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# Exact expressions for colloidal plane–particle interaction forces and energies with applications to atomic force microscopy

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## Abstract

We begin by deriving a general useful theoretical relationship between the plane–particle interaction forces in solution, and the corresponding plane–plane interaction energies. This is the main result of the paper. It provides a simple tool to obtain closed-form particle–plane forces from knowledge of plane–plane interaction energies. To illustrate the simplicity of use of this general formalism, we apply it to find particle–plane interactions within the Derjaguin–Landau–Verwey–Overbeek (DLVO) framework. Specifically, we obtain analytical expressions for forces and interaction energies in the van der Waals and the electrical double layer cases. The van der Waals expression is calculated here for benchmarking purposes and is compared with well-established expressions from Hamaker theory. The interactions for the electric double layer situation are computed in two cases: the linear superposition approximation and the constant surface potential. In both cases, our closed-form expressions were compared with existent numerical results. We also use the main result of this paper to generate an analytical force-separation expression based on atomic force microscope experiments for a tip and surface immersed in an aqueous solution, and compare it with the corresponding numerical results. Finally, based on our main result, we generalize the Derjaguin approximation by calculating the next order of approximation, thus obtaining a formula valuable for colloidal interaction estimations.

## 1. Introduction

*A priori* knowledge of the parametric functional form of force-separation curves in atomic force microscopy (AFM) is necessary for writing reconstruction algorithms that predict the morphology and chemical activity of the sample under study. In vacuum, the tip and the sample can be modelled as a collection of atoms. The total tip–sample force can thus be evaluated from the appropriate atom–atom forces. These elemental forces have been modelled via Morse

and 6–12 pair potentials. More sophisticated, quantum  $N$ -body approaches have also been used in the past [1, 2]. Nevertheless, a large number of applications occur for tip–sample interactions in aqueous solutions where the elemental atom–atom interactions are screened by ions at the surfaces. In addition, beyond applications in AFM, knowledge of the plane–particle interaction is significant *per se* in particle deposition [3], antireflection coating [4], data storage [5] and membrane separation processes [6]. Indeed, since the inception of colloid probe microscopy [7, 8], sphere–plane force measurements have become routine in aqueous solution studies.

Within the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory, the generation of closed-form expressions for these colloidal interactions has been hampered by the fact that analytical solutions to the Poisson–Boltzmann equation (PBE) are restricted to planar geometries. Numerical solutions for other geometries do exist, but it is always desirable to have analytical expressions.

To bypass the seemingly insurmountable problem of obtaining analytical solutions to PBE curved geometries, a major effort has been placed on constructing a set of reasonable assumptions that, when taken together with the planar solution to the PBE, generate good expressions for the interactions between colloidal particles.

The most celebrated effort in that direction, the Derjaguin approximation, provides an explicit recipe for the calculation of the interaction energy between two spheroidal particles, from the knowledge of the interaction energy between two planes. A detailed study of the limitations of this approach can be found elsewhere [9]; however, its major restriction is that the radii of curvature of the particles, at closest approach, must be much larger than the interaction length. This condition is not satisfied in many cases of interest. For example, typical interaction lengths in liquids are 1–10 nm—similar to the radius of curvature of sharp AFM tips. Thus, the Derjaguin approximation is expected not to provide a good tool for the analysis of AFM tip–sample forces.

A more recent approach, surface element integration (SEI), has been used to obtain colloidal interaction energies [10]. In particular, this method has been shown to be very robust when restricted to plane–particle interactions. This geometry is of intrinsic interest to AFM when studying extended samples, and for the interpretation of calibration procedures.

The paper is organized as follows. In section 2, we derive the central outcome of this paper, namely a relationship between generic plane–plane interaction energy, and its corresponding plane–sphere force. Then we apply that relationship to obtain closed-form sphere–plane interactions for the van der Waals (section 3) and for the electric double layer cases (section 4). Our results are compared with previous theoretical and numerical studies. In section 5, we study a model system based on recent experimental data and present explicit formulae for forces in that case. In addition, we perform comparisons with numerical computations in this case. In section 6, we obtain the next order correction to the Derjaguin approximation. Compared with Derjaguin's, this expression still provides a simple tool for estimation of interactions, while removing the restriction that the radius of curvature must be infinitely large. In section 7 we present our conclusions.

## 2. Theory

We begin from the interaction energy  $U$ , between a flat plane and a spherical particle as calculated using the SEI formalism [10, 11]:

$$U(D) = 2\pi \int_0^a \left[ E\left(D + a - a\sqrt{1 - \left(\frac{r}{a}\right)^2}\right) - E\left(D + a + a\sqrt{1 - \left(\frac{r}{a}\right)^2}\right) \right] r \, dr, \quad (1)$$

where  $E(h)$  is the interaction energy between two infinite planes, separated a distance  $h$ ,  $D$  is the distance of closest approach between the sphere and the plane, and  $a$  is the radius of the sphere.

