

# Electromagnetic Fluctuation-Dissipative Interactions of Mobile Particles and Nanoprobes Moving Near a Surface

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**Abstract.** Discussion of several experiments where could be probed the electromagnetic fluctuation-dissipative forces, is presented : measurements with the scanning probe microscopes, quartz crystal microbalance and interactions of neutral atomic beams with atomically flat surfaces. A comparison with the current theoretical models is given. It is shown that yet we can not relate the observed dissipative forces with the theoretical predictions with a confidence. Conditions where the fluctuation forces could be characterized more clearly, are proposed.

## Introduction

How do loose energy nanoscale objects in relative motion ? What are the involved dependencies on the separation, velocity, temperature and material properties ? These are the key questions of the macro-/nanotribology and many other important topics, such as adsorption/desorption processes on surfaces and damping motion of adsorbates, physical properties of thin films, the Brownian motion and energy loss of slow and fast charged and neutral atomic particles moving in close vicinity to a surface, the friction of nominally flat surfaces, heating effects in nanostructures via evanescent fields, etc.

Nowadays, great practical significance of these issues is stimulated by needs of the scanning probe microscopy [1-3], the quartz - crystal microbalance [4] and the surface force apparatus technique [5], frictional drag experiments between (2D) electron systems [6], possibilities of transmission and manipulation thin particle beams using nanochannels [7,8], energy modulation of particles in evanescent fields [9, 10], etc.

Even a short discussion of the above questions needs too much time to be presented here. We have a rather modest aim speaking about fluctuation dissipative forces (FDF) measured in recent experiments with the help of scanning probe microscopy (SPM), quartz crystal microbalance (QCM), beam interactions with surfaces, and making comparison with theoretical predictions. Also, we want to draw attention to some possible perspectives for future investigations. These issues seem to be mandatory because earlier and present day theoretical investigations of FDF have led to many controversial issues in publications of different authors (see [11] and references therein). Due to the lack of time, we leave aside the measurements of conservative van der Waals (vdW) forces .

## Theoretical Outline

An atomic or nanoparticle approaching a polarizable substrate, may engender collective excitations which act back upon the particle. Both virtual and real excitations occur, and the resulting interactions thereby has

both conservative and dissipative components. Consider, for example, a metal substrate and a particle moving parallel to its surface in  $x$ -direction. A moving bare charge produces the moving “image charge” on the metal surface, while a moving dipole (and high-order multipoles, both permanent and fluctuating) – the moving “image multipoles”. This motion of the induced charges is accompanied by Ohm’s law heating within the metal. Therefore, the friction is due to the conduction electrons, and the heat generated inside the metal is the source of this friction force on the particles outside the metal surface [12].

In the nonretarded and nonrelativistic case, the order of magnitude of attractive and drag friction forces for different particles can be determined from a simple analysis of physical dimensions. The needed quantities are: the height of the particle above the surface  $z_0 \sim (cm)$ ; the charge  $Ze \sim (gm^{0.5} cm^{1.5} sec^{-1})$ ; the dipole moment  $d \sim (gm^{0.5} cm^{2.5} sec^{-1})$ ; the conductivity of substrate  $\sigma \sim (sec^{-1})$ ; the particle velocity  $V \sim (cm/sec)$ ; the (spherical) particle radius  $R \sim (cm)$ ; the attractive and friction forces  $F_z, F_x \sim (gm cm sec^{-2})$ . One should make difference between the quantum-induced fluctuation forces, and thermal-induced ones. So, in the last case for a neutral spherical particle we get the following expressions for normal (tangential) force and heat flow :

$$\begin{aligned}
 F_z &\sim \frac{(k_B T) R^3}{z_0^4} & (a) \\
 F_x &\sim \frac{(k_B T) R^3 V}{z_0^5 \sigma} & (b) \\
 \dot{Q}^{\text{fl}} &\sim k_B T \frac{R^3}{z_0^3} \sigma & (c)
 \end{aligned} \tag{1}$$

where  $k_B$  is the Boltzmann’s constant and  $T$  is the particle temperature (the surface is assumed to be “cold”:  $T = 0$ ). Obviously, the dependence  $F_x \sim 1/\sigma$  is a simple consequence of the Ohm’s law. For normal component of the quantum-induced fluctuation force  $F_z$ , one has to replace  $k_B T$  by  $\hbar \omega_0$ , ( $\omega_0$  is the characteristic absorption frequency), but the corresponding tangential component  $F_x$  can not be obtained in such a simple manner. Formulae (1) give the first terms of the corresponding velocity expansions in powers of  $\lambda^2 = (V/z_0 \sigma)^2$ . Therefore, if  $\lambda \ll 1$ , (1) allows to get correct results. By the way, the applicability of the nonretarded approximation implies  $z_0 < c/\omega_0$ , with  $c$  being the speed of light. Therefore, even for normal metals, at  $\omega_0 = 10^{16} sec^{-1}$ , we get  $z_0 < 30 nm$ , while for more resistive materials the nonretarded approximation is valid at much greater distances. Anyhow, the nonretarded and nonrelativistic approximation is applicable in the experimental situations which we address to in this paper.

