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Dynamics of vibrating atomic force microscopy

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Abstract. An atomic force microscope which is operated in the oscillating mode is an example of an impact oscillator. The description of such dynamical systems can be reduced to a mathematical mapping, which displays a square-root singularity. A direct consequence of this property is the emergence of an infinite series of period-doubling bifurcations. This extremely characteristic phenomenon should be observed in atomic force microscopes. We consider an atomic force microscope in which the tip–substrate forces are modelled by a liquid-bridge interaction. By integrating the dynamical equations we show that the atomic force microscopy (AFM) dynamical behaviour has the same characteristic bifurcation scenario as the square-root map. We point to the remarkable role of the energy that is dissipated upon impact. We finally suggest ways to improve the operation of AFM.

1. Introduction

Atomic force microscopy (AFM) is often used in the vibrating or tapping mode [1] in order to cope with strong adhesive forces that may cause the tip to stick to the surface, or to prevent damage to soft surfaces. When the excursion of the oscillating cantilever becomes large enough for the tip to reach the surface, its dynamical state will change, for example, through a shift of its resonance frequency or a change of its amplitude. These changes are used to detect the proximity of the surface.

An atomic force microscope that is operated in the vibrating mode can be viewed as an impact oscillator. In its simplest guise, collisions in an impact oscillator are instantaneous velocity reflections off a hard wall. When no collisions occur, the impact oscillator is a driven harmonic oscillator, but it derives a strong nonlinearity from the collisions. Consequently, an impact oscillator displays a bewildering variety of periodic and chaotic states [2]. Although the tip–substrate interaction in AFM is far more complicated than an instantaneous velocity reflection, we believe that much can be learned from the dynamics of ideal impact oscillators. It is important to understand this dynamical behaviour in order to make more sensitive atomic force microscopes.

The analogy between impact oscillators and AFM was first exploited in experiments by Burnham *et al* [3]. The resonances found were identified as those predicted for impact oscillations, but information about the structure of the phase diagram was still scant. As we will argue, it is precisely the phase diagram that reveals telltale signs of impact oscillator dynamics. The phase diagram shows the boundaries of various dynamical behaviours in the

driving frequency–amplitude plane. A point in this diagram determines the dynamical state of the impact oscillator, which could be a periodic or chaotic state.

In non-contact vibrating AFM the focus has been on resonance shift as a way of detecting the proximity of the surface. Depending on the dynamical state, these shifts can vary more or less strongly with the mean tip–sample separation. In [4] an analytical study was performed for the behaviour of an oscillator in an attractive surface potential. Rather than changes of the resonance frequency, or changes of the oscillation amplitude, we will consider the much more drastic bifurcation of one dynamical state into another one as the surface is approached.

For impact oscillations a special situation arises when the collisions are with zero velocity, so-called *grazing impacts*. For impacts which are close to grazing it is possible to arrive at an extreme reduction of the mathematical description of the dynamics in the form of a Poincaré mapping. Such a mapping is a simple algebraic expression that evolves the dynamical state at discrete time t_n into the one at t_{n+1} . To understand grazing impact dynamics, it is then no longer necessary to integrate differential equations. This simplification enabled an almost exhaustive foray of the possible dynamical states of impact oscillators [5]. Fortunately, the reduction of grazing impact dynamics to iterates of a mapping is of particular relevance for AFM. Atomic force microscopes are preferably operated in the grazing regime where the atomic landscape is just felt by the vibrating tip.

Grazing impact mappings were first found by Nordmark [6] for instantaneous impacts with non-yielding walls, and were extended to yielding walls in [7][†]. Clearly,

[†] In a quite general context, the case of collisions with a yielding wall is also discussed in [8].

these simple interaction models are challenged in AFM. For example, when AFM is performed in air, the substrate is covered with a water film and the tip–substrate interaction is mediated by hydrodynamic forces. The question is whether the dynamics due to this more complicated interaction show the same characteristic phenomena as that of an ideal impact oscillator.

In order to answer this question we will consider a realistic impact model in the context of a continuous-time differential equation. Our work was inspired by Berg and Briggs [10], who discuss the dynamics of impact oscillators in the presence of a liquid-mediated interaction between the impacting tip and the surface. Whereas in [10] only a few points in the phase diagram were considered, we will concentrate on its overall structure and demonstrate that it survives in the presence of more complicated impact forces.

