# **Rebounds in a Capillary Tube**

# David Quéré\* and Élie Raphaël

Laboratoire de Physique de la Matière Condensée, URA 792 du CNRS, Collège de France, 75231 Paris Cedex 05, France

### Jean-Yves Ollitrault

Service de Physique Théorique, CE Saclay, 91191 Gif-sur-Yvette Cedex, France

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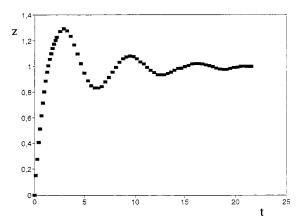
In experiments of capillary rise, oscillations of the liquid column can occur, as recently reported for liquids of low viscosity (*Europhys. Lett.* **1997**, *39*, 533). Here we analyze these oscillations (criterion of existence, shape, and damping). We start with an ideal fluid for which the oscillations are found to be indefinite and parabolic. Then, we take into account the dissipation at the entrance of the pipe: it is shown that it implies a different equation of motion depending on the direction of the liquid column (rise or fall). The first rise remains of parabolic shape, but a dissymmetry is found between this rise and the fall. Finally, the classical viscous friction inside the pipe is incorporated in the description and shown to accelerate the damping.

#### 1. Presentation

If at t = 0 a small tube of radius r is put in contact with a wetting liquid (of surface tension  $\gamma$ , density  $\rho$ , and viscosity  $\eta$ ), the liquid rises in the tube up to a height  $z_0$ , where the capillary force ( $F = 2\pi r \gamma$ ) balances the weight of the liquid column  $(\pi r^2 \rho g z_0$ , with g the gravitational acceleration). The dynamics of the rise is often dominated by the viscous friction of the liquid in the tube and described by Washburn's law:1 (i) at the beginning of the rise ( $z \ll z_0$ ), the meniscus position *z* increases with time as  $\sqrt{t}$ ; (ii) when approaching  $z_0$ , the column height zrelaxes exponentially.

We have recently observed with a high-speed camera the rise of liquids of small viscosities.<sup>2</sup> The following two points were observed: (i) The beginning of the rise is linear in time, which was explained by considering inertia; if balanced with the capillary force F, it yields a constant rising velocity  $c(c = (2\gamma/\rho r)^{1/2}$ , typically 20 cm/s, as observed experimentally). This regime has also been described in microgravity experiments by Dreyer et al. for liquids invading Hele-Shaw cells.  $^{3}$  (ii) Oscillations around  $z_{0}$  can occur if the viscosity is small enough. Such observations are reported in Figure 1, for ether ( $\gamma = 16.6$  mN/m,  $\rho = 710 \text{ kg/m}^3$ , and  $\eta = 0.3 \text{ mPa·s}$ ) rising in a glass tube of radius  $r = 689 \mu m$ . The meniscus position is plotted versus time, in dimensionless units: zhas been scaled by  $z_0$  and t by  $\tau = z_0/c$  ( $z_0$  and  $\tau$  are respectively 7.3 mm and 28 ms in the experiment). Oscillations are clearly visible, before damping due to the liquid viscosity and stopping at the height of capillary rise.

Here we try to understand these data. We start from an ideal liquid, for which it is shown that parabolic oscillations are expected. Then, we successively examine two causes of dissipation: energy loss at the entrance of the pipe and viscous friction inside it. A numerical integration of the equation of motion is performed, but we mainly stress some limiting cases (realistic from an



**Figure 1.** Capillary rise experiment for ether ( $\gamma = 16.6 \text{ mN/m}$ ,  $\rho = 710 \text{ kg/m}^3$ , and  $\eta = 0.3 \text{ mPa·s}$ ) rising in a glass tube of radius  $r = 689 \,\mu\text{m}$  (capillary rise height is  $z_0 = 7.3 \,\text{mm}$ ). The points are experimental data, obtained by a direct observation with a high-speed camera. The height z (scaled by  $z_0$ ) is plotted versus time t (scaled by  $\tau = z_0/c$ , where c is the inertial velocity of imbibition defined in the text; for ether in this tube, we have  $\tau = 28 \text{ ms}$ ).

experimental viewpoint) where analytical laws can be derived. We also propose a criterion for the existence of oscillations.

