Brownian Motion of Microscopic Solids under the Action of Fluctuating Electromagnetic Fields

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The Brownian motion of a microscopic solid under the action of fluctuating electromagnetic fields was detected using atomic-force microscopy. The distance dependence of the noise spectrum of free cantilever oscillations, of the resonance frequency, and of the damping coefficient were investigated under ultrahigh vacuum conditions. An analytic expression for the damping coefficient of a metallic tip–sample system was obtained on the basis of fluctuating electrodynamics. Our calculation is in good agreement with experimental data.

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The Brownian motion of small particles in liquids was discovered by Robert Brown in 1827 as a direct manifestation of molecular chaos. The phenomenon was explained by Einstein and by Smoluchowski [1,2] in terms of random fluctuations of a particle coordinate under the action of stochastic impacts of molecules or inhomogeneities in liquids or gases. For an elementary charge a relevant solution of the problem of Brownian motion of an electron under the influence of thermal fluctuating fields taking into account a radiative reaction force may be found in textbooks (see, for instance, Ref. [3]). Experimentally and theoretically, scattering of electrons by thermal electromagnetic fields near a solid surface was investigated in Ref. [4]. Up to this day, the problem of stochastic motion of charged or neutral small bodies under the action of random forces of any nature is an interesting topic in science, in particular, in nanophysics.

The development of scanning probe microscopies induced many successful applications of local probe methods in various fields of science and technology. As an example, the highly sensitive atomic force microscopy (AFM) was used in Ref. [5] to study the tip-sample interactions by means of noise spectra analysis of stochastic cantilever motion.

Here we present the first results of the investigation of a freely vibrating motion of an AFM cantilever near a clean solid surface under ultrahigh vacuum conditions. Variations of the resonance frequency and the damping coefficient, as a function of the distance between a tip and sample surface, were detected. It will be shown that the experimental results may be explained using the concept of Brownian motion of a microparticle under the action of thermal fluctuating electromagnetic fields generated by the sample being investigated. For a metal-metal system an analytical formula for the damping coefficient was obtained which is in good agreement with experimental results.

A general theory of equilibrium thermal fluctuations of an electromagnetic field was published by Rytov [6] and further developed in Refs. [3,7]. A fluctuating electromagnetic field is induced by random charges and currents which result from quantum or thermal fluctuations. This field is present inside any absorbing matter and also outside of it, as progressive waves radiated by a body and as nonradiating states exponentially decaying with the distance from a body, the latter representing quasistationary or evanescent fields. An interesting extension of the theory was provided by Carmanati and Greffet [8].

The results of Rytov's theory are valid for any ratio of the wavelength of an electromagnetic field λ and the characteristic spatial scale d of a system. This theory contains Planck's law as an asymptotic case for $\lambda \ll d$. In accordance with Planck's law, the spectral density of energy of the radiative part of a thermal electromagnetic field is given by $u_{0_w} = \omega^2\Theta(\omega, T)/\pi^2c^3$, with $\Theta(\omega, T) = (\hbar\omega/2)\coth(\hbar\omega/2k_BT)$, where k_B is the Boltzmann constant, T is the absolute temperature, and c is the speed of light in vacuum. The energy distribution of the spectrum of blackbody radiation is independent of the properties of the material emitting this radiation. However, in close proximity to the sample surface or in small volumes, when $\lambda \leq d$, the spectral composition of the thermal fluctuating fields is changing significantly. For example, in Refs. [3,6,7] it was shown that, at the distance d from the boundary of a half-space filled with a material with a dielectric constant $\epsilon(\omega) = \epsilon'(\omega) + \imath\epsilon''(\omega)$, the spectral energy density of an electromagnetic field generated by such a sample for $kd = 2\pi d/\lambda \ll 1$ is proportional to $(kd)^{-3}$. As in the low-frequency limit for dielectrics $\epsilon'(\omega) \sim \epsilon_0^{'}$; $\epsilon''(\omega) \sim \epsilon_0^{''}\omega$, where $\epsilon_0^{'}$ and $\epsilon_0^{''}$ are real numbers, we have

