Nanosubharmonics: The Dynamics of Small Nonlinear Contacts

N. A. Burnham, A. J. Kulik, and G. Gremaud
Département de Physique, Ecole Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

G. A. D. Briggs
Department of Materials, Oxford University, Parks Road, Oxford OX1 3PH, United Kingdom
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We observed the generation of subharmonics and chaos in a nanometer-sized mechanical contact. To first order, the behavior matches that of macroscopic systems, with some intriguing secondary differences. As the occurrence of periodic behavior (subharmonics) is related to the coefficient of restitution, it may be possible to image local energy dissipation with nanometer resolution.

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Countless generations of children have performed empirical observations of bouncing balls, but it was not until early this century that Raman began to study systematically the height to which steels balls dropped on glass would rebound [1]. He used the term coefficient of restitution to describe the ratio of the relative velocities after and before impact. In materials that can undergo plastic deformation, the energy lost is directly related to the dynamic hardness, which may differ from the static hardness by as much as an order of magnitude [2]. Contemporary problems range from hitting baseballs [3] to ships striking against a quay [4]. Two new aspects are brought together in this paper. First, there is great interest in the behavior of nonlinear systems subject to periodic excitation, and there exists a well-developed mathematical apparatus to describe the response. Second, much attention has recently been placed upon studies of the mechanical behavior of contact between materials on a very small scale, which can be investigated experimentally using scanning probe microscopes. An important motivation is to find out how what happens on a small scale relates to what happens on a big scale, and in particular whether phenomena that are predicted by macroscopic continuum models can be found in very small contacts.

A seminal analysis of the dynamic problem has been given by Pippard [5]. He considered a loudspeaker laid on its back, with a lightly loaded pin held against it by means of a spring. The contact between the loudspeaker and the pin is characterized by infinite stiffness in compression and infinite compliance in tension; the contact can push but it cannot pull. The accelerating force due to the spring is taken as independent of deflection. When the loudspeaker is excited with a small amplitude at a given frequency, the pin remains in contact and moves with the same amplitude. If the amplitude is increased, the acceleration of the loudspeaker may be greater than the acceleration that the spring can impart to the pin, so that contact is lost. The motion will then consist of a series of impacts between the loudspeaker and the pin. It is still possible for the motion to be periodic if each impact occurs at the same phase and the velocity of the pin immediately following an impact is the reverse of its velocity immediately before the impact. The difference between the relative velocities after and before impact, described by a coefficient of restitution less than unity, is compensated by the upward velocity of the loudspeaker at the moment of impact. More than one cycle of the motion of the loudspeaker may occur between impacts, and so the motion of the pin can exhibit subharmonics of the exciting frequency, as depicted schematically in Fig. 1(a). A given periodic solution may be either stable or unstable against small variations in the time interval between impacts. If the coefficient of restitution is ε, the maximum acceleration of the loudspeaker is a_0 and the free acceleration of the pin is a, then a subharmonic of period n can be stably excited if

$$\frac{1-\varepsilon}{1+\varepsilon}\pi n < \frac{a_0}{a} < \frac{1-\varepsilon}{1+\varepsilon}\pi n\left[1+\left(\frac{1+\varepsilon^2}{1-\varepsilon^2}\frac{2}{\pi n}\right)^2\right]^{1/2}. \quad (1)$$

This analysis predicts a rich variety of behavior. In general, as the loudspeaker amplitude is increased successively higher period subharmonics may be excited, spaced by amplitudes for which no solution exists and at which the motion is aperiodic, i.e., chaotic. This is equivalent to moving up the y axis of Fig. 1(b) for a given value of ε. But for values of ε close to unity, corresponding to almost elastic impact, there may be several stable values of n.

