Hysteresis of the cantilever shift in ultrasonic force microscopy

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We propose a technique based on ultrasonic force microscopy that exploits the hysteresis in cantilever jumping to and from a sample while varying the ultrasonic amplitude. Both the elastic modulus and the work of adhesion can be determined by comparision with a relation derived between their ratio and the cantilever shift at the jump-in point. The method is applied to measurements on an aluminum thin film.
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The application of periodic strain to a sample with an atomic force microscope (AFM) tip can be used to map elastic moduli and adhesion in complex composite materials. In particular, ultrasonic force microscopy (UFM) shows promise for mapping hard materials such as metals or semiconductors using standard, compliant AFM cantilevers;[1,2] high frequency ($\sim$MHz) ultrasonic vibration, of controlled amplitude A, is excited in the sample or cantilever base, and the tip–sample distance is therefore modulated at this excitation frequency. Rectification of this vibration shifts the average position of the cantilever owing to the nonlinear tip–sample interaction, and it is this shift that is mapped to produce a UFM image. The cantilever response in UFM is a complex function of the elastic properties, the work of adhesion and the amplitude A.[1,3,4] This has hindered the development of a general method to separately extract the elastic properties and the work of adhesion from the UFM signal.

UFM has recently been applied to quantitative elastic property measurement by careful study of the cantilever ''jump,'' in which the average cantilever position changes suddenly due to the hysteresis in the force–distance curve.[5,6] Dinelli *et al.* evaluated the contact stiffness by comparing the jump positions for different applied loads.[5] Inagaki *et al.* measured an unexpectedly large contrast in the UFM signals of two semiconductors, and interpreted this as a consequence of such jumps.[6] In this letter, we propose and demonstrate a method for quantitative UFM signal analysis that allows the separate determination of the elastic modulus and the work of adhesion by exploiting the hysteresis of the cantilever jump.

We take into account both elastic and adhesive forces using the Johnson–Kendall–Roberts–Sperling (JKRS) model.[7] This model strictly holds in the limit of soft samples and large contact radii, but can be applied to a reasonable approximation to a wide range of materials and tips.[8] Consider the contact between a sphere (of radius R, Young's modulus E_2, and Poisson's ratio ν_2) and a flat surface (of Young's modulus E_1 and Poisson's ratio ν_1) with applied load F. W is the work of adhesion, and K is the reduced elastic modulus defined by $4/(3K)=(1-\nu_1^2)/E_1+(1-\nu_2^2)/E_2$. The contact is characterized by the contact radius a and the indentation δ. For convenience, we use the normalized JKRS model represented by

$$\bar{F}=\bar{a}^3-\bar{a}\sqrt{6\bar{a}}, \tag{1}$$

$$\bar{\delta}=\bar{a}^2-\frac{2}{3}\sqrt{6\bar{a}}, \tag{2}$$

where $\bar{a}=a/(\pi R^2 W/K)^{1/3}$ is the normalized contact radius, $\bar{F}=F/(\pi RW)$ is the normalized applied load, and $\bar{\delta}=\delta/(\pi^2 RW^2/K^2)^{1/3}=\delta/\delta_0$ is the normalized indentation depth.[9]

When the tip is approaching the sample, at the moment of contact between the tip and the sample ($\bar{\delta}=0$) a sudden attractive (normalized) force $\bar{F}=-4/3$ is exerted between the tip and the sample. In a single cycle of deformation executed under static or near-static conditions, the sudden change of the tip–sample interaction makes the cantilever jump to a new equilibrium position in a time determined by the inverse of the fundamental resonance frequency of the cantilever and its damping. This kind of jump does *not* happen in a single cycle at frequencies appropriate to UFM. Because of the ultrasonic excitation, the inertia of the cantilever tends to keep the cantilever in periodic motion, exhibiting only an average response to the cyclically varying force (even if this cycle exhibits discontinuities in force). When the tip is moving away from the sample, the contact is kept for a while in the $\bar{\delta}<0$ region. According to the JKRS model, the tip–sample contact breaks when $\bar{\delta}=6^{-2/3}-(2/3)6^{1/3}\simeq-0.9086$. The force–indentation curve exhibits a bifurcation in the range $-0.9086<\bar{\delta}<0$, resulting in hysteresis.