#### 2. Ideal Rebounds

The total energy of a liquid column of height z is the sum of its kinetic, gravitational, and surface energies. It can be expressed in the same dimensionless units as above:

$$E = \frac{1}{2}zz^2 + \frac{1}{2}z^2 - z \tag{1}$$

where  $\dot{z}$  denotes the meniscus velocity. At rest ( $\dot{z} = 0$ ), the minimum of E gives the equilibrium position, which is of course z = 1.

If the liquid viscosity is zero, E is conserved and its value is E=0 (since at t=0 we have z=0). Then, eq 1 can be integrated once. The dynamic law z(t) for the height of the meniscus is found to be parabolic:

<sup>(1)</sup> Washburn, E. W. Phys. Rev. 1921, 17, 273.

<sup>(2)</sup> Quéré, D. *Europhys. Lett.* **1997**, *39*, 533. (3) Dreyer M.; Delgado, A.; Rath H. J. *J. Colloid Interface Sci.* **1994**, 163, 158,

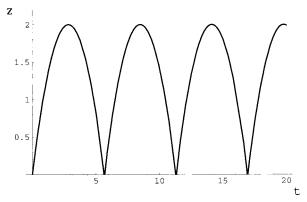


Figure 2. Rebounds in a capillary tube put in contact with an ideal fluid (no dissipation of any kind). The height z of the liquid column (scaled by  $z_0$ ) is plotted versus time t (scaled by  $z_0/c$ , where c is the inertial velocity of imbibition). Conservation of energy implies parabolic oscillations (eq 2) of height 2 and period  $4\sqrt{2}$  (about 5.6).

$$z(t) = \sqrt{2} t \left(1 - \frac{t}{4\sqrt{2}}\right) \qquad 0 \le t \le 4\sqrt{2}$$
 (2)

Then, at  $t = 4\sqrt{2}$ , the liquid column is in the same state as that at t = 0 (when it comes back to z = 0, it has no inertia, since it has no mass), so that it follows a similar parabolic trajectory. A wetting liquid put in contact with a tube indefinitely oscillates in this tube (transforming surface energy in kinetic and gravitational energies, and vice versa), as noticed by Tabor.4 These oscillations are parabolic, because of the nonconstant mass of the oscillator, and the height varies from 0 to 2 during half a period, which is  $T_{1/2} = 2\sqrt{2}$ . This ideal regime is plotted in Figure

The experiments indeed show oscillations (and during the first half period, z(t) is parabolic), but the maximal height is much lower than z = 2. Besides, after the first maximum is reached, the liquid column does not fall to zero. Even if the viscous friction along the tube does not play an important role in this first regime, we now show that another source of dissipation can explain these behaviors, while conserving the parabolic behavior close to t = 0.

### 3. Equations of Motion

Even for a liquid of negligible viscosity, some energy is dissipated because of the singular pressure loss<sup>5</sup> at the entrance (if the liquid rises) or at the exit (if it goes down). The reason is that there is an abrupt contraction between the reservoir and the pipe (in the experiment, the ratio of sections is of order 10<sup>-4</sup>), as noticed by Szekeley. 6 Then, it is well-known that balancing the rate of momentum with the force and writing the Bernoulli equation do not lead to the same result: considering that energy is conserved (the Bernoulli equation) underestimates the pressure drop. Physically, this irreversible loss of energy is due to eddies forming on the sides of the tube entrance.