This early description has been extended in two significant ways. Mehta and Luck have studied the time evolution of a bouncing ball on a vibrating platform. They furthered earlier work for $\varepsilon = 0$ [6] to the case $0 < \varepsilon < 1$ [7]. In this case a ball will come to rest after a finite time (albeit after an infinite number of bounces) [8]. Then the ball loses all memory of its previous history, just as it does when $\varepsilon = 0$, and so periodic motion is inevitably stable. General trajectories are eventually periodic; the time taken to explore phase space and find the periodic trajectory seems to vary as $(1-\varepsilon)^{-\upsilon}, \upsilon \approx 5$. Hindmarsh and Jeffries have considered an impact oscillator in which

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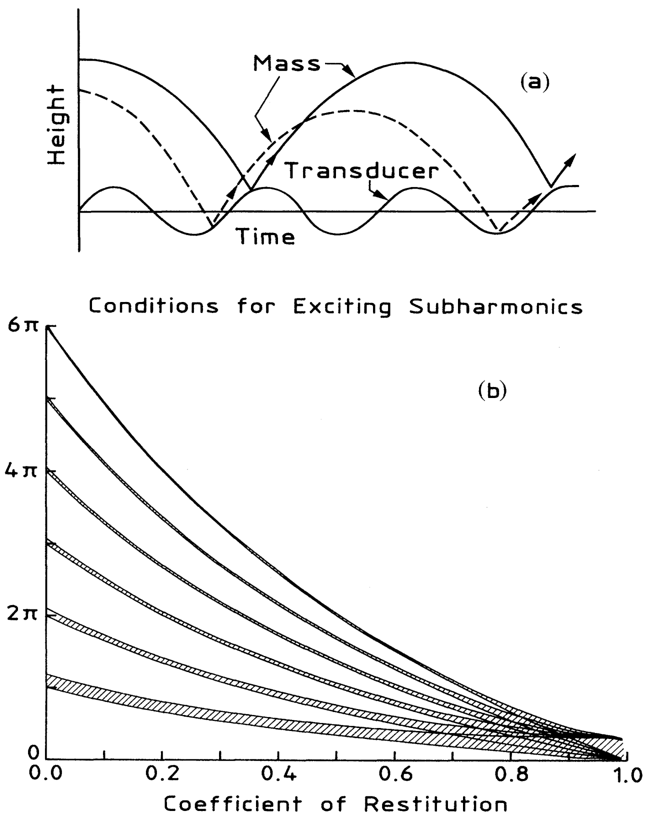

FIG. 1. (a) The heights of the mass and the transducer as a function of time. The broken (solid) line represents a nonperiodic (periodic) trajectory of the mass. In our experiment, the mass was the tip of an atomic force microscope (AFM) cantilever. (b) Regimes of stable periodic trajectories for the mass as a function of ε and the ratio a/a_0. The shaded areas are where the conditions of Eq. (1) are met.

a mass is mounted on a viscous spring, the other end of which is driven sinusoidally [9]. The mass collides with a rigid wall. This system exhibits behavior which has many similarities with the Pippard configuration, but there is a significant difference which depends on the position of the wall. If the wall is closer than the rest position of the mass so that it forces the spring into compression, then with increasing amplitude of excitation the regimes of stability for subharmonics occur in order of increasing n, as with the pin on the loudspeaker. But if the wall is beyond the rest position of the mass, then the order is inverted, e.g., the stable regime for period 3 occurs for a lower amplitude range than for period 2. The stability of this system has also been studied [10].

By using an atomic force microscope (AFM) [11], these subharmonic phenomena can be studied on a nanometer scale. The acceleration term a_0 in Eq. (1) varies linearly with the vibration amplitude and as the square of its frequency, so that by using a high frequency the displacement amplitude required to separate tip and sample is considerably reduced. In pioneering work on the detection of high-frequency vibrations in an AFM, the movement of the cantilever below its resonant frequency was measured [12]. Since the stiffness of the contact between the tip and the sample is nonlinear (because the area changes with load), the static deflection of the cantilever gives an indication of the high-frequency vibration amplitude. In practice it is difficult to measure static displacement because of drift, and so the signal to the transducer was modulated at a relatively low frequency. The contact acts as a mechanical diode, demodulating the surface vibration in a manner analogous to a crystal radio receiver. In this way measurements and images of elastic properties of material surfaces have been made at frequencies up to 114 MHz [13].