Kolosov *et al.* proposed the following formula,[1] based on a spring and mass model, for the cantilever shift in UFM in the limit of high-frequency excitation:

$$k_c y_0=\frac{1}{T}\int_0^T F(-\delta_{\mathrm{av}}-A\cos\omega t)\,dt, \tag{3}$$

where $T=2\pi/\omega$ is the period of the ultrasonic vibration, A is the amplitude of the sample vibration, k_c is the static cantilever stiffness, $F(-\delta)$ is the tip–sample force, δ_{av} is the average indentation, and $y_0=y_{00}+\Delta y$. Here, y_{00} is the position of the end of the cantilever with no ultrasonic excitation and Δy is the average outward cantilever shift owing to the nonlinear interaction in the presence of ultrasonic excita-

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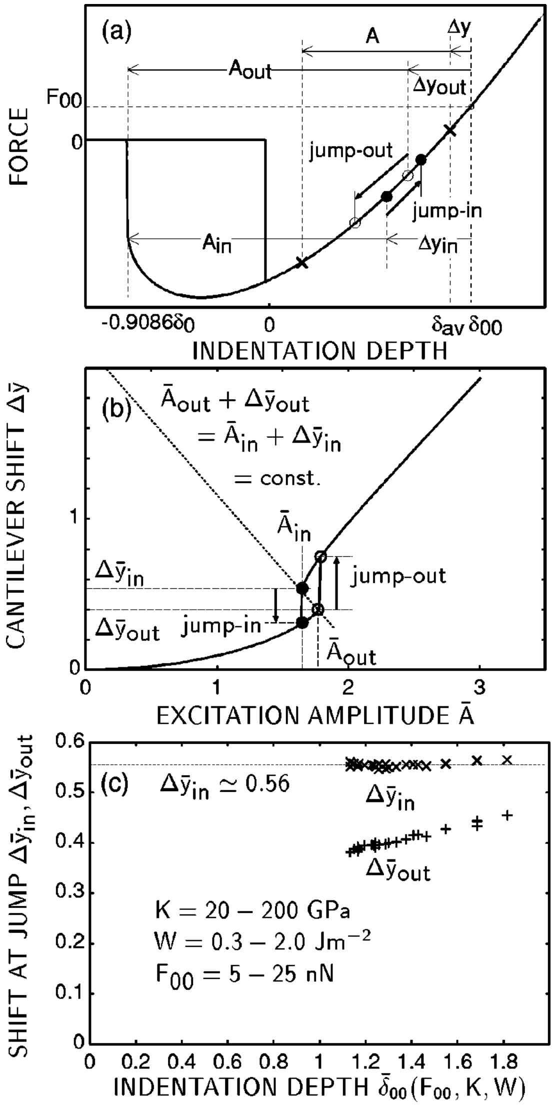

FIG. 1. (a) JKRS force–indentation curve. Open and closed circles denote the positions where the cantilever jumps out and in, respectively. (b) Normalized JKRS UFM cantilever shift as a function of normalized amplitude for a typical hard material ($K=100$ GPa and $W=0.5\ \mathrm{Jm}^{-2}$). Dotted curve: $\bar{A}+\Delta\bar{y}=$const., on which both jump in and jump out take place. (c) Normalized JKRS jump-in ($\Delta\bar{y}_{\rm in}$) and jump-out ($\Delta\bar{y}_{\rm out}$) cantilever shifts as a function of normalized initial indentation $\bar{\delta}_0$, for the range of F_{00}, K, and W considered.

tion. The indentation $\delta_{\rm av}$ can be expressed as $-\delta_{\rm av}=y_0-z_0$, where z_0 is the static sample displacement. Equation (3) is based on the assumption that the cantilever is subject to an average force over an excitation cycle. In UFM calculations, it is standard procedure to determine y_0 from Eq. (3), and then to extract Δy using y_{00} calculated from $k_c y_{00}=F(y_{00}-z_0)$.

The hysteresis of the cantilever jump can be understood from Eq. (3). Figure 1(a) shows the JKRS F-δ relation. If no ultrasonic excitation is applied, the cantilever stays at the indentation $\delta_{00}=z_0-y_{00}$ defined by the initial applied load F_{00}. On increasing A the average cantilever position shifts outwards, as shown by the general point marked by $\delta_{\rm av}$ in Fig. 1(a). Here $\Delta y=\delta_{00}-\delta_{\rm av}$, and the indentation oscillates in the range $\delta_{\rm av}-A$ to $\delta_{\rm av}+A$. Figure 1(a) also shows that the hysteresis loop begins to be traced out when Δy reaches $\Delta y_{\rm out}$, determined by

$$\Delta y + A = \delta_{00} + 0.9086\delta_0, \tag{4}$$

which applies at $\Delta y=\Delta y_{\rm out}$ when the average force abruptly changes, resulting in the jump out [open circles in Fig. 1(a)]. Similarly, when the amplitude is subsequently decreased, the system ceases to trace out the hysteresis loop at the shift $\Delta y_{\rm in}$ determined by Eq. (4) with $\Delta y=\Delta y_{\rm in}$, resulting in the jump in (solid circles).