If the ratio of the section of the tube over the section of the reservoir is close to zero, the pressure loss tends to a very simple expression which is (still in dimensionless units)5

$$\Delta P = \frac{1}{2} \dot{z}^2 \tag{3}$$

Thus, the energy dE lost for a displacement dz of the liquid column is simply equal to the kinetic energy of the liquid entering (or leaving) the tube. But the key point is the fact that this energy loss does not have the same expression when the liquid rises (dz > 0) or when it goes down (dz < 0). It is respectively written

$$dE = -\Delta P \, dz \tag{4a}$$

during the rise and

$$dE = \Delta P \, dz \tag{4b}$$

during the fall; thus, dE is negative in each case, as it must be. The latter equation is rather unusual in capillary "rise" experiments because, in order to apply it, it requires a downward motion of the liquid (or a fall), indeed observed in Figure 1 after the first maximum. Physically, eq 4b expresses the fact that the kinetic energy of the liquid column is lost in the (infinite) reservoir during the fall.

If viscous friction is present inside the tube, there is also a loss of energy for a displacement dz, which is classically given by the Poiseuille-Hagen law:

$$dE = -\Omega z \dot{z} dz \tag{5}$$

where  $\Omega$  is a dimensionless number proportional to the liquid viscosity  $\eta$  ( $\Omega = 8\sqrt{2}\eta\gamma^{1/2}r^{-5/2}\rho^{-3/2}g^{-1}$ ). Of course, eq 5 is valid whatever the direction the liquid column moves  $(dE \text{ is always negative, since } dz \text{ and } \dot{z} \text{ always have the}$ same sign). Finally, putting together eqs 1, 4, and 5 yields the equation of motion, which has a different expression depending on the direction of the liquid:

$$z\ddot{z} + \dot{z}^2 = 1 - z - \Omega z\dot{z}$$
 (dz > 0) (6a)

$$z\ddot{z} = 1 - z - \Omega z\dot{z} \qquad (dz < 0) \tag{6b}$$

## 4. Nearly Ideal Rebounds

**4.1. First Steps.** During the first steps after the contact, the parabolic Poiseuille-Hagen profile assumed to derive the friction term (eq 5) is not yet established. Thus, we shall consider  $\Omega = 0$  in all this section. This regime should last for the time necessary for setting-up the Poiseuille profile inside the pipe, that is for the boundary layer to diffuse on a length of order r, the pipe radius. This time dimensionally writes  $\tau^* \sim \rho r^2/\eta$ , and the numerical coefficient, evaluated by Schiller, is of order 0.115.8 Calculated for the data, it is about 130 ms (or 4.6 in dimensionless units), which indeed corresponds in Figure 1 to the time scale below which the column develops the first oscillation.

For  $\Omega = 0$ , the equations for the motion (eqs 6a and 6b) can be integrated once. We find

$$\frac{1}{2}z^2z^2 + \frac{1}{3}z^3 - \frac{1}{2}z^2 = A \tag{7a}$$

for the rise and

$$\frac{1}{2}\dot{z}^2 + z - \ln z = B$$
 (7b)

for the fall (A and B are two constants of integration).

<sup>(4)</sup> Tabor D. Gases, liquids and solids, 3rd ed.; Cambridge University Press: Cambridge, U.K., 1991. (5) Brun, E. A.; Martinot-Lagarde, A.; Mathieu, J. *Mécanique des* 

Fluides: Dunod, Paris, France, 1968.

(6) Szekeley, J.; Neumann, A. W.; Chuang, Y. K. *J. Colloid Interface* 

Sci. 1971, 35, 273.

<sup>(7)</sup> Goldstein, S. Modern Developments in Fluid Dynamics; Oxford University Press: Oxford, U.K., 1952.

<sup>(8)</sup> Schlichting, H. Boundary layer theory, McGraw-Hill: New York, 1968.