The expression for  $\dot{Q}^{\text{fl}}$  shows that heat exchange through evanescent modes of fluctuating electromagnetic field, where a small particle is placed in close vicinity to a surface, may significantly differ from that one due to black body radiation, being independent of distance and having another temperature dependence ( $\dot{Q}^{\text{fl}} \sim T^4$ ).

Certainly, Eqs.(1) describe the simplest case, while in general one the corresponding dependencies on material properties and the probe’s curvature radius are more complex [11]. Thus, for the paraboloidal SPM tip of radius  $R$ , to linear – velocity order, the thermal-induced FDF can be written in the form

$$F_x = -C R V / z^3 . \tag{2}$$

where  $z$  is the tip – sample clearance, the damping constant  $C$  depends on temperature, material properties and may correspond to both frictional ( $C > 0$ ) and accelerative ( $C < 0$ ) tangential force. The

last case takes place when the dielectric response functions characterize overlapped absorption peaks of the materials in contact. A possibility for the moving (neutral) particle to be heated is due to the fact that it has the internal degrees of freedom, while the bare charge and dipole molecule (without of the dipole moment reorientation) undergo only frictional forces in close vicinity of the surface.

In addition, in the nonlinear – velocity regime, a possibility of resonance particle – sample coupling occurs [11]. The corresponding conditions implies relation  $v \sim z_0 \Delta\omega$ , where  $\Delta\omega = |\omega_s - \omega_p|$ ,  $\omega_s$  and  $\omega_p$  are the characteristic frequencies of the surface and particle, respectively. In this case formulae (1) become incorrect because  $\lambda \sim 1$ , while the corresponding tangential force and heat flux are scaled as  $F_x \sim T / z_0^4 \Delta\omega$ ;  $Q \sim T / z_0^3 \Delta\omega$  (if  $\omega_s, \omega_p < k_B T / \hbar$ )

## Discussion of Experiments

**Measuring of dissipative forces in modulation mode of scanning probe microscopes.** Let us discuss the results recently obtained in [13-15]. It is worthwhile noting that in real experimental situation different mechanisms contribute the damping of vibrating nanoprobes. To date, there is no clear distinction between them and much work must be done in order to reach clearer understanding of the involved processes. For instance, in modulation mode (tapping mode) SPMs, an important role may have energy dissipation due to breaking/forming of the adhesional bonds (even in the vdW region) – a mechanism being more representative for the contact mode SPMs [16, 17]. Other possibilities have been discussed in [18, 19]. Below we neglect any of them.

In [13], the dissipative forces were measured in case of a silicon probe oscillating along a normal to mica surface (in UHV at  $T = 300 K$ ). The cantilever had stiffness  $k = 40 N/m$ , the curvature radius of the tip was  $R = 20 nm$ , the set point amplitude  $A = 32 nm$ , the quality factor  $Q = 22815$ , and the ground frequency  $f = 296.6 kHz$ . From this work, the mean dissipation power was equal to about 0.1 and 0.02  $pW$  at the minimum approach distances to the surface of 0.1 and 0.5  $nm$ , respectively. Therefore, the corresponding distance dependence is scaled as  $\bar{P} \propto h^{-1}$ . For comparison, the internal dissipation power of free vibrations is estimated to be  $P^{(i)} = \pi k A^2 f / Q = 1.7 pW$ .