As the mapping embodies the essence of impact dynamics we will introduce it first in section 2. For small excitation amplitudes the phase diagram of the map is characterized by regions where stable periodic orbits exist that have a single impact per period and whose period length is a multiple M of the excitation period. These so-called *maximal periodic orbits* will be introduced in section 2.2. In section 2.3 we will introduce the differential equation on which the mapping is based, and extend it with a model for liquid-film-mediated interaction between tip and substrate. We will demonstrate the scaling of the differential equation and give the relation between the parameters of the equation and the parameters of the map. It appears that a single scale parameter, namely the distance d between the equilibrium position of the cantilever and the substrate, determines the relative size of the liquid-mediated impact forces. In section 5 we will compare the phase diagram of the realistic model for AFM with that of the ideal impact oscillator. We will compute the energy loss on impact and discuss the applicability of the restitution model. A possible improvement of AFM is presented in section 6.

2. Dynamical equations

2.1. Mapping

An impact oscillator is a two-degrees-of-freedom system that is described by the position $x(t)$ and velocity $\dot{x}(t)$. It is a periodically driven system, and to completely determine the state of the oscillator it suffices to specify the position and the velocity at stroboscopic times $t_n = n2\pi/\omega$, where ω is the driving frequency. If we relate x_n to the position at time t_n and y_n to the velocity, the dynamical state of the oscillator is given by the sequence $(x_n, y_n), (x_{n+1}, y_{n+1}), \dots^\dagger$. For grazing impacts the mapping which expresses the state at time $n+1$ in that at time n reads [7]

$$\begin{cases} x_{n+1} = \alpha x_n + y_n + \rho \\ y_{n+1} = -\gamma x_n \end{cases} \text{ if } x_n \leq 0 \quad (1)$$

$$\begin{cases} x_{n+1} = -c_1 \sqrt{x_n} + c_2 x_n + y_n + \rho \\ y_{n+1} = c_3 x_n \end{cases} \text{ if } x_n \geq 0. \quad (2)$$

[†] The relation between the map variables (x_n, y_n) and the state $(x(t_n), \dot{x}(t_n))$ involves a nonlinear transformation, the specific form of which is irrelevant for our current purposes [7].

In the mapping (1), (2) the line $x_n = 0$ separates impacting ($x_n > 0$) and non-impacting ($x_n < 0$) orbits. If no collision occurs around t_n , the linear dynamics of the harmonic oscillator pertains and the linear map (1) applies. If, instead, an orbit crosses the borderline $x_n = 0$, the nonlinear map (2) applies. In (1), (2) ρ plays the role of the driving amplitude, such that impacts first occur at $\rho = 0$ when ρ is increased from a negative value. For a harmonic oscillator, the parameters α, γ and $c_1, \dots, c_3$ are set by the driving frequency, the Q -factor of the oscillator and the restitution coefficient; they will be specified below. In the ideal impact oscillator, collisions at, say, time t_0 correspond to an instantaneous velocity reflection.

$$\dot{x}(t_0^+) = -r\dot{x}(t_0^-), \quad (3)$$

where the restitution coefficient $0 < r \leq 1$ allows for energy loss at impact.

Instead of integrating the differential equation, impacts can now be studied by iterating the mathematical prescription (1), (2). This has greatly facilitated the study of the possible dynamical states and their bifurcation properties [5]. A key feature of the impact map (2) is the presence of a square root. It implies that the Jacobian of the map, which gauges the stretching of the $(x, \dot{x})$ phase space, has a *square-root singularity* at $x = 0$. This singularity spawns the characteristic dynamical behaviour of grazing impact oscillators.

The map (1), (2) applies to any oscillator that crosses a boundary. Although the association of the parameters $(\alpha, \gamma, c_1, \dots, c_3)$ to those of the underlying differential equation is no longer simple when the oscillator is not harmonic, the square root remains. The derivation of the map (1), (2) rests on a sharp distinction between non-impacting and impacting orbits. A further assumption is that the energy loss upon impact is concentrated at $x = 0$ through the restitution rule (3). While these assumptions are reasonable for impacts in macroscopic systems, they are clearly challenged in AFM. For example, in AFM the energy loss is not concentrated at the impact instance, and impacts may not be instantaneous. The question then is whether vibrating-mode AFM still displays the characteristic features of the dynamics generated by (1), (2).