It is also possible to measure the cantilever movement directly at high frequencies [14], and this was the technique adopted here (without modulation) for observing nanosubharmonics. A high-frequency transducer is mounted on the bottom of a sample and elastic waves cause the top surface to vibrate. The tip of the cantilever acts as the bouncing mass, and its displacement may be measured by the deflection of a laser beam reflected from the back of the cantilever. The bandwidth of the optical detection extended up to 1 MHz. This is considerably above the resonant frequency of the free AFM cantilever, which therefore approximates well to a mass supported by a light spring. The cantilever deflection signal could be examined with a digital oscilloscope and also a spectrum analyzer. The most reproducible results were obtained with smooth, flat samples such as calcite and graphite, and cantilever tips of silicon (oxide). The experiments were performed in ambient conditions. Quasistatic force-distance curves were measured with the high-frequency excitation present, and as expected it was found that the pull-off force (the tensile force necessary to overcome the adhesion between the tip and the sample and separate them) decreased with increasing amplitude.

The subharmonic behavior of the cantilever was studied as a function of the excitation amplitude and also the mean deflection of the cantilever and hence the mean restoring force. A representative measurement of the response of the cantilever is presented in Fig. 2. Figure 2(a) contains two oscilloscope traces. The upper trace is the excitation signal to the transducer, of period 1.06 μs and amplitude at the transducer 30 V, giving a surface displacement of order 1 nm and causing the tip to bounce a few nanometers. The separations at which attractive forces are first detected are generally larger than this. The lower trace shows the displacement of the cantilever, which has a period of 8.48 μs, i.e., period 8. The spectrum analysis of the lower trace is given in Fig. 2(b). The excitation frequency appears as a peak at 948.75 kHz. The period 8 peak is at 117.5 kHz, and is 24 dB stronger. The other peaks are harmonics of the period 8 peak (bouncing motion is not expected to be sinusoidal); the

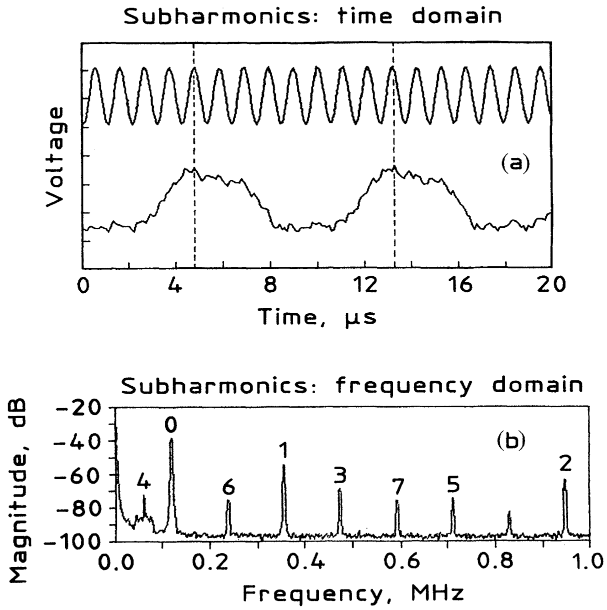

FIG. 2. (a) Oscilloscope traces of the excitation (upper trace) of the transducer and response (lower trace) of the cantilever tip, i.e., the mass. (b) Fourier transform of the cantilever response in (a). The peaks are numbered according to their magnitude. Peak number 2 represents the excitation frequency, peak 0 is the $n = 8$ subharmonic, and peak 4 is the free cantilever resonance. The other peaks are harmonics of peak 0.