Assuming the corresponding force to be defined by (2), and the harmonic cantilever movement  $z(t) = A \cos(2\pi f t)$ ,  $\mathfrak{z}(t) = -2\pi f A \sin(2\pi f t)$ , the mean dissipation power is given by

$$\bar{P}(d) = f \int_0^{1/f} F(t) \mathfrak{z}(t) dt = 4\pi f^2 A^{-1} C \int_0^\pi \frac{\sin^2(x)}{(d/A + \cos(x))^3} dx = \frac{2\pi^2 f^2 C}{A((d/A)^2 - 1)^{3/2}}. \quad (3)$$

where  $d$  is the cantilever support distance. The minimal approach distance is  $h = d - A$ , so at  $h \ll A$  from (3) we get

$$\bar{P}(h) = \sqrt{\frac{A}{2}} \pi^2 f^2 C / h^{3/2}. \quad (4)$$

Then, using the above estimates for  $\bar{P}$  at  $h = 0.1$  and  $h = 0.5 nm$ , we obtain  $C = 4.5 \cdot 10^{-28}$  to  $10^{-27} J \cdot sec$ .

On the other hand, in the case of silicon-mica contact we have [11]

$$C = \frac{27}{4} k_B T \frac{(\epsilon - 1) \tau}{(8\pi \sigma \tau + 3(\epsilon + 1))^2}. \quad (5)$$

where  $\sigma$  is the tip conductance,  $\epsilon$  and  $\tau$  are the substrate static dielectric permittivity and relaxation time, corresponding to the Debye approximation. From (5) we get the maximal  $C$  at

$$\sigma \cdot \tau = 3(\epsilon + 1) / 8\pi. \quad (6)$$

Using (5),(6) yields  $C = 0.08k_B T \tau$ , then at  $\epsilon = 6$  (which is typical to mica), to fit the experimental data we must assume  $\tau = 1.4 \cdot 10^{-6}$  to  $3 \cdot 10^{-6}$  sec and  $\sigma = 6 \cdot 10^5$  to  $2.7 \cdot 10^5 \text{ sec}^{-1}$ . The obtained parameters seem to be somewhat problematic being addressed to usual characteristics of pure silicon and mica at room temperature :  $\sigma = 10^{10} \text{ sec}^{-1}$  and  $\tau = 10^{-9} \text{ sec}$ , respectively. With these material parameters, the friction force will be a factor  $10^{-3}$  less than the experimental estimates. However, one should bear in mind that (5) defines only a part of the damping coefficient due to absorption in the low frequency range of electromagnetic spectra.

In case of parallel vibration of the nanoprobe, the mean dissipation power is given by  $(x(t) = A \cos(2\pi f t))$

$$\overline{P}(h) = \frac{C}{h^3} f \int_0^{1/f} x^2 dt = \frac{2\pi^2 A^2 f^2 C}{h^3}. \quad (7)$$

Therefore, at  $h = 0.5 \text{ nm}$  and the same experimental conditions, we would get  $\overline{P} \approx 0.9 \text{ pW}$  and evidently, the dissipation would be more intensive.

In the second experiment [14], the authors measured conservative and dissipative interactions between an aluminium tip and an Au(111) surface in UHV. The used set of parameters is:  $f = 267.2 \text{ kHz}$ ,  $A = 24 \text{ nm}$ ,  $k = 40 \text{ N/m}$ ,  $R = 35 \text{ nm}$ ,  $Q = 19050$ . An analytical fit of the damping coefficient according to (2) give  $C R \approx 8 \cdot 10^{-35} \text{ J} \cdot \text{m} \cdot \text{sec}$ , with the experimental error of about  $\pm 40\%$ . Then, using (4), the mean dissipation power is  $0.55 \pm 0.22 \text{ pW}$  at  $h = d - A = 0.5 \text{ nm}$ .

In this case, despite good agreement with the distance dependence predicted by formula (2), the experimental results prove to be in much worse agreement with the theoretical ones. So, from the expression analogous to (5), we would get  $CR = 10^{-46}$  to  $10^{-45} \text{ J} \cdot \text{m} \cdot \text{sec}$  [11]. With account of nonlocal contribution to the metal dielectric response [11], the constant  $C$  can be several orders of magnitude larger, but nevertheless, the dissipation power seems to be negligibly small.

In paper [15], the authors studied Brownian motion of an aluminium coated nanotip near a surface of gold in UHV. The measured damping coefficient  $\mathbf{g}/m_{\text{eff}}$  at distance to the snap-on point of about  $5 \text{ nm}$  was equal to  $150 \text{ s}^{-1}$  (with the effective mass of the oscillator  $m_{\text{eff}} = 10^{-8}$  to  $10^{-10} \text{ g}$ ). The corresponding friction force ( $1.5$  to  $0.015 \text{ nN}$ ) is again very large, so that it can unlikely be attributed to the FDF. This follows both from our calculations [11], and from [19], too. However, the authors of [15] have different opinion.