**4.2. Parabolic Rise.** If the meniscus position is z = 0at t = 0, A is fixed (A = 0) and the relation between the velocity and the position can be specified. In this particular case, it follows a parabolic law  $(z = \pm (1 - 2z/3)^{1/2})$ . Furthermore, the particular value A = 0 makes a second integration possible. A solution of remarkable simplicity is found for the position as a function of time:

$$z(t) = t\left(1 - \frac{t}{6}\right) \qquad 0 \le t \le 3 \tag{8}$$

As in eq 2, the first half oscillation is a parabola, of maximum height 1.5 (instead of 2) and duration 3 (instead of  $2\sqrt{2}$ ), in qualitative agreement with the data of Figure 1. The starting velocity is 1 (instead of  $\sqrt{2}$  in eq 2). A remarkable point is that the same result is obtained if inertia is balanced with the forces acting on the liquid column (the capillary force minus the weight).<sup>2</sup> It is written

$$\frac{\mathrm{d}(MV)}{\mathrm{d}t} = F - Mg \tag{9}$$

where V is the meniscus velocity ( $V = \dot{z}$ ) and M is the mass of the column, which is proportional to z. Introducing the same scaling for z and t as above, eq 9 reduces to eq 6a (with  $\Omega = 0$ ).

A question of interest is the way the liquid column "finds" the parabolic solution. When touching the liquid surface with a capillary tube, both the height and the velocities are zero, whereas eq 8 implies a velocity 1 for t = 0. Close to contact, both the viscous term and the weight (the z term in eq 6a) can be ignored, so that the equation of motion is

$$z\ddot{z} + \dot{z}^2 = 1 \tag{10}$$

Equation 10 has no solution satisfying  $z = \dot{z} = 0$  at t = 0, because of the singularity at z = 0. Close to the origin (z $\rightarrow$  0), the solution satisfying z(0) = 0 is z = t, as mentioned in section 1. On the other hand, if we look for a solution of zero initial velocity, we must introduce an initial position *a* at t = 0. The corresponding solution is  $z(t) = (a^2 + t^2)^{1/2}$ . In this case, the regime of constant velocity z=1 is reached in a time of order *a*, after a regime of pure acceleration: close to t = 0, we have  $z(t) \approx a(1 + t^2/2a^2)$ .

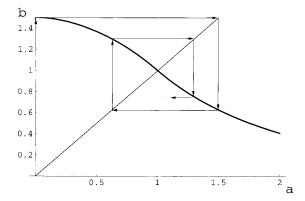
4.3. Oscillations. After reaching its maximum height z=1.5, the liquid column goes down following eq 7b. The solutions cannot be expressed analytically any more, and each portion of the trajectory must be calculated numerically. However, the extrema for zcan be obtained by simple recurrences. At these points, the velocity is zero, and we get from eqs 7

$$\frac{1}{3}z_{\min}^3 - \frac{1}{2}z_{\min}^2 = \frac{1}{3}z_{\max}^3 - \frac{1}{2}z_{\max}^2$$
 (11a)

for the rise and

$$z_{\min} - \ln z_{\min} = z_{\max} - \ln z_{\max}$$
 (11b)

for the fall. Starting from z = 0, we obtain as a series of extrema 1.5, 0.63, 1.30, 0.75, 1.21, 0.81, and so on. In Figure 3a, the extremal heights b are plotted as a function of the starting height a for each sequence of events (rise or fall). On each axis, z < 1 corresponds to the rise and z > 1 to the fall. In the same picture, a simple construction is presented, which allows a visualization of the column trajectory (and of the damping), starting from z = 0, as in the experiments. The damping due to the pressure loss



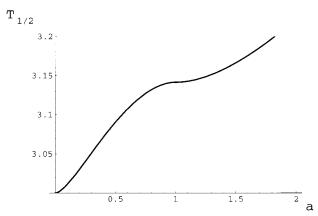


Figure 3. Nearly ideal rebounds (dissipation occurring only at the entrance of the tube), as a function of the initial position a: (a, top) Position b of the liquid column after half a period (i.e. a rise or a fall); the arrows indicate what happens versus time starting from a = 0. (b, bottom) Half the period of oscillation  $T_{1/2}$ . Both curves are drawn with the same dimensionless units as above and considering a zero initial velocity.

at the entrance (or exit) of the tube is rather soft: it is easy to show that the envelope of z(t) is a hyperbola, decreasing like  $3\pi/2t$ . For classical viscous damping, this envelope is an exponential, of short range, and thus possibly able to prevent oscillations from occurring, as usually observed.