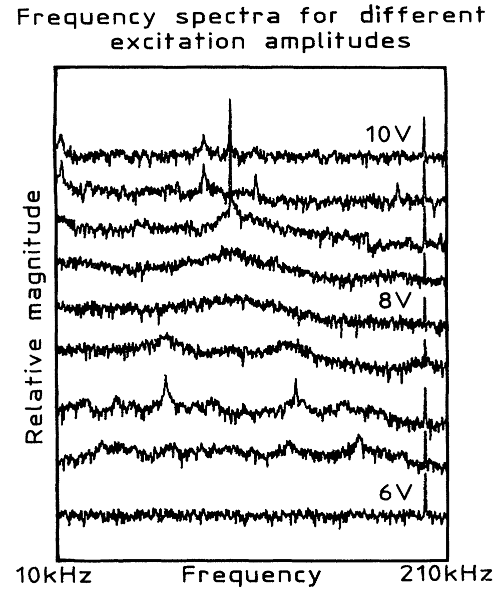

FIG. 3. Frequency spectra for different excitation amplitudes. The amplitude was decreased by 0.5 V after each spectrum was acquired.

strongest is the third harmonic which is 15 dB smaller. For an ideally parabolic trajectory the qth harmonic of the bouncing frequency would have an amplitude $\propto q^{-2}$. We found that periods of powers of 2 were favored (the highest achieved was period 16), which may relate to period doubling bifurcations leading to chaos [4]. Chaos could always be observed at high enough amplitudes of the driving signal. For certain parameters it was possible to observe chaos followed by periodic behavior, though contrary to the predictions of [7] this would then revert back to chaotic behavior, giving irregular alternation between periodic and aperiodic behavior. If a given subharmonic was being excited, changing the excitation amplitude or the mean static force would cause the behavior to become aperiodic. This corresponds to having ε not too close to unity and dropping out of the condition for that subharmonic before the condition for the next value of n is reached in Eq. (1), i.e., moving up or down the y axis of Fig. 1(b) for a particular ε.

A result from a systematic study of the dependence on excitation amplitude is shown in Fig. 3. The traces show spectral analyses of the cantilever response, starting with the highest excitation amplitude at the top. The excitation frequency was 204 kHz, and the three highest amplitude traces (10, 9.5, and 9 V) indicate period 2 behavior. This experiment was performed with a static tensile force, i.e., the cantilever pulling upwards against adhesive forces. In such conditions sidebands were often seen as in the second trace; the origin of these is not understood. As the excitation amplitude was decreased further the period 2 peak disappeared, and although a small peak was still present at the fundamental frequency the oscilloscope trace revealed that the behavior had become aperiodic. Further reduction of the amplitude led to the period 3 behavior seen with its harmonic in the seventh trace (7 V). At 6.5 V aperiodic behavior was again observed, and at 6 V the cantilever no longer bounced upon the surface but remained in contact (though with a smaller displacement amplitude than the transducer because of the contact compliance). This switching between stable harmonics and aperiodic behavior is just what is predicted by Eq. (1), but the order is wrong: the higher periods should be seen at higher amplitudes. The inversion of the order was found to be related to a tensile cantilever force, and corresponds to the order of subharmonics predicted for an offset impact oscillator with the wall placed beyond the equilibrium position of the mass [9].

Different behavior could also be obtained by varying the static force of the cantilever. This is another way of changing a_0/a in Eq. (1). Figure 4 shows a series of spectra obtained by ramping the mean position of the sample down away from the cantilever. The excitation frequency was 204 kHz. The two top and bottom traces are completely flat except for noise. The top two traces represent a repulsive load high enough such that insufficient acceleration was given to the cantilever for it

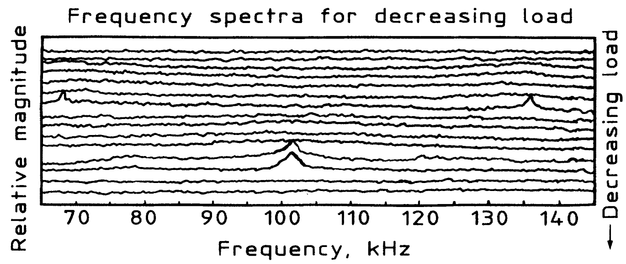

FIG. 4. Frequency spectra as a function of load. The sample was retracted by 21 nm after each spectrum was acquired, corresponding to a decrease in the force of about 46 nN. The excitation frequency was 204 kHz.