We have calculated the cantilever response including both jump in and jump out for a wide range of F_{00}, K, and W ($F_{00}=5$–25 nN, $K=20$–200 GPa, and $W=0.3$–2.0 Jm^{-2}) using a normalized version of Eq. (3) with $k_c=0.5\ \mathrm{Nm}^{-1}$. Figure 1(b) shows the calculated normalized UFM cantilever shift $\Delta\bar{y}=\Delta y/\delta_0$ as a function of the normalized amplitude $\bar{A}=A/\delta_0$ for a typical hard material ($K=100$ GPa and $W=0.5\ \mathrm{Jm}^{-2}$) with $F_{00}=10$ nN. When $\bar{A}$ is increased, the cantilever jumps out when $\Delta\bar{y}$ is equal to $\Delta\bar{y}_{\rm out}$ (open circles), whereas it jumps in at $\Delta\bar{y}_{\rm in}$ when $\bar{A}$ is decreased (closed circles). As discussed, both the jump-in and the jump-out points fall on the line $\Delta\bar{y}+\bar{A}=$ const. in Fig. 1(b). For typical AFM cantilever stiffnesses ($k_c<10\ \mathrm{Nm}^{-1}$), the curve in Fig. 1(b) is insensitive to the value of k_c.

Figure 1(c) shows the calculated normalized shifts at jump in and jump out versus the normalized initial indentation depth $\bar{\delta}_{00}$ for various values of F_{00}, K, and W in the ranges previously defined. A given set of F_{00}, K, and W determines $\Delta y_{\rm in}$, $\Delta y_{\rm out}$ and $\bar{\delta}_{00}$. The normalized cantilever shift at jump in, $\Delta\bar{y}_{\rm in}\approx 0.56$, is constant to a very good approximation (± 0.01) and is effectively independent of the choice of F_{00}, K, W, and hence $\bar{\delta}_{00}$. This leads to a universal relation between W, K, and the shift at jump in: $\delta_0=(\pi^2 R W^2/K^2)^{1/3}\approx\Delta y_{\rm in}/0.56$. This allows the ratio W/K to be directly obtained from the data for $\Delta y_{\rm in}$ and a knowledge of the tip radius R. The initial indentation δ_{00} can then be found from Eq. (4) applied at jump in if the amplitude at jump in, $A_{\rm in}$, is known. Together with the known initial applied force F_{00}, the values of W and K can then be separately derived from the JKRS equations.

We applied this method in an UFM experiment. A commercial AFM (Seiko SPI-3700) was modified to allow ultrasonic vibration of the sample in a direction normal to the sample surface. A piezoelectric transducer (0.2 mm thick) mounted below the sample introduces this vibration at 1.5 MHz, chosen well away from any mechanical resonance of the transducer. The output of the AFM position-sensitive optical detector is fed to a digital sampling oscilloscope (Iwatsu–LeCroy LC574) and averaged over 5000 times. The transducer vibration amplitude is calibrated with a Michelson interferometer. A rectangular silicon cantilever of dimensions $130\times35\times0.7\ \mu\mathrm{m}^3$, stiffness $0.6\ \mathrm{Nm}^{-1}$, nominal tip radius $R=10$ nm, and fundamental flexural resonance frequency 75 kHz is used. The ultrasonic vibration amplitude is modulated at 400 Hz (see Fig. 2), chosen to be well below this flexural resonance and higher than the reciprocal of the (constant force) feedback time constant ($\sim$16 ms). The sample is a 100 nm polycrystalline aluminum film evaporated on a silicon substrate.

The curves in Fig. 2 show the measured excitation wave form and also the UFM cantilever shift as a function of time for an average load of 10 nN ($\approx F_{00}$).[10] This wave form

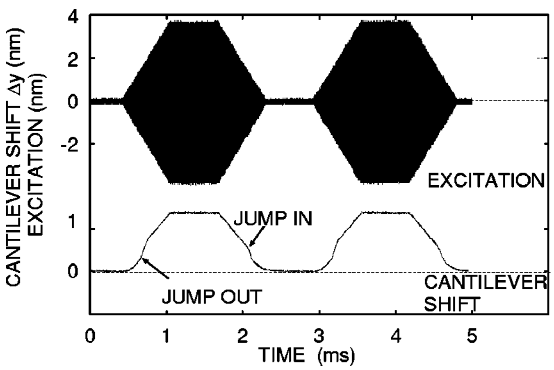

FIG. 2. Measured excitation wave form (top) and UFM cantilever shift (bottom) as a function of time.

allows the hysteresis of the response to be probed. The shift and excitation amplitude could be calibrated by displacing the sample stage by known amounts. Figure 3 shows the experimental UFM cantilever shift as a function of excitation ultrasonic amplitude A, clearly indicating hysteresis. The positions of jump-in and jump-out fall on the curve $\Delta y + A = \text{const.}$ as expected.