$$u_w \sim \frac{\epsilon_0^{''}k_BT}{d^3(1 + \epsilon_0^{'})^2}. \tag{1}$$

The asymptotic result (1) is valid only for poor conductors where $|\epsilon| \geq 1$. In analogy, from the results of Refs. [3,6,7] it is possible to find the low-frequency limit for good conductors, where $|\epsilon(\omega)| \gg 1$ and $\epsilon(\omega) \simeq \imath(4\pi\sigma/\omega)$, with σ the conductivity of the material. For the same case $kd \ll 1$, we have

$$u_w \sim \frac{ck_BT}{d^4\sqrt{\sigma\omega^3}}. \tag{2}$$

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In this formula, ω is restricted at the low values, because for good metals the thickness of a skin layer $\delta = c/\sqrt{2\pi\sigma\omega} \ll \min\{d, R, h\}$, where R, h are the radius of curvature and the thickness of a sample. From (1) and (2) it follows that the quasistationary (evanescent) part of the thermal field contains information about the electrodynamical properties of materials, in contrast to the case of the radiational part of the thermal field. These formulas testify that the spectral density energy of thermal fields is shifted to the low-frequency part of the spectrum and sharply increases with decreasing distance from the sample surface. In the high-frequency limit for any material the spectral density of energy decays exponentially with the frequency.

The Brownian motion can only be detected for very small particles, since for massive bodies the random force action of molecules will be averaged out. Therefore, we tried to register the Brownian motion of a microscopic solid (for example, a micrometer-sized probe of an atomic-force microscope located at a distance d from a sample surface). This seems appropriate since, as indicated by (1) and (2), the energy of a thermal fluctuating electromagnetic field increases near a solid surface, especially at the low-frequency part of spectrum, including the typical range of mechanical frequencies of AFM cantilevers, i.e., 10^3–10^5 s^{-1}.

Our calculations aim to estimate the damping coefficient of a system consisting of two metallic solids separated by a vacuum gap. Let us consider our system as a discrete dissipative system. The fluctuations of the vertical coordinate of a cantilever are described by the function $z(t)$. These fluctuations are induced by external stochastic forces $f(t)$. The relevant part of the tip that is involved in van der Waals (vdW) interactions originates from an apex volume approximately determined by the tip radius, $p \approx 10^{-5}$ cm. As the wavelength of an electromagnetic fluctuating excitation is much larger than the radius of the tip, we will use a simple oscillator model of the system as described by the Langevin equation,

$$m_{\text{eff}} \frac{d^2z}{dt^2} + \gamma \frac{dz}{dt} + m_{\text{eff}}\omega_0^2 z = f(t), \qquad (3)$$

where m_{eff} is the effective mass of an oscillator, γ is the friction parameter, and ω_0 is the eigenfrequency of the system. The Fourier transform $z(\omega)$ is connected with the Fourier transform of a random force $f(\omega)$ by the well-known expression $z(\omega) = \alpha(\omega) f(\omega)$, where $\alpha(\omega)$ is the susceptibility of the system. The power density spectra $\langle |z(\omega)|^2 \rangle$ and $\langle |f(\omega)|^2 \rangle$ of the stochastic processes $z(t)$ and $f(t)$ are expressed via the susceptibility of the system in accordance with the fluctuation dissipation theorem (FDT) [9].

From (3) and FDT, it is easy to find the expressions for the susceptibility

$$\alpha(\omega) = \frac{m_{\text{eff}}(\omega_0^2 - \omega^2) + \imath\gamma\omega}{m_{\text{eff}}^2(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2}. \qquad (4)$$

Thus an analytical expression for the shape of the resonance peak in frequency space [power density spectrum (PDS)] can be given,

$$\langle |z(\omega)|^2 \rangle = \frac{k_B T \gamma}{\pi[m_{\text{eff}}^2(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2]}. \qquad (5)$$

Then, from (3), (4), and FDT we have a general formula for the friction coefficient

$$\gamma = (k_B T)^{-1} \int_0^\infty \langle f(t) f(0) \rangle \, dt. \qquad (6)$$

