High frequency tribological investigations on quartz resonator surfaces

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We report on the application of quartz crystal resonators for friction measurements in the MHz regime. The dissipative processes are studied by approaching a small sphere to the quartz surface and measuring the shifts in frequency and bandwidth as a function of the sphere–sample distance. Once elastic contact is established, the frequency and the bandwidth can be well described by an increased stiffness of the quartz–sphere system. In the range of distances where contact is just being formed, we observe strongly different behavior for high-friction metal–metal interfaces and low-friction fluoropolymer interfaces. For high-friction interfaces there is an excess dissipation of energy which we attribute to frictional processes in the contact zone. © 1999 American Institute of Physics. [S0021-8979(99)02407-X]

I. INTRODUCTION

Quartz crystal resonators are quite unique objects in the sense that their resonances display extremely little damping.1,2 This is in part due to the intrinsically low dissipation of crystalline quartz. Other factors are the lack of emission of longitudinal sound and the fact that dissipation of mechanical energy into the mounting structure can be prevented by confining the oscillation to the central portions with “energy trapping.”2 Because of the extreme sharpness of the resonances, quartz resonators have unsurpassed capabilities as frequency control elements and have gained widespread use as such. Slight disturbances, on the other hand, are also detected with high sensitivity. In fact, part of the problem in manufacturing frequency standards from quartz resonators is to shield the resonators from mechanical, acoustical, thermal, or electric disturbances. Changes of frequency with temperature,2 pressure,3 stress,4 mass loading,5 viscosity,6 and others have been widely exploited for sensoric purposes.

In this article we report on the change of frequency and bandwidth upon touching the face of the quartz blank with a small sphere. As we show, the disturbance is small enough to be treated as a perturbation. The resonance frequency increases in all cases investigated so far. We interpret this as an effect of “added stiffness.” In the same way as a thin coating increases the effective mass of the resonator, the contact with the sphere increases its effective spring constant. When the sphere just barely touches the quartz surface we observe dissipative processes in the contact zone. The bandwidth increases more strongly than the frequency in this range. These processes allow for the investigation of interfacial friction between the quartz surface and the sphere. Quartz resonators give access to tribology in a range of speeds and frequencies unachievable with other instruments like the surface forces apparatus (SFA)7,8 or the atomic force microscope (AFM).9,10 Quartz resonators have been used previously to investigate the frictional behavior of adsorbed molecules at cryogenic temperatures.11–13 Note, however, that this is an entirely different regime of friction where the shear stress caused by the film’s inertia is extremely weak.14 In this regime, the effect of shear stress is to slightly modify the activation barriers for surface diffusion. In the experiments reported here the shear stress can be estimated to be well in the MPa range where yielding and wear are certainly possible. The relative speed between the sphere and the substrate can be as high as 1 m/s, the accessible frequencies being between 4 and 100 MHz. These speeds and time constants come close to the conditions given in magnetic disk drives.15 Due to the short timescale, some of the relaxation processes observed with the SFA and AFM should be suppressed for quartz resonators. A new regime of friction can be accessed experimentally, the investigations of which should have large impact for practical applications.

II. EXPERIMENT

Figure 1 gives a sketch of the experimental setup. We used planar quartz blanks with a fundamental frequency of 4 and 10 MHz. The natural roughness of the surface as determined from AFM images is some tens of nanometers. A sphere with a radius \( R \) of 3.5 mm is approached to the quartz surface from above by means of a piezoactuator. The actuator (P-732.ZC from Polytec) has capacitive displacements sensors and is feedback controlled to a resolution of 1 Å with an absolute accuracy of 1 nm. At this point, we do not have a normal force sensor implemented. However, the normal force can be estimated from the known vertical movement of the sphere after contact is established. The latter is about 2 \( \mu \text{m} \). Since the quartz plate is by far the weakest element in the structure, the normal force is largely determined by the quartz plate’s bending rigidity. The effective spring constant of the circular disk suspended around its rim with an external load acting on its center is given by \( k = \frac{86 \pi E t^3}{R_p^3} \left( 3 R_p^2 (1 - \nu^2) \right) \), with \( E \) the Young’s modulus, \( \nu \) the Poisson’s number, \( t \) the thickness, and \( R_p \) the radius of the plate.16 Using \( E \sim 86 \text{ GPa} \), \( \nu \sim 0.11 \), \( t = 400 \mu \text{m} \), and \( R_p \sim 7 \text{ mm} \), we arrive at
a vertical stiffness of about $5 \times 10^5$ N/m. With a maximum displacement of 2 \( \mu \)m this yields a maximum force \( F \) of about 1 N. With this normal force, we can in turn estimate the contact radius \( r_c \) from the Johnson–Kendall–Roberts (JKR) equation\(^{18} \) to be less than 20 \( \mu \)m. Here we have assumed that \( F / R \) is larger than the interfacial energy which is certainly true for the maximum force. Given the substantial normal force, one may wonder whether the frequency shifts could be induced by the bending stress in the quartz rather than by the increased stiffness. However, bending stress should decrease the frequency rather than increasing it\(^4 \) and can therefore not be the dominant effect.