2.2. Maximal periodic orbits

At small driving amplitudes, the excursion of the impact oscillator will not be large enough to hit the wall and it displays the trivial period-1 non-impacting periodic orbit. When the driving amplitude ρ is increased, impacts will first occur at $\rho = 0$, and the dynamical state of the oscillator will change. As explained in [5, 9], a direct consequence of the square-root singularity is the emergence of a series of transitions of non-impacting period-1 orbits (p_1) to period M (p_M) orbits that have one impact for each M periods of the drive. These p_M orbits may survive to driving amplitudes $\rho < 0$ when the driving amplitude is subsequently smoothly decreased. Thus, they may show an apparent hysteresis. Since we view this *period-adding* $p_1 \leftrightarrow p_M$ sequence as highly characteristic for impact oscillators, our goal will be

Figure 1. Bifurcation diagram of the ideal impact oscillator with restitution coefficient $r = 0$, and parameter values $\alpha = 0.7412$, $\gamma = 0.6912$, $c_1 = 1$, $c_2 = 0.1611$ and $c_3 = 0.6476$ as a function of $\sigma = \rho/1.187$. The points are 100 iterates of the map (1), (2), taken at each value of the reduced driving amplitude σ . When the driving strength σ is started from a negative value and increased quasistatically, a period-1 orbit is encountered that does not impact. At $\sigma = \sigma_1 = 0$ a transition to a maximal period-3 orbit is seen. This state undergoes a secondary bifurcation at $\sigma = \sigma_2$ to a period-3 orbit which impacts twice in each period. Chaos is seen for $\sigma \in [0.089, 0.209]$. When σ is subsequently decreased, the maximal p_3 orbit persists to $\sigma = \sigma_3 (\approx -0.002)$.

to study its survival in the presence of more complicated impact forces, such as those found in AFM.

For a particular parameter setting, the transition $p_1 \rightarrow p_{M=3}$ is illustrated in figure 1 where, instead of the parameter ρ , we have used the scaled bifurcation parameter σ which will be specified below. The bifurcation diagram was computed from the mapping (1), (2) by first setting the bifurcation parameter σ , then iterating the map many times to let transients die, and then drawing the next 100 iterates. As the map is a stroboscopic picture, a single value of x_n denotes a period-1 orbit, and so on. For negative σ we encounter the trivial non-impacting period-1 orbit. First impacts occur when σ is quasistatically increased to $\sigma = \sigma_1 = 0$, and the non-impacting orbit gives way to a period-3 orbit that has one impact per period of the driving. When σ is increased further, the p_3 orbit is destroyed due to an additional impact at $\sigma = \sigma_2$. When the driving strength is subsequently lowered, the p_3 maximal periodic orbit is traced to $\sigma_3 < 0$, until it loses stability and yields to the non-impacting p_1 orbit.

The sketched bifurcation structure is characteristic for impact oscillators with instantaneous impacts. We proved [7] that it remains in the case of impacts with a resilient wall, but with the energy loss concentrated at impact. We will demonstrate that this dynamic structure is also preserved in the case of more complicated interactions.

2.3. Differential equation

In order to be able to specify the parameters used in (1), (2), we will now formulate the relevant continuous-

Figure 2. Oscillating AFM. The cantilever with mass m is driven with a harmonic force $F \cos(\omega t)$. Its deflection is x , its velocity $\dot{x}$. It can collide with the substrate at $x = 0$. In equilibrium (drawn with dashed curves), the separation between the tip and the surface is d . The tip interacts with the substrate through a liquid bridge. In our model the tip bounces elastically off the rigid surface when $D = 10^{-10}$ m. Impact energy loss is due to viscous damping of the liquid bridge.

time differential equation. This will also allow us to introduce the liquid-bridge model for the tip-substrate interaction.