In our experiments a commercial UHV AFM (Omicron) operated at room temperature was used. All presented data were obtained at a background pressure of less than 5×10^{-10} mbar. The cantilever was a standard 320-μm-long SiN contact triangular-shaped cantilever (Park Scientific Instruments) with a spring constant of $k = 0.01 \pm 0.005$ N/m. An Au(111) crystal was used as a sample, sputtered and tempered two days before the measurement. The AFM tip was sputtered and coated with at least 10 nm of Al to avoid residual charges. A schematic diagram of the experiment is shown in Fig. 1. The cantilever near the surface is driven by intrinsic thermal, quantum stochastic, and electromagnetic forces. After detection the signal is transformed in real time to its power density spectrum via fast Fourier transform using a commercial spectrum analyzer (LeCroy 334A, 500 MHz). A single measurement consists of a series of spectra obtained at different distances from the surface. The resonance peaks at different distances from the sample surface are presented in Fig. 2. These peaks are described by Lorentzian curves. The χ^2 fits on the data were done to extract all parameters available from the resonance peaks (5). The amplitude $A = k_B T \gamma / \pi m_{\text{eff}}^2$, the damping coefficient γ/m_{eff}, and the resonance frequency $f_0 = \omega_0/2\pi$ show a significant distance dependence. In Fig. 3 the dependence of the

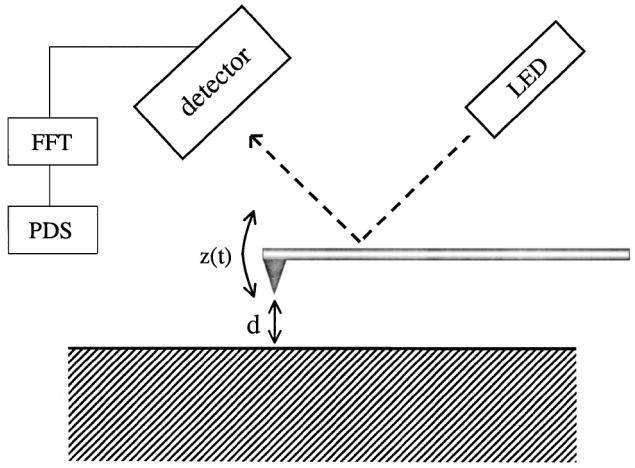

FIG. 1. Sketch of the experimental setup. The signal proportional to $z(t)$ is detected via a 4-quadrant detector and transformed into its PDS via FFT in real time.

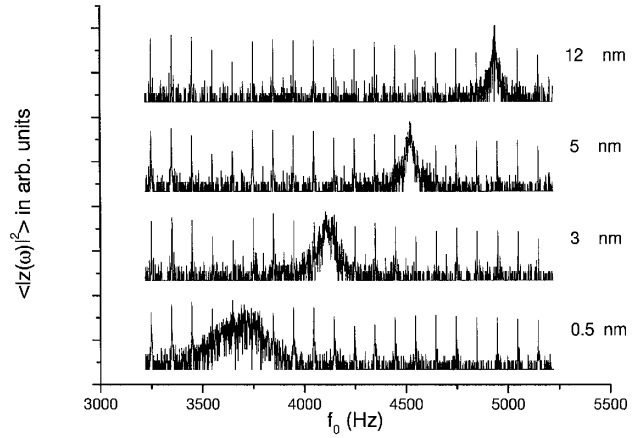

FIG. 2. Typical noise spectra at different distances from the surface. The resonance frequency distance dependence and the broadening of the resonance peak can be seen. The sharp peaks correspond to electronic noise. The distances given in the figure correspond to the snap-on point as a reference point. $z^2(\omega)$ is given in the spectrum analyzer's intrinsic logarithmic scale for the PDS.

damping coefficient on the distance d is shown. These data were well reproduced in several measurements.

In order to evaluate the influence of surface residual potentials, force-voltage measurements were done at different distances from the surface to find the force-voltage minimum. The voltage which corresponds to the minimum of these curves and which compensates all residual potentials is equal to 260 ± 50 mV.

Any distance-dependent measurements were done from approximately 100 nm to the point of the jump in the cantilever to the surface ("snap-on" point). The snap-on point is determined by calculating the effective spring constant and f_0 under the assumption of a simple $1/d$ dependence of the interaction energy (see Ref. [10]).