The resonances are probed with an impedance analyzer (HP 5100A). We fit complex Lorentzians to the susceptance \( Y(\omega) \). The frequency and the bandwidth are extracted as fit parameters. The accuracy is better than 1 Hz for frequency as well as bandwidth. One measurement takes about 5 s. A third fit parameter also obtained is the amplitude of the resonance ("oscillator strength"). Generally speaking, the amplitude of resonance is susceptible to various kinds of disturbances and is therefore not usually analyzed or interpreted in great detail. In the experiments reported here, we find a discontinuous jump of the amplitude by some percent at a sphere–sample distance close to the first contact (\( z \approx 0 \) in the bottom part of Fig. 2). This jump is superimposed onto an irregular behavior as it is observed in other experiments as well. The fact that the jump is larger than the other irregularities and that it occurs at some specific sphere–sample distance makes us believe that it is connected to the contact. At present we do not have a clear interpretation of this phenomenon. Separate experiments suggested that the jump may be related to the establishment of electrical contact between the two surfaces.

III. MODELING

Provided that the interactions between the resonator and its environment are acoustic in nature, equivalent circuits have proven a very valuable tool for understanding and modeling quartz crystals.\(^{19–21} \) Equivalent circuit models are one-dimensional in nature, i.e., the acoustic waves inside the quartz are assumed to be planar with the lateral boundaries ignored. Anharmonic sidebands which have nodal planes

FIG. 1. Schematic of the experimental setup. The experimental parameters are the shifts in frequency \( \delta f \), the change in half bandwidth \( \delta f' \), and the amplitude A.

FIG. 2. A typical set of data. The origin of the \( z \) scale has been arbitrarily set to the onset of frequency changes. Negative distances correspond to the sphere pushing onto the quartz. Neither the frequencies nor the bandwidths (part a) give any indication of discontinuities which could have been induced by a jump into contact. When contact is established, the frequency increases which is interpreted in terms of an increased stiffness of the quartz–sphere system. As opposed to frequency and bandwidth, the oscillation amplitude does show a discontinuity, the significance of which is unclear at the moment.
across the quartz surface are not contained in this model. Given that the interaction with the sphere is very local, one may question the assumption that the modes of interest are still given by planar waves. When using the equivalent circuit one assumes that the influence of the sphere does not fundamentally change the mode pattern. In Fig. 3, we show conductance spectra $G(\omega)$ close to the fundamental of a 10 MHz quartz blank. Clearly, the peak height of the different anharmonic sidebands varies. Different modes are affected to different degrees depending on whether or not the sphere is touching the quartz surface close to a nodal line. Even though the peak height varies, the general pattern does not. This proves that the mode shapes are largely maintained even in contact with the sphere. The equivalent circuit model holds.

Figure 4 shows the widely used Butterworth–van Dyke equivalent circuit with an added element accounting for interactions. The Butterworth–van Dyke circuit is derived from the more complicated (but also more rigorous) Mason equivalent circuit with a number of approximations discussed, for example, in Ref. 19. One can show that an acoustic interaction with the environment may be represented by a load impedance $Z_L$, in series with the other elements of the acoustic branch. The resonance frequency $f^*$ is given by the condition where the mechanical branch of the circuit vanishes. Here, the $f^* = f^i + i f^v$ (f$^i$ the half width at half maximum) is a complex frequency containing the resonant fre-

![Fig. 3. Conductance spectra $G(\omega)$ close to the fundamental of a 10 MHz quartz resonator. All peaks apart from the one on the very left-hand side are anharmonic side bands. While the peak heights vary after contact with the sphere, the positions vary only slightly. The mode structure remains essentially unchanged.](image)

![Fig. 4. Equivalent circuit models. (a) The classical Butterworth–van Dyke circuit supplemented by an additional element to account for acoustic coupling to the environment. (b) Illustration of the electromechanical analogy. Because in mechanics the displacements (currents) are additive rather than the stresses (voltages), two serial mechanical elements have to be placed in parallel in the equivalent electrical circuit. (c) The entire equivalent circuit for a quartz resonator in contact with a small sphere.](image)

quency and the half bandwidth as its real and its imaginary part, respectively.