The model that we have in mind is shown in figure 2. It consists of a cantilever with mass m , spring constant K and linear damping ν . It is driven sinusoidally with amplitude F and frequency ω . Denoting by x the position of the point of the spherical tip closest to the wall, the equation of motion is

$$m\ddot{x} + \nu\dot{x} + K(x + d) + I(x, \dot{x}) = F \cos(\omega t + \phi), \quad (4)$$

where $I(x, \dot{x})$ is the interaction with the substrate that is located at $x = 0$; overdots indicate time derivatives. The distance between the tip and the substrate is $D \equiv |x|$. In the ideal impact oscillator the infinite interaction force I results in instantaneous velocity reflections with restitution coefficient r . If we scale time t with the period $2\pi/\omega$ of the external driving and the deflection x with the equilibrium distance d to the substrate, then (4) becomes in dimensionless units

$$\ddot{x} + \tilde{\nu}\dot{x} + \Omega^2(x + 1) + \tilde{I}(x, \dot{x}) = \tilde{F} \cos(2\pi t + \phi), \quad (5)$$

where $\tilde{\nu} = 2\pi\nu/m\omega$, $\Omega^2 = 4\pi^2 K/m\omega^2$, $\tilde{F} = (4\pi^2/m\omega^2 d)F$ and

$$\tilde{I}(x, \dot{x}) = \frac{4\pi^2}{m\omega^2 d} I\left(dx, \frac{d\omega}{2\pi}\dot{x}\right). \quad (6)$$

The driving force can be further normalized with respect to the driving strength $\tilde{F}_g$ needed for grazing impact in the ideal case, $\tilde{F}_g = [(\Omega^2 - 4\pi^2)^2 + 4\pi^2\tilde{\nu}^2]^{1/2}$, and $\phi = \cos^{-1}[(\Omega^2 - 4\pi^2)/\tilde{F}_g]$. Accordingly, we will use $\sigma = (\tilde{F} - \tilde{F}_g)/\tilde{F}_g$ as the reduced driving strength. It is also convenient to introduce a normalized driving frequency ξ as $\xi = fT$, where $T = 2\pi/[\Omega^2 - (\nu/2)^2]^{1/2}$ is the period of the free damped oscillations of the oscillator and $f = \omega/2\pi$ is the driving frequency.

From the non-dimensionalization (6) we reach the important conclusion that for an impact force I that is *linear*

in the separation, the dynamical behaviour is *independent* of the equilibrium tip-sample distance d . In general, the dynamical state may change at impact when F reaches F_g . Impacts can either occur for large cantilever amplitudes and large separation d , or at small amplitudes and small d . If the impact force is linear, it not possible to distinguish these situations. Experiments in AFM, therefore, give only information about nonlinear impact forces. For such a *nonlinear* force, the separation d is a key parameter that determines its effect on the dynamics. For velocity-dependent impact forces, another important parameter is the ratio of the oscillation time to that of an intrinsic interaction time scale. In the case of liquid-bridge interactions, this time scale would be set by the viscous dissipation.

If the impact force is such that it results in instantaneous velocity reflection, the dynamics of the ideal impact oscillator is approximated by that of the mapping (1), (2). The dynamical state can then be found quickly by iterating the mapping. The operating point in parameter space is set by the values of α, γ that are completely determined by the Q -factor of the cantilever and the reduced driving frequency ξ as $\gamma = \exp(-\tilde{\nu}) = \exp(-4\pi/\xi Q(1 - Q^{-2})^{1/2})$ and $\alpha = 2\gamma^{1/2} \cos(2\pi/\xi)$, where $(1 - Q^{-2})^{1/2}$ is the ratio between the frequency of the freely swinging cantilever and its eigenfrequency $(K/m)^{1/2}$.

3. The phase diagram

Let us assume a Q -factor $Q = 40$, which is a typical value for an atomic force microscope operated in air, and choose a restitution coefficient $r = 0$ (which we will argue below). For these parameters, the phase diagram of the ideal impact oscillator is sketched in figure 3. It consists of tongues in which maximal periodic orbits p_M are stable with periods $M = 3, 4, \dots$. The tongues form a geometric progression with their period tending to infinity and their size shrinking to zero as we approach the point of linear resonance $\xi = 1$.

In a typical experiment the excitation frequency is set and the driving amplitude is varied to values large enough for impacts to occur and back again. Thus, the phase diagram is traversed at a fixed (reduced) driving frequency ξ , starting from a point $\sigma < 0$ to a positive value of σ and back again to $\sigma < 0$. An example of such a scan has already been shown in figure 1, where the used parameters $\alpha, \gamma, c_{1,\dots,3}$ correspond to the case $\xi = 0.85$ and $Q = 40$.