Besides, we can consider possible role of the resonance coupling effect. At the experimental conditions [14], the maximal tip velocity is estimated to be  $V_m = 2\pi f A = 0.04 \text{ m/s}$ . Then at  $z_0 = 0.5 \text{ nm}$ , the resonance dynamic condition implies  $\Delta\omega = V_m / 0.62 z_0 = 1.3 \cdot 10^8 \text{ sec}^{-1}$ . Such frequencies, in principle, may be characteristic for the acoustic surface phonons. Nevertheless, the corresponding mechanism needs to be elaborated in more details, because it necessitates electron-phonon coupling.

**Sliding friction of adsorbates (quartz crystal microbalance experiments).** In typical experimental situation [4, 20], adsorption of gas atoms onto the microbalance produces shifts in both the ground frequency  $f_0$  and the quality factor  $Q$  of the quartz oscillator. Characteristic slip time  $\mathbf{t}$  and friction parameter  $\eta$  (the shear stress per unit velocity) are determined by

$$\mathbf{d}(Q^{-1}) = 4\rho\mathbf{t}d f_0, \mathbf{h} = \mathbf{r}/\mathbf{t}. \quad (8)$$

where  $\rho$  is the mass of the adsorbate per unit area.

From [20, 21], the friction of inert gas films (mono- and bilayers) on Ag/Au(111) surfaces is characterized by  $\mathbf{t} = 2$  to  $3 \text{ ns}$ . To date, there is no well-accepted theoretical interpretation of the QCM results. So, Krim and coworkers guess that more preferable mechanism of friction is the phononic one [22, 23], while other authors [25 - 27] argue that on systems like Xe on Ag, the electronic mechanisms should dominate.

As far as electronic friction is concerned, Persson and Volokitin [24], using the high - order perturbation theory, have proposed the expression for the drag friction force of physisorbed atoms on a metal surface, which proves to be independent of temperature ( $F_x \sim V / z_0^{10}$ ). Using this expression and the time-dependent local density approximation, Liebsch [28] obtained  $\tau = 3 \text{ ns}$  at  $z_0 = 0.24 \text{ nm}$ . In this case  $z_0$  is assumed to be the separation from the metal jellium edge. Persson [25] has found  $\tau = 25 \text{ ns}$  at the same  $z_0$ , making use a simpler model of the surface, while from the corresponding QCM observations it follows  $\tau = 1 \text{ ns}$  [29]. One can note a reasonable agreement between the experiment and the theory, but the dependence  $F_x \sim z_0^{-10}$  seems to be extremely sensitive to even small variations of the distance. Evidently, since we do not control  $z_0$  independently, the theoretical estimations cannot be accepted with a confidence. For instance, an obvious shortcoming of the theory is due to its inability to describe dependence of the slip time on the adsorbate coverage.

Another point of ambiguity of the electronic friction models is related with recent results [30], where the authors obtained a new formula for the friction coefficient (see Eq.(42)). According to this one, the slip time is estimated to be  $\tau \sim z_0^8 (\sigma / T)^2$ , where  $\sigma$  is the surface conductance. Then in the case where the temperature is tuned to the conductivity so as to give maximum friction for Xe physisorption, at  $z_0 = 0.24 \text{ nm}$ , we get  $\tau = 2.4 \text{ ns}$ . In drastic contrast to [24], the above formula is very sensitive to temperature and surface resistivity.

It is worthwhile noting in this turn, that one should also take account of an important linear - order (in polarizability and electric field) contributions to the friction force, which are determined by (1b) or more general formulae [11], when the dependence  $\tau(z_0)$  is less strong ( $\tau \sim z_0^5 / T$ ), and what is more important – the slip time depends on structure of the adsorbed film via the dielectric response function. The corresponding effects are important even at small temperatures of the order of  $1 \text{ K}$ .

For instance, a solid Xe film is known to have strong exciton absorption bands and those ones due to intraband transitions [31]. On the other hand, the solidified film must be characterized by new phononic modes, and when cooling (below freezing point), this may lead even to positive contribution to the tangential force (under sufficient electron-phonon coupling), if the film temperature is lower than that of the surface [11]. Such an effect can be, in principle, responsible for small friction of solidified incommensurate films [32], because any structural transformations caused by heat transfer influence the dielectric response functions and, therefore, modify FDF. However, yet we did not clearly understand role of heating effects related with evanescent fields in these 2D-systems. For instance, for resistive materials, which for Eq.(1c) is valid, the heat flow can be by about 9 orders of magnitude higher than that from black body radiation (see also [32, 33]).