The lower part of Fig. 4(a) corresponds to the electric capacitance across the electrodes and is of minor importance in this context. The upper, acoustic elements are given by an inductive element $i \omega L_1$ ($L_1$ being proportional to the mass per unit area of the quartz blank $m_q$), a capacitive element $1/(i \omega C_1)$ ($1/C_1$ being proportional to the stiffness $G_q/d_q$ with $d_q$ the thickness and $G_q$ the shear modulus of the quartz), and a resistive element $R_1$ ($R_1$ being proportional to the viscous dissipation inside the quartz). All these elements have the dimension of stress/velocity×area. They are the acoustic analogs of the electric impedances which have the dimension of voltage/current. Note that in acoustics the impedance is usually normalized to area, while this is not done for electrical circuits. Also, a serial acoustic network transforms to a parallel electric network because in a serial acoustic network the velocities (‘‘currents’’) are additive rather than the stresses (‘‘voltages’’) [Fig. 4(b)].

Once the load impedance $Z_L$ is known, the induced frequency shift is simply given by

$$\delta f^* = \delta f + i \delta f^v = \frac{i f_0 Z_L}{\pi Z_q},$$

with $f_0$ the fundamental frequency and $Z_q = 8.8 \times 10^6$ kg m$^{-2}$ s$^{-1}$ the acoustic impedance of AT-cut quartz. In the derivation of Eq. (1) it was assumed that $Z_L$ is much smaller than the other elements of the circuit which is to say that the disturbance (or the normalized frequency shift $\delta f/f$) is small. Note that $Z_L$ is a complex quantity such that both the shift in frequency and in bandwidth are given by Eq. (1).
It is the conceptual simplicity of Eq. (1) which makes the equivalent circuits so attractive to the experimentalist.

Whereas simple mechanical analogs for the elements $L_1$, $C_1$, and $R_1$ exist, the mechanical analogs are not always as obvious for the impedance $Z_L$. For semi-infinite media like liquids, one has $Z_L = Z_{ac}$ with $Z_{ac}$, the acoustic impedance of the material. For more complicated configurations, one has to resort to calculating the stress at the quartz surface with a suitable acoustic model. For example, one finds $Z_L = i Z_f \tan(\omega d_f/\nu_f)$ for a film with thickness $d_f$, acoustic impedance $Z_f$, and speed of sound $\nu_f$. The only situation where the mechanical analog of the impedance $Z_L$ is simple and amenable to intuition is the case of a very thin film, in which one has $Z_L = i m f$ with $m_f$ the film’s mass per unit area. Interestingly, no situation has been reported so far where the impedance was given by a term $\kappa/(i\omega)$. While a small “added mass” is routinely measured with quartz crystal microbalances, no such “added stiffness” as given by the spring constant $\kappa$ has been reported in the literature. In this article we show that a small sphere touching the quartz surface represents a disturbance of this kind.

In the following we use a rather crude wave picture to estimate the order of magnitude of the spring constant $\kappa$. The model also justifies the use of a term $\kappa/(i\omega)$ for the load impedance and clarifies the nature of the interaction. Figure 5 shows a sketch. The radius of contact is assumed to be much smaller than the wavelength of sound. Although we have no direct way to experimentally determine the contact radius, an estimate can be made with the JKR model. Straightforward application of the JKR equation to a contact radius in the range of 10 μm. This is much less than the wavelength of sound at 12 MHz in brass which is 170 μm. We assume that a spherical sound wave emanates from the point of contact into the sphere. For the purpose of this estimation, we also assume that there is just one elastic modulus $K$ in the sphere, i.e., that longitudinal waves and shear waves travel with the same speed. With these simplifications, the deformation of the sphere may be approximated as