In an upward scan, first impacts occur at $\sigma = 0$, which is at $\xi = 0.85$ inside the $M = 3$ stability tongue. When σ is increased further, the p_3 orbit is destroyed at $\sigma = \sigma_2$ due to an additional impact. This is the upper boundary of the p_3 stability tongue. When the driving strength is subsequently lowered, the p_3 maximal periodic orbit is traced to $\sigma_3 < 0$. It loses stability and yields again to the non-impacting p_1 orbit. Obviously, the interval $[\sigma_3, \sigma_2]$ in figure 1 is the cross section of the $M = 3$ stability tongue at $\xi = 0.85$ in figure 3.

† The other parameters are $c_1 = \text{sgn}(P_{12})$, $c_2 = \alpha - 2(1+r)P_{22} + (1+r)^2 P_{22}^2$, $c_3 = (1+2r)\gamma - (1+r)^2 P_{22}^3$, where P_{12} and P_{22} are elements of the time-one propagator of the harmonic oscillator [9]: $P_{12} = \gamma^{1/2} \sin(2\pi/\xi)/(2\pi/\xi)$ and $P_{22} = \alpha/2 - \gamma^{1/2} \ln \gamma^{1/2} \sin(2\pi/\xi)/(2\pi/\xi)$. Finally, the relation between the bifurcation parameter ρ in (1), (2) and the driving strength σ is $\rho = \sigma(1 - \alpha + \gamma)/(8\pi^2(1+r)^2 P_{12}^2)$. All parameters are for the underdamped case $Q > 1$.

Figure 3. Phase diagram of the ideal impact oscillator (1), (2) with quality factor $Q = 2\Omega/\tilde{\nu} = 40$ and restitution coefficient $r = 0$. It was computed from the mapping (1), (2). The stability tongues of maximal periodic orbits p_M , $M = 3, \dots, 8$ are indicated. The insets show the $M = 3$ and 4 orbits in the continuous-time $(x, \dot{x})$ plane. The points $(\xi, \sigma) = (0.84, 0.01)$ and $(\xi, \sigma) = (0.88, 0.01)$, where these orbits were computed from the differential equation (4) are indicated.

The phase diagram of figure 3 is characteristic for impact oscillators with instantaneous impacts. We proved [7] that it remains in the case of impacts with a resilient wall with the energy loss concentrated at impact. We will now demonstrate that this dynamic structure is also preserved in the case of more complicated interactions.

4. Liquid-bridge impact force

The forces between objects on a mesoscopic scale are discussed extensively in [11]. They consist of forces induced by absorbed liquid films, and forces due to direct solid to solid interaction. For liquid-mediated forces the viscosity is a natural sink of impact energy and no restitution model needs to be considered. Solid to solid interactions may induce hysteretic stick and release phenomena [12]. As our focus is on dynamics, which may display hysteresis of its own, we will not consider the latter forces now.

Figure 4 shows the impact force $I(x, \dot{x})$ of the liquid-bridge model described by Berg and Briggs [10]. Following their paper, the impact force is the sum of several constituents, the most important of which is an attractive part due to the liquid-bridge capillarity, $I_{\text{cap}} = -4\pi R\gamma/(1 + D/2r_K)$ [11], where R is the tip radius and $D = |x|$ the distance between the tip and the surface, and a viscous dissipation that is proportional to the velocity $I_{\text{visc}} = -3\pi\eta R^2 \dot{D}/(2D)$. At very small tip-sample separations the van der Waals repulsion $I_{\text{vdW}} = BR/(180D^8)$ becomes important. At a separation $D = 10^{-10}$ m we assume that the tip has reached the rigid substrate under the absorbed liquid layer. The substrate is thought to reflect the velocity of the tip instantaneously and elastically.

Figure 4. Sum of impact force $I(x, \dot{x})$ and elastic force Kx acting on the cantilever. The cantilever displacement $x(t)$ traces a sinusoidal motion which just grazes the substrate. The upper curve corresponds to motion towards the surface, the lower curve corresponds to motion away from it. The difference between the two curves is due to the viscous damping in the liquid bridge. For clarity, the effect of viscosity was multiplied by a factor of 10^2 . Inset: blow-up of the region very close to the surface. At a tip–substrate separation $D = 10^{-10}$ m the impact force changes into an infinitely steep hard-wall interaction.