The times corresponding to the successive rises and falls, or half-periods  $T_{1/2}$ , can also be calculated. They are plotted in Figure 3b as a function of the initial height a. For example, starting from z = 0, we find  $T_{1/2} = 3$ , as expressed in eq 8. It is the only value which can be calculated analytically, together with what happens close to the equilibrium value (z = 1). There we read in Figure 3b:  $T_{1/2} = \pi$ . For such small oscillations, eqs 7a and 7b can be linearized. Setting  $z = 1 + \epsilon$  (with  $\epsilon \ll 1$ ) yields  $\ddot{\epsilon} = -\epsilon$ . Thus, left close to equilibrium, the column oscillates sinusoidally with a period  $T = 2\pi$ .

Finally, the solution of eqs 7a and 7b can be calculated and drawn (Figure 4, with  $\Omega = 0$ ). All the different features stressed above (long-range damping, dissymmetry between the rise and the fall, existence of asymptotic analytical solutions for  $t \rightarrow 0$  and  $t \rightarrow \infty$ ) can be observed in this plot. But some discrepancy remains when comparing with the data in Figure 1, so that we we now have to treat the whole problem, including viscous friction inside the tube.

# 5. Viscous Fluids

5.1. Short-Time Behavior. Though eq 8 predicts the observed parabolic rise of the column, it overestimates the maximum height: 1.3 is observed in Figure 1 instead

**Figure 4.** Dynamics of the liquid column (same dimensionless coordinates as above), obtained by integrating eqs 6 (taking as initial conditions a=0.001 and z=0).  $\Omega=0$  is the so-called "nearly ideal" case (the viscous friction is neglected inside the tube).  $\Omega \geq 0$  corresponds to real fluids: then, the damping is quicker, because of the viscous friction inside the tube. The curve  $\Omega=0.3$  (thick line) closely agrees with the data of Figure 1;  $\Omega=2$  is the critical damping from which oscillations disappear.

of the predicted value 1.5. This difference can be interpreted as a local effect of the viscosity: the boundary layer develops from the contact line, so that viscous dissipation immediately takes place close to it, forcing the liquid wedge to join the solid with a nonzero dynamic contact angle  $\theta(z)$ . Thus, the driving force is smaller than that considered up to now: on the right-hand side of eq 10, 1 must be replaced by  $\cos \theta$ , providing a rising velocity close to the beginning of  $(\cos \theta)^{1/2}$  instead of 1.

Hoffman showed that  $\theta$  is determined by the capillary number (Ca =  $\eta/\gamma$ ). In the experiment, the starting velocity is c=23 cm/s, which gives Ca =  $2\times 10^{-3}$ . From Hoffman's curves,  $\theta$  can be evaluated to be around 30° (rather small because of the small viscosity of ether), from which the dynamics of the rise is deduced. Integrating eq 6a with an effective (and constant) driving force of  $\cos\theta$  (instead of 1) and still considering  $\Omega=0$ , we obtained a parabolic rise in close agreement with the data (in particular with a maximum at z=1.3). Hence, for  $t<\tau^*$ , it can be considered that the whole liquid column is inertial while viscosity only acts locally on the contact angle, imposing a lower starting velocity.