$$u(r) \approx u_0 \frac{r_c}{r} e^{-ikr} \quad \text{for } r > r_c,$$

$$u(r) \approx u_0 \quad \text{for } r < r_c,$$

(2)

where $u$ is the displacement, $r$ is the radial coordinate originating at the point of contact, $k$ is the wave number, $r_c$ is the contact radius, and $u_0$ is the amplitude of oscillation. All tensor indices have been dropped here for simplicity. The mechanical impedance is defined as

$$Z^s = \frac{\sigma}{\partial u/\partial t},$$

where $\sigma$ is the stress. Note that the impedance is by definition normalized to the area. Outside the contact area ($r > r_c$) we find for the stress

$$\sigma \sim K\nabla u \sim Ku_0 r_c \left( -\frac{1}{r^2} - \frac{ik}{r} \right) e^{-ikr} \quad \text{for } r > r_c,$$

(4)

where $K$ is the elastic modulus. Numerical factors like $2\pi$ have been dropped. For $r < r_c$ we again assume that the stress is constant everywhere and equal to the stress at $r = r_c$ evaluated according to Eq. (4), that is

$$\sigma \sim \frac{Ku_0}{r_c} (-1 - ikr_c) \quad \text{for } r < r_c.$$

(5)

At this point we have to account for the effect that the acoustic impedance is normalized to the area of the wave. In order to stay consistent with this convention we include a factor $r_c^2 / r_c^2$ ($r_c$ the radius of the active area which is about the same as the electrode area) into the impedance and write

$$Z_e = \frac{r_c^2}{r_c^2} \frac{\sigma}{\partial u/\partial t} - \frac{1}{r_c^2} \frac{K r_c}{i\omega} (-1 - ikr_c).$$

(6)

Inserting Eq. (6) into Eq. (1) one finds

$$\delta f^s = \delta f + i \delta\Gamma$$

$$= \frac{if_0}{\pi} \frac{Z_L}{Z_q} = \frac{f_0}{\pi Z_q} \frac{K r_c}{r_c^2} (1 + ikr_c)$$

$$\sim \frac{f_0}{\pi Z_q} \frac{1}{r_c^2} \frac{\kappa}{\omega} (1 + ikr_c).$$

(7)

The product of the modulus $K$ and the contact radius $r_c$ defines the spring constant $\kappa$. A similar relation results from simple scaling arguments. The parameter $\kappa$ should have the dimension N/m and the only way to construct such a quantity from the intrinsic parameters of the experiment is to write $\kappa \sim Kr_c$. Note that a spherical sound wave is essential in the calculations given in Eqs. (2)–(7). The first term in the brackets results from the differentiation of the factor $1/r$ in Eq. (2). The observed increase of resonance frequency therefore is an effect of near field acoustics. Plane waves radiated into an elastic medium in contact with the quartz will always decrease the resonance frequency. As long as the radius of contact $r_c$ is smaller than the wavelength of sound $\lambda$, the interaction is mostly elastic. The term $ikr_c$ accounts for the dissipation of energy via radiation of sound into the sphere. While the term $ikr_c$ is less than 1 in experiment, it is not negligible either. Some acoustic energy is lost via sound waves, thereby increasing the bandwidth. When firm contact is established these sound waves are the dominating channel for dissipation. This process of dissipation is different from
friction. It has been included as a separate element into the equivalent circuit in Fig. 4(c). The ratio of the increase in bandwidth $\Delta f$ and in frequency $\delta f$ ("loss tangent") is equal to the quantity $kr_c$ and can therefore be used to assess the area of contact.

Friction is accounted for by a separate resistive element in the equivalent circuit. This element has to be added in parallel to the term for elastic contact [see Fig. 4(b)]. Modeling friction with equivalent circuits implies that the friction forces depend linearly on the sliding speed. While this linear model has been used previously in quartz–crystal work, it is by no means obvious. Both the time-honored Amonton's law and recent results of nanotribology state that the friction force often is independent of the sliding velocity. When searching for nonlinearities in our experiments, we found them to be astonishingly small. This issue certainly needs further investigation. For the time being, we adopt the linear model as a hypothesis, the validity of which remains to be tested. Nonlinearities cannot be treated in the framework of equivalent circuits.

