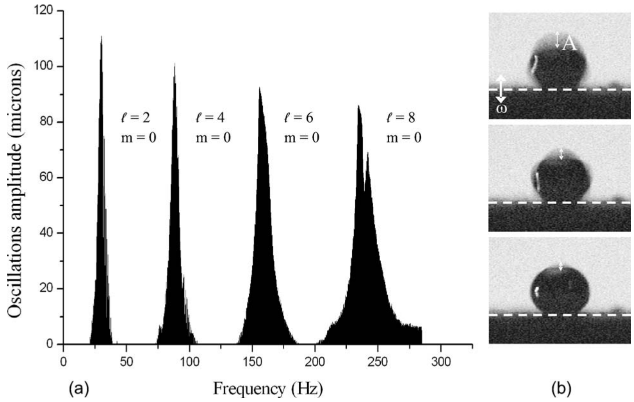

FIG. 3. (a) Frequency spectrum of a mercury droplet of radius $R=1.6$ mm showing the first eigenfrequencies for even values $\{\ell=2k,|m|=0\}$. (b) Photographs of vibration modes for mercury droplet.

$2\ \mathrm{Hz\,s^{-1}}$ and by using a high speed data acquisition (code written with Labview 7.0). Figure 3 shows the first eigenmodes for a mercury droplet of radius $R=1.6$ mm, as a function of the oscillating frequency. From the frequency spectrum, only even values of $\ell(\{\ell=2,|m|=0\},\{\ell=4,|m|=0\})$, $\{\ell=6,|m|=0\}$, and $\{\ell=8,|m|=0\}$ have been detected. We note here that odd values of degree $\ell=2k+1$ are not forbidden but their detections with the laser apparatus are not possible as the harmonic function presents an antisymmetric oscillation (alternates from a positive, at $\theta=0$, to a final negative value at $\theta=\pi$). However, the determination of odd eigenmodes has been possible by following the trace of a reflection dot on the surface droplet with a charge coupled device camera, as shown in Fig. 2. It appears for all $\ell=2k+1$ eigenfrequencies that the position of the gravity center is conserved. Conversely, for even values of ℓ, where the spherical harmonic presents a symmetric oscillation, we have a displacement of the gravity center with the forced oscillations. Here, the gravitational term does not affect the dispersion relation [Eq. (4)]. The normalized pulsation $\omega(\gamma/\rho R^3)^{-1/2}$ has been plotted in Fig. 4 as a function of ℓ for three sets of data with different radii R (circles, $R=1.0$ mm; triangles; $R=0.8$ mm; and squares, $R=0.5$ mm). The experimental data show a good agreement with Eq. (4), justifying the correction term $1+\sqrt{(2\ell+1)/4\pi}$ added to the Rayleigh formula. We noticed that a fit using Rayleigh's formula for a spherical and levitating droplet deviates from our experimental data. From these sets of data, we have also plotted ω as a function of R to determine the power law related to the radius R^n. For the three different droplet radii, we found a power law of $n\approx-\frac{3}{2}$, according to the first term of Eq. (4) (data not shown). Moreover, we have investigated the experimental gap between forced (frequency fixed at 35 Hz) and free oscillations (see for levitating liquid droplets [14] and for wetting drops [15–17]) of mercury droplet for the first eigenfrequency $\{\ell=2,|m|=0\}$ [Fig. 5(a)]. From the corresponding fast Fourier transform (FFT) [5(b)], we obtained a value for the ratio between the dispersion relation for forced oscillations [Eq. (4)] and free oscillations given by Rayleigh of 0.78 ± 0.02, which corresponds to the correction term $\sqrt{1/(1+\sqrt{5/4\pi})}$.