to overcome the repulsive load and lose contact with the sample. Starting with the third trace from the top down to the third trace from the bottom, the response moves through aperiodic, period 3, aperiodic, and finally period 2 behavior. For the bottom two traces, the sample position was far enough away from the cantilever such that all contact was lost. Some hysteresis was observed, so that the transitions occurred at slightly different points during a reverse loading sequence.

Although the exact conditions determining the thresholds and the sequence of the different subharmonics remain an intriguing problem, in general these results seem to indicate that it is indeed possible to obtain on a nanoscale the various phenomena that are predicted by macroscopic bouncing contact models. The difference between big and small may lie in the interpretation of the coefficient of restitution. In macroscopic experiments, ε is dominated by bulk viscoelastic or plastic properties, which can also account for the dependence of ε on the impact parameters. But if adhesion is present, then ε is less than unity even for perfectly elastic materials [15]. This is because of the hysteresis introduced into the loading-unloading force-distance relationship; when the surfaces are pulled apart work must be done against adhesive forces at separations where during approach the initial attraction was much smaller. This effect can be enhanced by local viscoelastic or plastic processes as the surfaces are peeling apart. As the load on the contact is reduced, it is expected that adhesive effects will increasingly dominate the coefficient of restitution ε, and hence the properties which can be measured and imaged using the phenomena described here. By calibrating the amplitude of the surface displacement, it will be possible to use a scanning probe microscope to determine ε locally with nanometer resolution, and to apply this to the study of dissipative processes such as the practical problems of the impact of powders and toner particles on surfaces and stick-slip motion.

Christine Mayencourt's measurements of subharmonic behavior at the macroscale with a pin on a loudspeaker stimulated the rest of this work. Neville Robinson's periodic explanations of chaos theory were not at all chaotic.

[1] C. Zener, J. Phys. (Paris), Colloq. **46**, C10-1–4 (1985).
[2] D. Tabor, Rev. Phys. Technol. **1**, 145–179 (1970).
[3] D.T. Kagan, Am. J. Phys. **58**, 151–154 (1990).
[4] J.M.T. Thompson and H.B. Stewart, *Nonlinear Dynamics and Chaos* (John Wiley and Sons, New York, 1991).
[5] A.B. Pippard, *The Physics of Vibration I* (Cambridge University Press, Cambridge, 1978).
[6] A. Mehta and J.M. Luck, Phys. Rev. Lett. **65**, 393–396 (1990).
[7] J.M. Luck and A. Mehta, Phys. Rev. E **48**, 3988–3997 (1993).
[8] C. Budd and F. Dux, Philos. Trans. R. Soc. London A **347**, 365–389 (1994).
[9] M.B. Hindmarsh and D.J. Jefferies, J. Phys. A **17**, 1791–1803 (1984).
[10] S. Foale, Proc. R. Soc. London A **347**, 353–363 (1994).
[11] N.A. Burnham and R.J. Colton, *Scanning Tunneling Microscopy and Spectroscopy: Theory, Techniques and Applications,* edited by D.A. Bonnell (VCH, New York, 1993), pp. 191–269.
[12] O. Kolosov and K. Yamanaka, Jpn. J. Appl. Phys. **32**, L1095–1098 (1993).
[13] K. Yamanaka, *Advances in Acoustic Microscopy I,* edited by G.A.D. Briggs (Plenum, New York, 1995), pp. 301–341.
[14] U. Rabe and W. Arnold, Appl. Phys. Lett. **64**, 1493–1495 (1994).
[15] K.L. Johnson and H.M. Pollock, J. Adhesion Sci. Technol. **8**, 1–9 (1994).