We used a tip radius of $R = 20$ nm; the other parameters were taken from [10]†. In order to non-dimensionalize the tip–surface interaction, the only additional information that we need is the stiffness K of the cantilever (0.32 N m^{-1}) and its free swinging frequency $(1 - Q^{-2})^{1/2}(K/m)^{1/2}$ (90 kHz).

Figure 4 was computed with the equilibrium distance between the substrate and the cantilever set at $d = 50$ nm. It has two curves: one corresponds to motion towards the substrate, the other one to motion away from it. They differ because of the viscous damping of the liquid bridge. In order to illustrate the viscous damping, we used a simple sinusoidal cantilever excursion with amplitude of 50 nm (which just grazes the substrate). The reduced frequency is $\xi = 0.85$. Of course, true impacting orbits are not sinusoidal and the damping will depend on the position in a much more complicated way. It must be emphasized that the used expressions for the hydrodynamic forces are only valid at small tip–substrate separations $D \ll R$. Still, we use them outside this regime. Because these forces decrease rapidly with increasing D , we believe that the error made is acceptable. Moreover, it is not understood in detail how liquid bridges break at large separation D .

In this model the impact energy loss is due to the deformation of the liquid bridge. It is seen in figure 4 that at equilibrium separations smaller than $d = 50$ the force acting on the cantilever is highly anharmonic. Naturally, the effects of the liquid-mediated tip–surface interaction become

† These parameters are surface tension $\gamma = 7.3 \times 10^{-2} \text{ N m}^{-2}$, Kelvin radius $r_K = 7.8 \times 10^{-10} \text{ m}$, dynamic viscosity $\eta = 10^{-3} \text{ N m}^{-2} \text{ s}$ and van der Waals constant $B = 10^{-79} \text{ J m}^{-2}$. Following [10], we also include an entropic contribution to the impact force $I_{\text{struct}} = A_0 \tau \exp(-(D-\delta)/\tau) \{1 - \exp(-a^2/(2R\tau))\}$ with $a = R \sin(\cos^{-1}(1-2r_K/R))$ and $A_0 = 40 \text{ N m}^{-1}$, $\delta = \tau = 6 \times 10^{-10} \text{ m}$.

Figure 5. Bifurcation diagram of an oscillating atomic force microscope that impacts through a liquid bridge with the surface. It was computed by integrating (4) using a cantilever quality factor $Q = 40$ and reduced driving frequency $\xi = 0.85$. Shown are stroboscopic points x taken at a fixed driving phase. At each (reduced) driving strength σ we first integrated over many periods of the excitation to let transients die out, and then drew the next 100 stroboscopic points. This figure should be compared to figure 1 of the ideal impact oscillator. At $\sigma = \sigma_3$ it displays for decreasing σ the same $p_3 \rightarrow p_1$ transition as the mapping (1), (2).

weaker with increasing d and for $d = \infty$ we reach the case of the ideal impact oscillator with $r = 1$. At small enough d , the capillary force I_{cap} becomes stronger than the elastic restoring force Kx , and the tip may stick to the surface. For the parameters used, the relative force minimum in figure 4 at $x \approx -4$ nm is at zero when $d = 22$ nm.

5. The phase diagram of AFM

In order to see the evolution of the phase diagram with the strength of the liquid-bridge interaction, we first show the dynamics at $d = 200$ nm where the effect of the liquid bridge is relatively weak and next study the case $d = 50$ nm which is more strongly disturbed.

In figure 5 we show the bifurcation diagram of the AFM that impacts through a liquid bridge with equilibrium separation $d = 200$ nm. A variety of periodic and chaotic states can be recognized in this diagram, which at first sight is completely different from that seen in figure 1 for the ideal impact oscillator. However, both diagrams show *the same* $p_3 \rightarrow p_1$ transition at small σ . Clearly, at small driving strength σ , when the orbit is closest to grazing, the ideal impact oscillator and the AFM that interacts through a liquid bridge share *the same* basic bifurcation structure.