**5.2. Condition for Oscillations.** For  $t > \tau^*$ , viscosity acts on the whole column. The Poiseuille viscous force cannot be neglected any more and the whole eqs 6a and 6b must be considered. Depending on the value of  $\Omega$  (which is proportional to the liquid viscosity), different behaviors are expected; in particular, the case of a very large  $\Omega$ corresponds to Washburn's analysis. We first look for the condition for which oscillations are likely to be observed. Close to the threshold in  $\Omega$  and at the position where an oscillation appears, z can be linearized; setting  $z = 1 + \epsilon$  $(\epsilon \ll 1)$ , we find for both eqs 6a and 6b  $\ddot{\epsilon} + \Omega \dot{\epsilon} + \epsilon = 0$ , which leads to oscillations if  $\Omega \leq 2$ . In our experiment for example, this criterion is largely fulfilled, since we have  $\Omega \sim$  0.2. A dimensional condition on  $\Omega$  can also be derived by comparing the time  $\tau$  necessary for the column to pass the capillary rise height  $z_0$  with a velocity c ( $\tau \sim z_0/c$ ) with the time for setting a Poiseuille flow  $\tau^* \sim \rho r^2/\eta$ : oscillations are expected if  $\tau < \tau^*$ , hence for  $\Omega$  smaller than a number of order unity.

**5.3. Viscous Damping.** Equation 6 can be finally solved numerically, with the initial conditions specified above (z = a and z = 0). Solutions are drawn in Figure 4 for a = 0.001 and different values for  $\Omega$  (0, 0.3, and 2). For  $\Omega = 2$ , it is indeed observed that there is no oscillation (the most frequent situation in practice, valid for small pipes or large viscosities). If increasing  $\Omega$ , the curve would gradually tend toward Washburn behavior, with smaller and smaller inertial corrections. An estimate for these corrections was proposed by some authors to explain deviations from Washburn's law observed close to the origin (z = 0). <sup>6,10</sup> An idea for example was to consider the flow inside the reservoir which supplies the tube. The size of the concerned region is of order r (it is limited in space because the fluid velocity in the reservoir quickly falls with distance x from the tube entrance, as  $x^{-\frac{1}{2}}$ ). Thus, the correction related to this flow consists of adding a term of order  $r/z_0$  (0.09 in our experiment) to z, in the right member of eq 10.6 The numerical solution is found to be very close to the previous one, because of the modest role of the term z, in particular during the starting stage of the

In the case we are interested in here ( $\Omega$  < 2), oscillations indeed develop. Slight shifts of the half-period can be observed, a consequence of the dependence of  $T_{1/2}$  on the amplitude (Figure 3b). More quantitatively, the curve  $\Omega=0.3$  is found to be in excellent agreement with the data (for which we rather have  $\Omega=0.2$ ): both the position of the extrema and the visible dissymmetry between the rise and the fall are described. Thus, considering as sources of dissipation both the loss at the entrance of the pipe and the viscous friction inside it allows us to describe the data.

#### 6. Conclusion

We have interpreted our data on rebounds in capillary rise by considering the time  $\tau^*$  necessary to set up the Poiseuille profile in the tube.

(1) Before  $\tau^*$ , the liquid column behaves "nearly ideally". (a) During the rise, the position of the meniscus follows a parabolic law as a function of time which can be understood very simply (eq 8). As a little correction to this simple model, we took into account the existence of a dynamic contact angle at the liquid front, whose effect is to slow the liquid column. (b) The fall is not parabolic any longer because the equations of motion are not the same, depending on the column direction (eqs 6a and 6b). This dissymmetry comes from the fact that the pressure loss at the place where the tube meets the reservoir is quadratic in velocity. As a remarkable point, the equations for the motion can be integrated once, despite the existence of a source of dissipation: equations looking like equations of conservation are obtained (eqs 7a and 7b), from which all the "nearly ideal" oscillations can be described (for example, graphically).

(2) Above  $\tau^*$ , viscous friction along the liquid column must be considered. The threshold in viscosity above which oscillations disappear was calculated. Below this threshold, oscillations are damped (as seen from a numerical integration of eqs 6). The dissymmetry between the rise and the fall is still visible and softens as the oscillations are damped. Then, the motion stops.

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