IV. RESULTS AND DISCUSSION

Figure 2 shows a typical trace of the raw data. Displayed are the frequency shift and the shift in bandwidth. First, and importantly, the frequency increases upon contact. As discussed in the Sec. III we associate this increase with an increase in the overall stiffness of the system. According to Eq. (7) the increase in the friction coefficient should be proportional to the contact radius. The contact radius, in turn, should be proportional to the third root of the normal force provided that the normal force is larger than the interface tension times the radius of the sphere which is fulfilled for most of the experiments. Following this line of reasoning, we have inserted a third-root law according to the JKR theory as a dotted line into the data set. This illustrates that the elastic coupling is correlated with the contact radius.

At no point in the frequency and bandwidth data does one observe any discontinuity. Apparently, there is no jumping into contact. This contrasts to AFM experiments where jumping into contact frequently occurs. In this experiment, the vertical stiffness of the system is larger than typical force gradients originating from van der Waals forces. The vertical stiffness is given by the bending rigidity of the quartz plate and is in the order of $5 \times 10^5 \text{ N/m}$. Force gradients from van der Waals forces, on the other hand, are given by $A_R R/(3d^3)$ with $A_R \approx 4 \times 10^{-19} \text{ Nm}$ the Hamaker constant. Inserting numbers one finds that jumping is only expected at a distance of about 1 nm where the roughness of the quartz surface dominates the picture.

In Fig. 6 we show the frequency shift $\delta f$ and the loss tangent $\Delta f/\delta f$ for three different interfaces which are a gold–brass contact, a SiO$_x$–brass contact, and a fluorocarbon–fluorocarbon contact. The SiO$_x$ layer had been evaporated onto the gold electrode of the quartz resonator. The fluorocarbon surfaces had been generated by a plasma polymerization process. These interfaces strongly differ from each other in their tribological behavior. While metal–metal interfaces slide very badly, sliding is greatly facilitated for fluorocarbon interfaces due to their low surface energies. This difference is clearly reflected in the loss tangent $\Delta f/\delta f$ around $d=0$. While the metal–metal contacts display a strong maximum, no such peak is observed for fluoropolymers.

Figure 7 shows data taken on different harmonics in order to investigate the frequency dependence of the interaction. Displayed are data taken on the third and the fifth overtone of a 4-MHz quartz resonator. The quartz plate used is an "overtone resonator" with two flat, optically polished faces. Metal–metal contacts are known to have poor tribological properties while fluorocarbons slide very well.

Finally, we show the results of our first attempts to find nonlinearities in Fig. 8. If stick–slip motion was present, one should observe a dependence of the frequency shifts on the driving amplitude of the quartz oscillator. When changing the driving amplitude by a factor of 10 (that is changing the power by a factor 100) we could not see a variation of frequency shifts which we would consider significant. The
gests that the loss tangent is proportional to the contact radius, as predicted when the experiments are performed in the MHz regime. This sug-
 variations seen in Fig. 8 are within the margin of reproduc-

come to rest which may be pre-
for providing the fluorocarbon coatings.

FIG. 7. Dependence of frequency shifts and bandwidths on overtone order. The frequency shifts are lower on higher harmonics. This is in accordance with a load impedance of the form $\frac{1}{\sqrt{k c}} \frac{k}{f}$. The loss tangent $\frac{\Delta f}{f}$ increases on higher harmonics because the wavelength of sound is smaller and the term $k r$, [Eq. (7)] therefore increases. Note that the loss tangent increases with the normal force roughly like the frequency shift. This sug-

V. CONCLUSIONS

We have shown that quartz resonators can be used to study friction in a range of high speeds and high frequencies and thereby supplement other experimental techniques like the surface forces apparatus or the atomic force microscope in the study of tribology. As an initial step, we have clarified the consequences of an elastic contact between the quartz plate and small sphere in contact with it. The observed in-
crease in frequency is modeled by adding an element of the form $k/\omega$ into the equivalent circuit. The spring constant $k$ accounts for the added stiffness and is proportional to the contact radius and the sphere’s elastic modulus. When con-
tact is just barely established, excess dissipation of energy related to friction processes is observed. As expected, the excess dissipation is high for metal–metal contacts and lower for fluorocarbon interfaces.

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17The equation assumes an isotropic medium. We have used $E = c_{11}$ and $\rho = 1/2(\rho_1 + \rho_2 + \rho_3)$ with the numerical values from Ref. 2, p. 49.