This is further demonstrated in figure 6 which shows the corresponding phase diagram at $d = 200$ nm. It is strikingly similar to that of the ideal impact oscillator in figure 3 and shows the same geometrically converging series of period-adding transitions. In figure 6 the reduced driving amplitude $\tilde{\sigma} = \tilde{F}/\tilde{F}_g - 1$ is defined in terms of the driving strength $\tilde{F}_g$ where impacts first occur in the ideal impact oscillator. Naturally, the liquid bridge leads to an increase in the driving

Figure 6. Phase diagram of an oscillating atomic force microscope that impacts through a liquid bridge with the surface. It was computed from the differential equation (4). The equilibrium distance between tip and substrate is $d = 200$ nm. The stability tongues of maximal periodic orbits p_m are the grey regions. Full curve: reduced driving amplitude $\sigma = \sigma_1$ where impacts first occur.

amplitude where impacts first occur and the anharmonicity leads to a shift of the p_M stability tongues along the (reduced) frequency axis. The broad resonance-like behaviour of the (reduced) amplitude σ_1 where impacts first occur reflects the anharmonicity of the force acting on the tip.

The effect of the liquid bridge becomes more and more important as the equilibrium separation d decreases. Accordingly, the distortion of the phase diagram becomes more pronounced at smaller d . Figure 7 shows the phase diagram at $d = 50$. A similar geometric progression of period-adding transitions as in the ideal impact oscillator of figure 3 can be seen, but an intervening period-5 orbit appears out of place. The highest period found is $M = 6$, and it may be that the infinite series of period additions is truncated. It also appears that maximal periodic orbits p_M may yield to several different states through additional impacts at the upper boundary of the stability tongues. This is responsible for the ragged look of the $M = 2$ tongue.

In the presence of the liquid bridge, the instant of impact is not well defined, for example in figure 4, orbits with amplitudes >45 nm sense enough of the impact force that they could be viewed as ‘impacting’. Still, the bifurcation of the non-impacting p_1 orbit occurs first when the tip reaches the solid substrate under the liquid layer. At this point the cantilever–wall interaction changes discontinuously to hard and elastically repulsive. For an equilibrium distance $d = 50$ nm and a reduced driving frequency $\xi = 0.85$ excursions of 45 nm are reached at a reduced driving amplitude $\sigma = 0.21$, whereas the bifurcation in figure 7 occurs only at $\sigma = \sigma_1 = 0.38$.

In oscillating AFM, there are two sources of energy loss. One is through the linear damping of the cantilever beam as represented by the damping coefficient ν in (4); the other one is associated with impacts through the viscous substrate interaction. In the phase diagram in figure 3, the stability

Figure 7. Phase diagram of an oscillating atomic force microscope that impacts through a liquid bridge with the surface. It was computed from the differential equation (4). The equilibrium distance between tip and substrate is $d = 50$ nm. The stability tongues of maximal periodic orbits p_m are the grey regions. Full curve: reduced driving amplitude $\sigma = \sigma_1$ where impacts first occur. At the used equilibrium tip–substrate distance $d = 50$ nm the liquid-bridge interaction is strong (see also figure 4).

tongues do not depend sensitively on the linear damping ν , but their width is strongly determined by the energy loss that is associated with impacts. For perfectly elastic collisions their width even shrinks to zero. Comparison of the phase diagram of the ideal impact oscillator in figure 3 to that of the liquid-bridge dissipation is comparable to a restitution coefficient $r = 0$ in the first case (this is why we used $r = 0$ in figure 3).

For the ideal impact oscillator, the energy loss at each impact is

$$\frac{1}{2}(1 - r^2)v_{\text{in}}^2,$$

where v_{in} is the impact velocity. The average energy loss per cycle ΔE is the average $\langle \frac{1}{2}(1 - r^2)v_{\text{in}}^2 \rangle$ over many cycles. For the liquid-bridge model the average energy loss per cycle is

$$\Delta E = \left\langle \int_0^1 \dot{x} \tilde{I}(x, \dot{x}) dt \right\rangle.$$

In figure 8 the energy loss of the ideal impact oscillator is compared to that of the atomic force oscillator with the liquid-bridge model[†]. It is a surprising observation that the energy losses for these two cases differ by almost three orders of magnitude. Still, the widths of the p_M stability tongues are comparable. Clearly, the distributed dissipation of the liquid-bridge model has a much stronger effect on the dynamical state of the oscillator than a restitution loss which is concentrated at the instant of impact. The conclusion is that energy losses cannot be translated readily into an effective restitution coefficient, as was done in [10].

