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Fluctuational Electromagnetic Interaction Between a Moving Particle and a Flat Surface Covered with a Thin Adsorbed Film

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Abstract—The effect of a thin adsorbed film on the fluctuational electromagnetic tangential force of interaction between a moving nanoparticle and a flat solid surface was theoretically studied for the first time in a nonrelativistic approximation. Particular calculations were performed for a metal film on a dielectric and for a dielectric film on a metal. In both cases, the nanoparticle is assumed to be madder of a nonmagnetic metal. It is shown that, at a normal temperature, the presence of an adsorbed film may lead to an increase in the tangential friction force by one to two orders of magnitude for a certain relation between the particle distance from the surface and the film thickness. In the case of a dielectric film on a metal substrate, a decrease in the temperature is accompanied by exponential decrease in the viscous friction. For a metal film on a dielectric substrate, the tangential force exhibits a quadratic dependence on the temperature. © *2004 MAIK "Nauka/Interperiodica".*

In practical applications related to interpretation of the interaction between the probe of an atomic force microscope (AFM) and the sample surface, an important particular case is the substrate with a permittivity $\varepsilon_s(\omega)$ covered by an adsorbed film of thickness d and permittivity $\varepsilon_a(\omega)$ (Fig. 1). Recent measurements of the viscous dissipative force in "pure" dynamic silicon–mica [1], aluminum–gold [2], and gold–gold [3] contacts were performed at room temperature in vacuum. However, a comparison of these experimental data with the values predicted by the theory of fluctuational electromagnetic interactions revealed discrepancies reaching several orders on magnitude [4–6]. These discrepancies stimulate the search for factors missing in the theory, which would provide for a increase in the magnitude of fluctuational electromagnetic interactions. In particular, the role of heating (cooling) of the AFM probe by near fluctuational fields was recently considered in [7]. Another important factor can be the presence of adsorbed layers of foreign molecules. In particular, according to estimates [8], the presence of adsorbed K atoms at a concentration of 10^{18} m^{-2} on a Cu(001) surface leads to an increase in the coefficient of viscous friction between flat copper samples by seven orders of magnitude. It should be noted, however, that the estimation [8] was based on the theory of fluctuational dissipative interaction between semi-infinite media separated by a flat vacuum gap, while scanning probe microscopy (in particular, AFM) features the interaction between a nanoprobe with the curvature radius R and a flat surface. This system is more adequately described by the theory developed in [4–6].

Let us consider, for certainty, a probe moving parallel to a surface at a distance of z_0. We assume that the standard conditions of applicability of the nondelayed dipole approximation are satisfied [4–6], so that $R \ll z_0 \ll c/\omega_0$, where c is the speed of light in vacuum and ω_0 is the characteristic absorption frequency of the electromagnetic spectrum. The main difference of this problem from the case of interaction between a particle and a semi-infinite medium with flat boundary consists in the need for modification of the solution of the Pois-

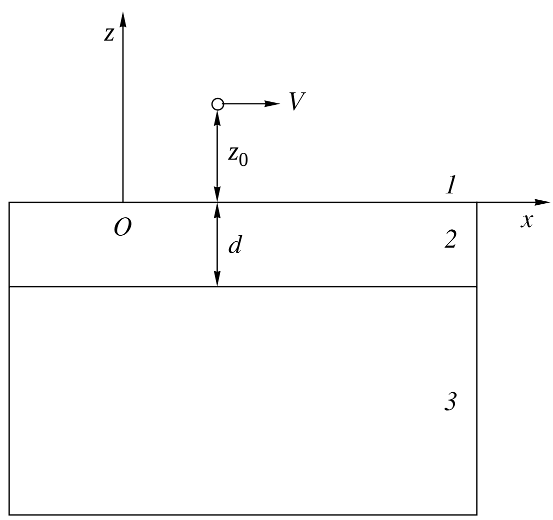

Fig. 1. Schematic diagram illustrating interaction between a particle and a surface covered by a thin adsorbed film: (*1*) vacuum; (*2*) film; and (*3*) substrate (see the text for explanations).