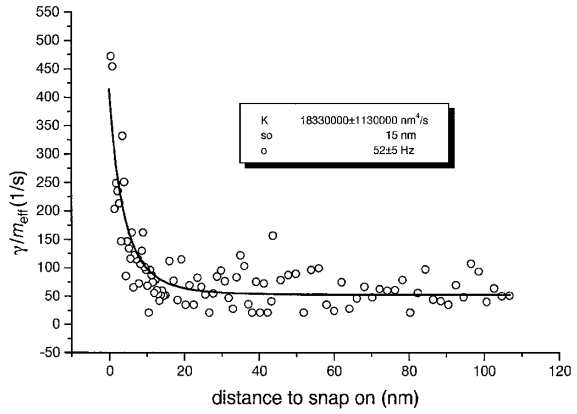

FIG. 3. Damping coefficient γ/m_{eff} vs distance to the snap-on point. The three closest points to the snap on are not included in the fitting of the functional dependence of γ/m_{eff}, but they are shown in the plot. Any measurements of distance have an error of up to 25% of the absolute value. The distance dependence is corrected for cantilever deflection.

Thus, the experimental f_0 versus distance curve can be fitted according to this expression with the snap-on point as a fitting parameter. Clearly, the harmonic oscillator model breaks down for distances very close to the snap-on point. In this case, the shape of the resonance peak is no longer Lorentzian.

We obtain an explicit form of the friction parameter for the case of a vibrating metal tip over the planar metal half-space under the action of an external force using an expression for the temporal variation of a total energy in the system given by

$$\frac{dE}{dt} = f(t)v - 2\Phi\,, \tag{7}$$

where v is the velocity of the tip and $\Phi = \gamma v^2/2$ is the dissipative function. Both the tip and the planar sample are assumed as electrically neutral but they may interact via vdW forces.

The thermal fluctuating electromagnetic fields of the half-space induce random charges and currents into the apex of the tip moved close to the sample surface. As a consequence, inside the neutral system the statistical average value, for example, the charge density $\langle \rho(\vec{r},t)\rangle$, is identically equal to zero, but the instantaneous value $\rho(\vec{r},t)$ and the mean square value $\langle |\rho(\vec{r},t)|^2\rangle$ is finite.

At first we consider the forced harmonic movement of a particle over the sample with the velocity $v(t) = \mathrm{Re}[v(\Omega)\exp(\imath\Omega t)]$, where Ω is the frequency of the cantilever near the surface sample. Because of this movement, the additional instantaneous current $j(\vec{r},t) = \rho(\vec{r},t)v(t)$ will appear in our system. This will induce the image current $j(\vec{r},t) = \tilde{\rho}(\vec{r},t)v(t)$ inside the metal sample, where $\tilde{\rho}(\vec{r},t) = -\rho(\vec{r},t)$. The Joule dissipation of the current $\tilde{j}(\vec{r},t)$ determines the losses in our system. This situation is similar to that of a free charge moving in an electrostatic field between the electrodes of a plane capacitor. If the material of the electrodes is not a perfect metal, the induced image current in the electrodes leads to the Joule dissipation of energy, and the movement of the charge is not conservative.

We identify the Joule losses Q_J of the image current $\tilde{j}(\vec{r},t)$ inside the metal half-space with the losses in the system of a vibrating cantilever near the sample with $Q_J = \langle 2\Phi\rangle$. By definition we have

$$Q_J = \int_V \left\langle \frac{\tilde{j}^2(\vec{r},t)}{\sigma_s} \right\rangle dV\,, \tag{8}$$

where σ_s is the conductivity of the sample. The integration is done over the volume of the induced image current. Using Fourier transforms, it can be shown that $\tilde{j}(\vec{r},\omega) = v(\Omega)[\tilde{\rho}(\omega+\Omega) + \tilde{\rho}(\omega-\Omega)]/2 \approx \tilde{\rho}(\omega)v(\Omega)$, if $\omega \gg \Omega$ for typical frequencies of cantilever vibrations of atomic-force microscopies. The approximation $\tilde{j}(\vec{r},\omega) \approx \tilde{\rho}(\omega)v(\Omega)$ implies a separation of cantilever vibrations and electromagnetic excitations in the system and allows us to obtain the necessary form of losses in an

easy way. Then, taking into account the expansion over the positive frequencies only, we have

$$Q_J = 2v^2(\Omega)\,\mathrm{Re}\int_0^\infty \int_V \frac{\langle|\tilde{\rho}^2(\vec{r},\omega)|^2\rangle}{\sigma_s}\,d\omega\,dV\,. \quad (9)$$

A comparison with the equality $Q_J = 2\Phi = \langle \gamma v^2\rangle$ results in the expression for the friction parameter

$$\gamma = 2\,\mathrm{Re}\int_0^\infty \int_V \frac{\langle|\tilde{\rho}^2(\vec{r},\omega)|^2\rangle}{\sigma_s}\,d\omega\,dV\,, \quad (10)$$

Now, the problem is to find the mean square values of the induced image charges $\langle|\tilde{\rho}^2(\vec{r},\omega)|^2\rangle$.