Impact energy loss through hydrodynamic interaction is but one loss mechanism. Another one is the excitation of

[†] As the map (1), (2) involves transformed variables, the energy loss ΔE was computed from the differential equation of the ideal impact oscillator.

Figure 8. Curve with $d = \infty$: average energy loss per cycle ΔE for the ideal impact oscillator with restitution coefficient $r = 0$. Curve with $d = 200$ nm, $10^3 \times \Delta E$ for the liquid-bridge atomic force oscillator. The energy loss has been computed for the dynamical states shown in figures 1, 5, respectively. The liquid-bridge energy loss is three orders of magnitude smaller than the restitution loss. The two branches of the hysteretic transitions can be clearly recognized in the two cases. The apparent noise in the curves is due to chaotic dynamics.

higher-order modes of the cantilever at impact, which can dissipate through the radiation of sound. For macroscopic impacting systems we have found this to be an important effect [9, 13], but we do not know how this works out at mesoscopic scales.

6. Improving the sensitivity of AFM

In conventional vibrating AFM, contact between tip and surface is detected through a change of the cantilever amplitude. We propose to use, instead, maximal periodic orbits as a more sensitive probe. These subharmonic resonances emerge at the driving strength $\sigma = \sigma_1$ where impacts first take place ($\sigma_1 = 0$ in the ideal impact oscillator). They could be detected readily by monitoring the frequency spectrum of cantilever excursions. For most excitation frequencies the $p_1 \rightarrow p_M$ transition looks like a subcritical bifurcation and the M th subharmonic peak that emerges in the frequency spectrum at $\sigma = \sigma_1$ will be sizable. It should be realized, however, that in this case the *backward* transition $p_M \rightarrow p_1$ to the period-1 state is hysteretic as it occurs at a value of $\sigma < \sigma_1$. Only at selected frequencies is the apparent hysteresis absent. In that case the transition looks like a forward bifurcation and the M th subharmonic peak in the spectrum will be zero at $\sigma = \sigma_1$. Because intrinsic nonlinearities in the detection system readily result in higher harmonics of the cantilever displacement signal, the possibility of measuring the surface proximity through *subharmonics* is an advantage.

In this paper we have considered direct excitation of the cantilever. A key experimental problem is a precise control of the cantilever oscillation amplitude (and phase). In weakly damped (high- Q) systems, the amplitude stability is

determined by the stability of the resonance frequency. This stability is often jeopardized by temperature drifts [9]. A completely equivalent experiment is to vibrate the substrate instead. It is readily appreciated that also in this case (4) applies, but with the effective forcing

$$Am\{(\omega^2 - K/m)^2 + (\omega v/m)^2\}^{1/2} \cos(\omega t + \phi + \tilde{\phi}), \quad (7)$$

with the phase $\tilde{\phi} = \tan^{-1}(\omega v/(K - m\omega^2))$ and where A is the amplitude of the driven substrate. The resonance factor $\{ \}^{1/2}$ in the effective forcing now compensates the resonance behaviour of the cantilever amplitude so that amplitude stability is no longer a problem. However, the only excitation of the cantilever is now through impacts, and the stability of the *phase* $\tilde{\phi}$ of the driving with respect to the phase of the cantilever becomes of crucial importance.

7. Conclusion

The liquid-bridge model provides one of many conceivable types of interaction in AFM. It is a continuous force everywhere except when the oscillating tip reaches the substrate. It is precisely at this point where the simple periodic state of the AFM bifurcates into another one. Such a transition is an example of a border-collision bifurcation [14]. It is a remarkable finding that the qualitative prediction of the map (1), (2) holds, even in the case of forces which vary rapidly near impact.

Another remarkable observation is that it is not the nonlinearity of the impact force which causes a bifurcation of the dynamical state of the AFM, but the mere impact. The nonlinearity causes only a distortion of the phase plane of the impact oscillator.

We do not believe that our general conclusion holds if attractive impact forces are very strong and lead to (temporary) sticking of the cantilever to the substrate. Several model forces are known that display such behaviour [11] and it may be worthwhile to study their effect on the generic bifurcation structure of vibrating AFM.

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