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son equation for the Fourier component $\Phi_{\omega\mathbf{k}}(z)$ of the electric potential induced in the medium by the moving fluctuating dipole (ω is the frequency; **k** is the two-dimensional wavevector parallel to the surface; *z* axis is perpendicular to the surface).

A solution for the function $\Phi_{\omega\mathbf{k}}(z)$ in this system (Fig. 1) constructed in the regions of vacuum (*1*), film (*2*) and substrate (*3*) must satisfy the conditions of continuity of the potential and the electric displacement at the boundaries $z = 0$ and $z = -d$. An analysis shows that all the general formulas [4–6] for the tangential force (F_x) and the rate of heating (dQ/dt) cased by the fluctuational electromagnetic field remain valid, provided that the dielectric response of the semi-infinite medium $\Delta(\omega) = (\varepsilon(\omega) - 1)/(\varepsilon(\omega) + 1)$ is replaced by the function

$$D(\omega, \mathbf{k}) = \frac{\Delta_1(\omega) - \Delta_2(\omega)\exp(-2kd)}{1 - \Delta_1(\omega)\Delta_2(\omega)\exp(-2kd)}, \quad (1)$$

$$\Delta_1(\omega) = \frac{\varepsilon_a(\omega) - 1}{\varepsilon_a(\omega) + 1}, \quad (2)$$

$$\Delta_2(\omega) = \frac{\varepsilon_a(\omega) - \varepsilon_s(\omega)}{\varepsilon_a(\omega) + \varepsilon_s(\omega)}. \quad (3)$$

As can be seen, formula (1) satisfies obvious limiting relations $D(\omega, \mathbf{k}) \longrightarrow \Delta_1(\omega)$ for $\varepsilon_a(\omega) \longrightarrow \varepsilon_s(\omega)$ for $d \longrightarrow \infty$; and $D(\omega, \mathbf{k}) \longrightarrow \Delta_2(\omega)$ for $d \longrightarrow 0$. In terms of function (1), the viscous tangential force acting upon the article can be written as (negative sign corresponds to retardation)

$$F_x = -\frac{3\hbar V}{2\pi}\int_{0,\infty}\int d\omega dk k^4 \exp(-2kz_0) f(\omega, k), \quad (4)$$

$$f(\omega, k) = \coth\frac{\hbar\omega}{2k_BT_1}\alpha''(\omega)\frac{dD''(\omega, k)}{d\omega} + \coth\frac{\hbar\omega}{2k_BT_2}D''(\omega, k)\frac{d\alpha''(\omega)}{d\omega}, \quad (5)$$

where $\hbar$ and k_B are the Planck and Boltzmann constants, respectively; $\alpha(\omega)$ is the dipole polarizability of the particle; T_1 and T_2 are the temperatures of the particle and the surface (in the general case, different); primed and double-primed quantities denote the real and imaginary components. In particular, the complex polarizability of a spherical particle of radius *R* and permittivity $\varepsilon(\omega)$ is

$$\alpha''(\omega) = R^3 \mathrm{Im}\frac{\varepsilon(\omega) - 1}{\varepsilon(\omega) + 2}. \quad (6)$$

Using relations (1)–(3), the complex displacement can be written as

$$D''(\omega, k) = \frac{\Delta_1''(1 - a^2|\Delta_2|^2) - a\Delta_2''(1 - |\Delta_1|^2)}{1 + a^2|\Delta_1|^2|\Delta_2|^2 - 2a(\Delta_1'\Delta_2' - \Delta_1''\Delta_2'')}, \quad (7)$$

where $a = \exp(-2kd)$ and the argument ω is omitted.

In the case of a particle moving perpendicularly to the surface [9], the numerical coefficient in the right-hand part of Eq. (4) is doubled. The corresponding formula, describing the dissipative part of the normal force of interaction between a particle and the surface, contains a conservative contribution related to the standard van der Waals interaction (with dynamic corrections). The corresponding expressions can be written by analogy with formula (4).