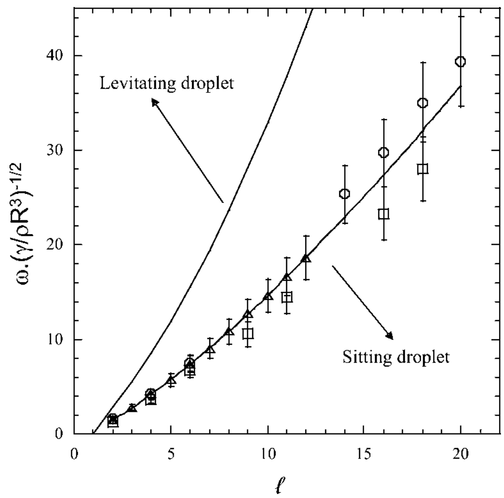

FIG. 4. Normalized pulsation ω as a function of the order ℓ for mercury droplets of radii $R=1.0$ mm (circles), $R=0.8$ mm (triangles), and $R=0.5$ mm (squares). The experimental data show a good agreement with Eq. (4). A fit using Rayleigh's formula for a levitated sphere deviates from our data.

We have extended our model of droplet oscillations under a pointlike force to the case of droplet bouncing, focusing, in particuiar, on the interesting problem of impact. This field of research has been put on the map recently as it leads to a wide range of industrial applications. In the literature, the experimental work of Richard *et al.* [9] has pointed out that the contact time of a nonwetting bouncing drop scales as the period of a drop vibration derived by Rayleigh, with a mea-

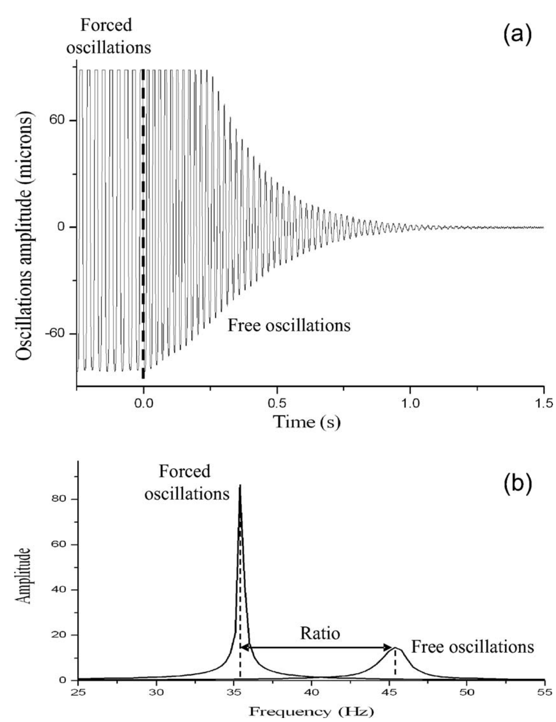

FIG. 5. Forced (frequency fixed at 35 Hz) and free oscillations of mercury droplets ($R=1.6$ mm) for the eigenfrequency $\{\ell=2,|m|=0\}$ (b) The corresponding FFT shows a ratio between forced and free oscillations of 0.78 ± 0.02.

sured experimental prefactor equal to 2.6±0.1. In the first approximation, we have considered that the bouncing can be described as one-half of the oscillation. Accordingly, the free oscillation prefactor, determined in the framework of Rayleigh, should be lower with a value of 1.65. Here, the decomposition on the spherical harmonics basis, as described in the first section of this paper, can be used to explain theoretically the difference in the prefactor values. However, in the case of a droplet bouncing, the oscillations are not provided by the oscillating substrate but by the transfer of the kinetic energy of the falling drop into an oscillating mode induced by a fixed surface. From this consideration, we have obtained three equations in the approximation of the two first spherical harmonics ($\ell=1$ and $\ell=2$) and small amplitude oscillations ($\zeta_\ell \ll R/\ell$):

$$N_1\zeta_1 + N_2\zeta_2 = 0, \tag{5}$$

$$\rho R\ddot{\zeta}_1 + F_c\frac{N_1}{R^2} = 0, \tag{6}$$

$$\rho R\frac{\ddot{\zeta}_2}{2} + \frac{4\gamma}{R^2}\zeta_2 + F_c\frac{N_2}{R^2} = 0, \tag{7}$$

where Eq. (5) represents the contact between the drop and the substrate surface, Eqs. (6) and (7) are the Rayleigh's equations applied to the translation (projection on $\ell=1$), and to the first harmonic oscillating mode (projection on $\ell=2$), respectively. From Eq. (6), we obtained the new expression of the contact force F_c,