At last, we shall briefly touch on recent QCM experiments on superconducting surfaces [34]. It was found that friction of solid nitrogen film drops abruptly (by about two times as compared to the normal state value) below the transition temperature (7.2 K) of lead. Evidently, this requires significant role of the electronic friction processes in this system [35]. The authors note that their theory cannot explain this experiment, because the electronic sliding friction decreases continuously when the system is cooled below  $T_c$ , in a way correlating with the fraction of electrons in the superconducting condensate. In our opinion, this conclusion follows from oversimplified character of the used model, relating the electronic friction exclusively with the  $dc$ -resistivity. However, in these systems an essential role may have electron-phonon coupling [34]. Really, the phonon frequencies ( $10^{12}$  to  $10^{13}$   $\text{sec}^{-1}$ ) exactly coincide the energy gap of lead:  $\omega_c = 4k_B T_c / \hbar = 3.8 \cdot 10^{12}$   $\text{sec}^{-1}$ . If, in normal state, there is a noticeable contribution to the energy dissipation from this frequency range, then below  $T_c$  we must observe a drop of the friction force and increasing the slip time. In favour of such a possibility one can mention the experimental results on energy dependence of the density states,  $N(\mathbf{w})$  for lead [36], where the nonmonotonous character of the  $N(\mathbf{w})$  has been clearly seen at  $\mathbf{w} > 2k_B T_c$ .

Still more intriguing results are to be expected when studying electronic friction on surfaces of high temperature superconductors, where a great variety of interesting features of the involved dielectric and absorption properties have been observed (see, for instance, [37] and references therein).

**Experiments on passage of neutral atomic beams near a surface.** By the way, the velocity-dependent effects may well become more significant for higher beam energies and particle velocities of the order of Fermi velocities in metals [38]. In this work we have calculated the expected drag force and stopping power for a well-collimated neutral beam of helium atoms passing in close vicinity to aluminium surface. A possible experimental arrangement similar to that has been exploited when measuring transmission of electrons through thin microchannels in metal foils (of 20-200 nm in diameter) [9] or, for instance, when the atomic beam moves above the metal (semiconductor) bar deposited on a substrate.

Unfortunately, in our previous calculations [38] the temperature factors have not been taken into account correctly. In view of the present day results, we do not expect a noticeable contribution of the surface plasmon coupling (for substrates of normal metals). However, the similar effects might be observed due to a resonance with the low-frequency surface excitations like surface plasmons in doped semiconductors and polaritons in dielectric materials. Also, the resonance stopping effects must be important for hot neutral molecules and clusters above cold substrates. The higher is the temperature  $T_1$  of the particles, the more of their inner states will satisfy the frequency-tuning conditions, thus giving rise to stronger particle-surface coupling, as well.

Moreover, one should bear in mind a possibility for the low-frequency nonlocal contribution to the dissipative force [11]. For instance, for a helium atom with the velocity of  $3 \cdot 10^6$   $\text{m/s}$ , passing above aluminium surface at  $z_0 = 1$   $\text{nm}$ , the estimated energy loss will be of the order of  $0.1$   $\text{eV}/\mu\text{m}$ . Much higher energy losses are expected for neutral atomic beams above the surface of poor conductors and dielectrics.

Evidently, in real experimental situation, some part of an output beam will be ionized, but atoms which remain in neutral state, would be characterized by negative or positive energy shifts differing from those of the outgoing ions. Consequently, the total energy distribution spectrum of the particles must reveal these features.

## Summary

In summary, we have shown that to date we have no clear experimental evidence in favour of the fluctuation-induced friction forces. The observed values of the dissipative forces in SPM-experiments are

much greater than theoretically expected FDF: for silicon(tip) – mica (surface) – by about  $10^2 \div 10^3$  times, for contact of normal metals – by about 10 orders of magnitude. There are strong indications that the electronic friction models should incorporate the temperature and structural effects while being applied to the QCM - experiments in an unambiguous way. In order to clearly see role of FDF in SPM and QCM experiments, one needs to study the characteristic dependencies of the dissipative forces on distance, temperature, geometrical and material parameters. Regarding experiments with SPMs in dynamic regime, we draw attention to lateral vibration mode, where the tip moves at a constant height from the surface. Another possibility to study FDF is related with measurements of the energy transmission spectra of neutral atomic(molecular) beams in close vicinity of atomically smooth surfaces. More preferable is using of semiconductors and dielectric materials. In conclusion, we guess that development of experimental technique presents a great challenge for studying properties of materials by measuring the fluctuation electromagnetic forces on the nanoscale.

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