The basic difference of formula (4) from formulas for the tangential interaction of a particle with a semi-infinite medium (see, e.g., [5, Eq. (4.31)] is the dependence of $D''(\omega, k)$ on the wavevector *k* (even without taking into account the possible nonlocal character of $\varepsilon_s(\omega)$ and $\varepsilon_a(\omega)$). In the general case, this circumstance leads to a more complicated dependence of F_x on the distance z_0, differing from the law $F_x \sim z_0^{-5}$ obtained in [4, 5].

Let us apply the above results to some particular cases.

1. Metal film on a dielectric substrate. In the microwave spectral range ($\omega \approx k_BT/\hbar$) far from the phonon resonances, we obtain $\varepsilon_a(\omega) \approx 1 + 4\pi\sigma_0 i/\omega$, where σ_0 is the static conductivity (obviously, $4\pi\sigma_0/\omega \gg 1$). Taking into account Eqs. (2) and (3), we obtain $\Delta_2'(\omega) \approx 1$, $\Delta_2'(\omega) \approx 0$, and $\Delta_1(\omega) \approx 1 + \frac{\omega}{2\pi\sigma_0}$. Then, Eq. (7) yields

$$D''(\omega, k) \approx \frac{\omega}{2\pi\sigma_0}\frac{1 + \exp(-2kd)}{1 - \exp(-2kd)}. \quad (8)$$

In the case of $z_0/d \gg 1$, substitution of formula (8) into Eq. (5) shows that the force F_x increases by a factor of z_0/d in comparison to the case of a “pure” surface. On the contrary, the dependence of F_x on z_0 becomes weaker ($F_x \sim z_0^{-4}$ instead of $F_x \sim z_0^{-5}$). Under the conditions of AFM experiments, the inequality z_0/d can be satisfied only for a sufficiently large distance of the point of close contact from the samples surface. At a distance on the order of 1 nm or below, the amplification effect ceases and, hence, the presence of adsorbed layers does not (on the average) significantly influence the interaction for the probe moving perpendicularly to the surface. Note also the quadratic dependence of F_x on the temperature and a weak (if any) dependence of this force on the properties of the substrate.

2. Dielectric film on a metal substrate. In this case, $\Delta_2'(\omega) \approx -1$, $\Delta_2''(\omega) \approx 0$, and $\Delta_1(\omega) = \Delta_1'(\omega) + i\Delta_1''(\omega)$. Then, Eq. (7) yields

$$D''(\omega, k) \approx \frac{\Delta_1''(\omega)(1-a^2)}{(1+a\Delta_1'(\omega))^2 + a^2\Delta_1''(\omega)^2}. \quad (9)$$

This formula shows the possibility of a resonance for $1 + a\Delta_1'(\omega) = 0$ and $\Delta_1'(\omega) < 0$. Let $\varepsilon_a(\omega)$ to have the standard form of

$$\varepsilon_a(\omega) = \varepsilon_\infty\left(1 + \frac{\omega_L^2 - \omega_T^2}{\omega_T^2 - \omega^2 - i\gamma\omega}\right) = \varepsilon_\infty + \frac{\omega_T^2(\varepsilon_0 - \varepsilon_\infty)}{\omega_T^2 - \omega^2 - i\gamma\omega}, \quad (10)$$

where ε_0, ε_∞, are the static and optical permittivities, respectively; ω_L and ω_T are the longitudinal and transverse phonon frequencies, respectively; and $\tilde{\gamma}$ is the damping factor. Using Eqs. (2) and (10), one can readily show that the resonance condition is satisfied for two surface modes of the phonon-polariton type. The dispersion equations of these modes (with omitted terms of higher orders in small parameter $\tilde{\gamma}$) have the following form:

$$\left(\frac{\omega}{\omega_T}\right)^{(1)} = \left(p\left(1 + \frac{\tilde{\gamma}^2}{q-p}(1+\exp(2kd)/r)\right)\right)^{1/2}, \quad (0 \le k \le k_{\max}), \quad (11)$$