$$F_c = -\rho R^3\left(\frac{\ddot{\zeta}_1}{N_1}\right). \tag{8}$$

The two eigenmodes composed by both translation and oscillation terms are obtained by solving the differential system composed by Eqs. (5) and (7). We found one trivial solution equal to zero and a theoretical prefactor with a value of 2.3. As a result, we have calculated a contact time close to the experimental value obtained by Richard *et al.* [9]. We strongly believed that the difference observed with the free oscillation prefactor is due to the coupling between the translation and oscillations modes. However, in order to obtain a complete description, this model has to be extended to more than two harmonic oscillating modes [18].

In summary, we have used the decomposition on the spherical harmonics basis to investigate theoretically forced and free oscillations of spherical droplets. Our model developed in the regime of capillary forces for droplet oscillations induced by a pointlike force is in good agreement with experimental results using mercury droplets. As the excitation can be decomposed on the basis of spherical harmonics, according to Eq. (2), this model can be generalized for all kinds of excitations of a sitting or levitating droplet. We have extended this model for the interesting case of droplet bouncing, in particular, for impact problems. In the approximation of the first harmonic oscillating mode, we have calculated a contact time longer than the experimental value of Richard *et al.* [9]. We strongly believed that the increase of the free oscillating prefactor is due to a coupling between translation and oscillation modes. However, in order to obtain a complete description, one needs to extend this model to more than two harmonic oscillating modes [18]. Another interesting perspective is to apply this model to the case of a partially wetting droplet under forced oscillations [19]. The wetting effect on droplet eigenfrequencies is a nontrivial phenomenon and this study is motivated by the range of applications in fields such as microfluidics, inkjet technology, and for dynamic contact angle and surface tension measurements.

We are grateful towards Sir S. Edwards and D. Quéré for their comments on the manuscript.

[1] A. M. Worthington, Proc. R. Soc. London **25**, 261 (1876).
[2] T. Onda, S. Shibuichi, N. Satoh, and K. Tsujiii, Langmuir **12**, 2125 (1996).
[3] J. Bico, C. Marzolin, and D. Quéré, Europhys. Lett. **47**, 220 (1999).
[4] A. L. Biance, C. Clanet, and D. Quéré, Phys. Fluids **15**, 1623 (2003).
[5] V. Bergeron, D. Bonn, J.-Y Martin, and L. Vovelle, Nature (London) **405**, 775 (1999).
[6] D. Richard and D. Quéré, Europhys. Lett. **50**, 769 (2000).
[7] K. Okumura, F. Chevy, D. Richard, D. Quéré, and C. Clanet, Europhys. Lett. **62**, 237 (2003).
[8] H. Z. Hertz, J. Reine Angew. Math. **92**, 156 (1881).
[9] D. Richard, C. Clanet, and D. Quéré, Nature (London) **417**, 811 (2002).
[10] P. G. de Gennes, F. Brochart-Wyart, and D. Quéré, *Gouttes, Bulles, Perles et Ondes* (Berlin, Paris, 2002).
[11] Lord Rayleigh, Proc. R. Soc. London **29**, 71 (1879).
[12] L. D. Landau and E. M. Lishitz, *Course of Theoretical Physics: Fluid Mechanics* (Pergamon Press, New York, 1959).
[13] P. S. de Laplace, *Oeuvres Complétes de Laplace* (Gauthier-Villars, Paris, 1912).
[14] M. Perez, Y. Brechet, M. Papoular, and M. Suery, Europhys. Lett. **47**, 189 (1999).
[15] C. Bisch, A. Lasek, and H. Rodot, J. Mec. Theor. Appl. **1**, 165 (1982).
[16] M. Strani and F. Sabeta, J. Fluid Mech. **295**, 263 (1995).
[17] D. W. DePaoli, T. C. Scott, and O. A. Basaran, Sep. Sci. Technol. **27**, 2071 (1992).
[18] G. Lagubeau and S. Courty (unpublished).
[19] S. Courty and G. Lagubeau (unpublished).