To do this, we use the solution from Ref. [3], of the problem of current induction inside a thin metallic rod by thermally fluctuating electromagnetic fields of surrounding solids. In the case where the radius of the cylindrical rod p (this is the radius of curvature of the moving probe) is much smaller than the wavelength of the thermal fields, it is possible to extract the mean square value of the induced current $I(\omega)$ for the case $kd \ll 1$,

$$\langle|I(\omega)|^2\rangle \approx \frac{3p^2c^3\sigma_t\Theta(\omega,T)}{4\omega^{5/2}d^4\sqrt{8\pi\sigma_s}}\,, \quad (11)$$

where we used a formula for the specific impedance of a cylindrical rod of radius p in considering the normal skin-effect; σ_t is the conductivity of the rod (tip).

To obtain the value $\langle|\rho(\vec{r},\omega)|^2\rangle$ for (10) we use the continuity equation $div\,j(\vec{r},\omega) = \imath\omega\rho(\vec{r},\omega)$. An integration over the tip apex and the multiplication to the complex conjugated value followed by statistical averaging yield

$$\langle|I(\omega)|^2\rangle = \omega^2\int\int\langle|\rho(\vec{r},\omega)|^2\rangle\,dV\,dV'. \quad (12)$$

After substitution of (11) and (12) into (10) we have

$$\gamma \approx \frac{3c^3\sigma_t}{2pd^4\sqrt{8\pi\sigma_s^3}}\int_{\omega_2}^{\omega_1}\Theta(\omega,T)\omega^{-9/2}\,d\omega\,, \quad (13)$$

where we assumed $V \sim p^3$. The integration in (13) is restricted over the frequencies from ω_2 ($\omega \gg \Omega$) up to ω_1 because $\omega \ll \sigma$ in the model for the dielectric constant for good metals, $|\epsilon| \gg 1$. Finally, up to a constant, which is taking into account the real geometry of a probing tip, and in the case $\hbar\omega \le k_BT$, we have

$$\gamma \approx \frac{\sigma_t c^3 k_B T\tau^{7/2}}{pd^4\sqrt{8\pi\sigma_s^3}}\,, \quad (14)$$

where $\tau = \omega_2^{-1}$ is the free parameter of the problem. It is evident that in our consideration we are neglecting the thermal fields of the tip itself compared to fields of a half-space. From (13) it is easy to obtain a related formula in the case $T = 0$.

The damping coefficient γ/m_{eff} from (14) is fitted to our experimental results using

$$\frac{\gamma}{m_{\mathrm{eff}}} = \frac{K}{(x + s_o)^4} + \gamma_0\,, \quad (15)$$

where K includes all factors from (14), $x + s_o = d$ in nm, s_o is the snap-on point, and γ_0 corresponds to the intrinsic damping of the cantilever, when $d \to \infty$. The snap on is not a real fitting parameter, because it is determined as described above and not changed for a given experiment. The result of the fitting procedure is shown in Fig. 3 by a solid curve. This fit was done for our experimental parameters $T = 300$ K, $k_BT = 4,\ 1 \times 10^{-14}$ erg, $c = 3 \times 10^{10}$ cm/s, $p = 10^{-5}$ cm, $\sigma_s = 3 \times 10^{17}$ s^{-1}, $\sigma_t = 10^{16}$ s^{-1}, and $m_{\mathrm{eff}} = 10^{-8}$–$10^{-10}$ g. For these values of m_{eff} we have $\tau \approx 10^{-12}$–10^{-13} s, close to the relaxation time of electrons in metals. Taking into account that (14) is valid up to a constant, it should be regarded as a good coincidence.

In conclusion, we discussed the Brownian motion of a microscopic solid under the action of fluctuating electromagnetic fields under ultrahigh vacuum conditions using atomic-force techniques. From an analysis of the noise spectrum of cantilever vibrations, the functional dependencies of resonance frequency and the damping coefficient on the size of the vacuum gap were obtained. The expression for the damping coefficient of a tip-sample system for metals was obtained on the basis of fluctuating electrodynamics, in good agreement with the experimental data.

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