$$\left(\frac{\omega}{\omega_T}\right)^{(2)} = \left(\frac{p + qr\exp(-2kd)}{1 + r\exp(-2kd)} - \frac{\tilde{\gamma}^2}{q-p}(q + p\exp(2kd)/r)\right)^{1/2}, \quad 0 \le k \le k_{\max}, \quad (12)$$

$$k_{\max}d = \frac{1}{2}\ln\left(\frac{r(q-p)^2}{\tilde{\gamma}(2\sqrt{p}(q-p) + \tilde{\gamma}(3p+q))}\right), \quad (13)$$

$$p = \frac{\varepsilon_0 + 1}{\varepsilon_\infty + 1}, \quad q = \frac{\varepsilon_0 - 1}{\varepsilon_\infty - 1}, \quad r = \frac{\varepsilon_\infty - 1}{\varepsilon_\infty + 1}, \quad \tilde{\gamma} = \frac{\gamma}{\omega_T}. \quad (14)$$

Figure 2 shows an example of dispersion relations (11) and (12) for a silicon carbide film on a metal substrate, calculated for the following parameters [10]: $\omega_T = 1.49 \times 10^{14}$ s^{-1}; $\omega_L = 1.8 \times 10^{14}$ s^{-1}; $\gamma = 8.9 \times 10^{11}$ s^{-1}; and $\varepsilon_\infty = 6.7$. In this case, formula (13) yields $k_{\max} = 1.16/d$. In the presence of a resonance at a frequency of $\omega = \omega_T x(kd)$, where $x(kd)$ is the function determined by

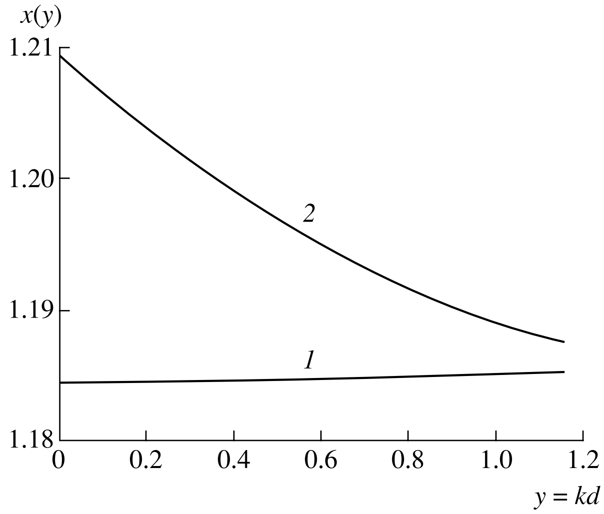

Fig. 2. Dispersion relations of the surface electromagnetic modes doe a dielectric film (SiC) on metal (Au) substrate. Curves *1* and *2* correspond to formulas (11) and (12), respectively.

the right-hand parts of Eqs. (11) and (12), formula (9) can be reduced to the following form:

$$D''(\omega, k) = 2\pi sh(2kd)\frac{\delta(\omega - \omega_T x(dx))}{\left|d\Delta_1'(\omega)/d\omega\right|_{\omega = \omega_T x(kd)}}. \quad (15)$$

$$\left|\frac{d\Delta_1'(\omega)}{d\omega}\right|_{\omega = \omega_T x(kd)} = \frac{1}{\omega_T}\frac{(p - x^2)^2 + \tilde{\gamma}^2 x^2}{\left|x^2(3ar + 4) - 2ar(p + q - \tilde{\gamma}^2) - 4p + 3\tilde{\gamma}^2\right|}, \quad (16)$$

where the argument of the function $x(kd)$ is omitted. Upon substitution of formulas (6) and (15) into Eq. (4), integration with respect to the frequency is simple, and the integral with respect to the wavevector can be calculated by numerical methods. It was interesting to compare the results of calculations of the forces of interaction between a metal particle and a metal surface with and without a dielectric film. Restricting the consideration to the case of equal temperatures ($T_1 = T_2$) and equal conductivities of the particle and substrate, we obtain from Eq. (4) for the "pure surface

$$F_x = -\frac{9}{32\pi}\frac{\hbar V R^3}{z_0^5}\left(\frac{k_B T}{\hbar\sigma_0}\right)^2. \quad (17)$$