The result of the calculation of K and W gives $K = 20$ GPa and $W = 1.6\ \text{Jm}^{-2}$. The corresponding predicted Δy-A relation is shown by the solid curve in Fig. 3. The fit for $\Delta y < \Delta y_{\text{in}}$ is reasonable, but degrades at higher shifts. The predicted shape of the hysteresis loop is also different from that observed. Moreover, the value of $K = 20$ GPa derived is smaller than that expected (~ 68 GPa) for this tip–sample combination.[11] Possible reasons for these discrepancies are (1) the presence of nanoasperities, an oxide layer or a water film, or (2) the effects of the high strain rates used, viscoelasticity, or plastic flow at the relatively high stresses involved (~ 0.2 GPa). Large deviations from theory in this field are common owing to these reasons.[5] However, the deviations become much less when tips of somewhat larger radii, of the order of 50–200 nm, are used because of the lower stresses ($\propto R^{-2/3}$ for the Hertz model), larger probed volumes ($\propto R$), and concomitant smaller influence of asperities.[12] We can therefore expect a much better accuracy and less sensitivity to impurity layers with the use of tips of larger radius.

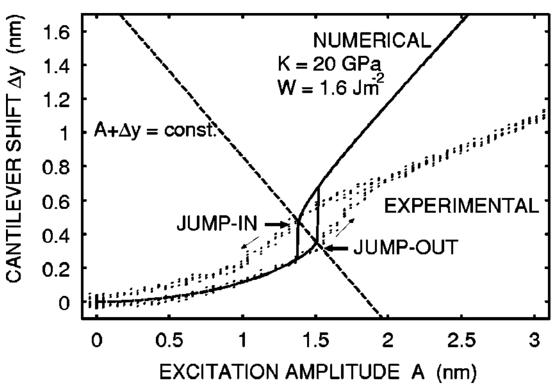

FIG. 3. UFM cantilever shift as a function of excitation amplitude. The dashed curve represents $A + \Delta y = \text{const.}$ and the solid curve the JKRS prediction with $K = 20$ GPa and $W = 1.6\ \text{Jm}^{-2}$.

In summary, we propose a method of UFM signal analysis allowing the separate determination of the elastic modulus and work of adhesion on the basis of the JKRS model by exploiting the hysteresis of the cantilever jumps as a function of ultrasonic amplitude. It should be possible to extend the general method to more complicated models of the force–distance curve and to a wide variety of both soft and hard materials. Moreover, the method could be combined with lateral scanning to produce an UFM imaging mode.

[1] O. Kolosov and K. Yamanaka, Jpn. J. Appl. Phys., Part 2 **32**, L1095 (1993).
[2] O. V. Kolosov, M. R. Castell, C. D. Marsh, G. A. D. Briggs, T. I. Kamins, and R. S. Williams, Phys. Rev. Lett. **81**, 1046 (1998).
[3] K. Yamanaka, H. Ogiso, and O. Kolosov, Jpn. J. Appl. Phys., Part 1 **33**, 3197 (1994).
[4] U. Rabe, V. Scherer, S. Hirskorn, and W. Arnold, J. Vac. Sci. Technol. B **15**, 1506 (1997).
[5] F. Dinelli, S. K. Biswas, G. A. D. Briggs, and O. V. Kolosov, Phys. Rev. B **61**, 13995 (2000).
[6] K. Inagaki, O. V. Kolosov, G. A. D. Briggs, and O. B. Wright, Appl. Phys. Lett. **76**, 1836 (2000).
[7] K. Johnson, K. Kendall, and A. D. Roberts, Proc. R. Soc. London, Ser. A **324**, 301 (1971); G. Sperling, Ph.D. thesis, Karlsruhe Technical High School, 1964.
[8] D. Maugis, J. Colloid Interface Sci. **150**, 243 (1992).
[9] In Ref. 6, the JKRS equations quoted contain a misprint.
[10] $F_{00} \approx 10$ nN because the change in force on switching off the ultrasound in the absence of feedback is $\Delta y_{\text{av}} k_c \sim 0.5\ \text{nm} \times 0.6\ \text{Nm}^{-1} = 0.3$ nN, where Δy_{av} is the average of Δy over time (in Fig. 2). Moreover, the downward shift of the sample stage (by Δy_{av}) due to the feedback has a negligible influence here, and is ignored.
[11] Decreasing R used in the fits by a factor of 3.4^2 would yield $K = 68$ GPa in agreement with the bulk value, but gives an unreasonably large value of W increased by a factor of 3.4 to $W = 5.4\ \text{Jm}^{-2}$. (The theoretical curve of Fig. 3 is unchanged by this.)
[12] U. Rabe, S. Amelio, E. Kester, V. Scherer, S. Hirskorn, and W. Arnold, Ultrasonics **38**, 430 (2000).