Using Eqs. (4)–(6) and (15)–(17), the ratio of forces F_x for the surfaces with and without an adsorbed film can be written as

$$H(\alpha, \beta) = \frac{\sigma_0}{\omega_W}Y(\alpha, \beta), \quad \alpha = \frac{z_0}{d}, \quad \beta = \frac{\omega_T}{\omega_W}, \quad (18)$$

where $\omega_W = k_B T/\hbar$ is the Wien frequency and $Y(\alpha, \beta)$ is a function determined by numerical methods (the dis-

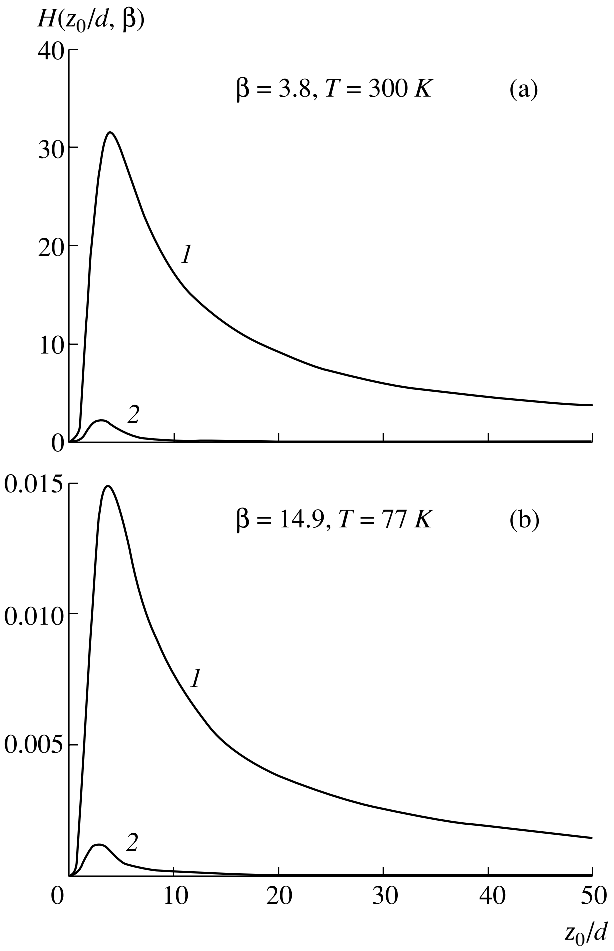

Fig. 3. The ratio of tangential forces for the interaction of a gold nanoparticle with a gold substrate with and without a silicon carbide film of thickness d at $T = 300$ (a) and 77 K (b). Curves *1* and *2* correspond to dispersion relations determined by formulas (11) and (12), respectively.

tance z_0 from the "pure" and coated surface is assumed to be the same).

Figure 3 shows the results of calculation of the ratio $H(\alpha, \beta)$ for a silicon carbide film on gold. Curves *1* and *2* correspond to the surface modes of two types determined by formulas (1) and (12). The calculation was performed for two temperatures: $T_1 = T_2 = T = 300$ K (Fig. 3a) and $T_1 = T_2 = T = 77$ K (Fig. 3b). As can be seen, the presence of a dielectric film on the metal substrate can increase the dissipative force at room temperature by one to two orders of magnitude, the maximum effect being observed for a certain relation between the distance from the particle to the surface and the film thickness (in our case, for $z_0/d \approx 3$–4). A decrease in the temperature reduces the tangential force three to four orders of magnitude (Fig. 3b). Thus, in the general case, the presence of an adsorbed film decreases the friction. This effect is related to the exponential temperature factor in Eq. (4) and the large value of parameter $\beta = \omega_T/\omega_W$ ($\beta = 14.9$ at $T = 77$ K). For dielectric films with lower values of the transverse phonon frequency ω_T (e.g., for ZnS), the temperature-induced decrease in the interaction force is less pronounced.

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