The force between the sphere and the plane is then obtained from

$$F(D) = -\frac{dU(D)}{dD} \quad (2)$$

or, explicitly using (1),

$$F(D) = -\int_0^a \frac{\partial E\left(D+a-a\sqrt{1-\left(\frac{r}{a}\right)^2}\right)}{\partial D} + \int_0^a \frac{\partial E\left(D+a+a\sqrt{1-\left(\frac{r}{a}\right)^2}\right)}{\partial D}. \quad (3)$$

Both integrals in equation (3) can be condensed into a single expression

$$I_\varepsilon(D) = \varepsilon \int_0^a \frac{\partial}{\partial D} E\left(D+a-\varepsilon a\sqrt{1-\left(\frac{r}{a}\right)^2}\right) r \, dr \quad (4a)$$

with  $\varepsilon = \pm 1$ , such that

$$F(D) = I_{-1}(D) + I_{+1}(D). \quad (4b)$$

Next, consider the change of variables  $r \rightarrow \theta$ ,

$$r = a \sin \theta, \quad \text{with } 0 \leq \theta \leq \frac{\pi}{2}. \quad (5)$$

Then,

$$I_\varepsilon(D) = \varepsilon a^2 \int_0^{\pi/2} \frac{\partial}{\partial D} E(D+a+\varepsilon a \cos \theta) \sin \theta \cos \theta \, d\theta. \quad (6)$$

Notice that

$$\frac{\partial}{\partial \theta} E(D+a+\varepsilon a \cos \theta) = -\varepsilon a \sin \theta \frac{\partial}{\partial D} E(D+a+\varepsilon a \cos \theta). \quad (7)$$

Thus, the integral in (6) can be written as

$$I_\varepsilon(D) = -a \int_0^{\pi/2} \frac{\partial}{\partial \theta} E(D+a+\varepsilon a \cos \theta) \cos \theta \, d\theta. \quad (8)$$

On integrating by parts,

$$I_\varepsilon(D) = -a \left\{ [E(D+a+\varepsilon a \cos \theta) \cos \theta]_0^{\pi/2} + \int_0^{\pi/2} E(D+a+\varepsilon a \cos \theta) \sin \theta \, d\theta \right\}. \quad (9)$$

That is,

$$I_\varepsilon(D) = aE(D+a+\varepsilon a) - a \int_0^{\pi/2} E(D+a+\varepsilon a \cos \theta) \sin \theta \, d\theta. \quad (10)$$

We now perform an additional change of variables  $x(\theta) = D+a+\varepsilon a \cos \theta$  (6), so that equation (10) becomes

$$I_\varepsilon(D) = aE(D+a+\varepsilon a) - a \int_{D+a+\varepsilon a}^{D+a} E(x) \, dx. \quad (11)$$

Using (11) and (4), and substituting into (3),

$$F(D) = 2\pi \left[ aE(D+2a) + \int_{D+2a}^{D+a} E(x) \, dx + aE(D) - \int_D^{D+a} E(x) \, dx \right]. \quad (12)$$

That is,

$$F(D) = 2\pi a[E(D) + E(D + 2a)] - 2\pi \int_D^{D+2a} E(x) dx. \quad (13)$$

Equation (13) is the central result of this article. It provides an economical prescription to evaluate the particle–surface force from knowledge of the energy–separation function between two planes. It presents clear advantages to the direct use of equation (3), in that equation (13) can be directly integrated analytically for many cases of interest while equation (3) can only be handled numerically. We show explicit applications in the remainder of the paper.

### 3. Van der Waals interaction

The van der Waals case has been solved a long time ago [12], and we only report it here as a validation of our formula, equation (13). The plane–plane van der Waals interaction is given by [13]

$$E(h) = -\frac{A_H}{12\pi h^2}, \quad (14)$$

where  $A_H$  is the effective Hamaker constant.

Substituting equation (14) into (13) provides

$$F(D) = -\frac{A_H}{6} \left[ \frac{a}{D^2} + \frac{a}{(D + 2a)^2} + \frac{1}{D + 2a} - \frac{1}{D} \right]. \quad (15)$$

This force comes, via equation (2), from the potential

$$U(D) = -\frac{A_H}{6} \left[ \frac{a}{D} + \frac{a}{D + 2a} + \ln\left(\frac{D}{D + 2a}\right) \right], \quad (16)$$

which is in agreement with the usual Hamaker expression.

### 4. Electric double layer interaction

In this section we use equation (13) in conjunction with published expressions for the electric double layer plane–plane interaction in two cases: (1) the linear superposition approximation [14] and (2) the constant surface potential [15].

In the linear superposition approximation the energy per unit area between two infinite planes is

$$E(h) = 32\varepsilon_0\varepsilon_r\kappa \tanh\left(\frac{ze\psi_1}{4kT}\right) \tanh\left(\frac{ze\psi_2}{4kT}\right) \left(\frac{kT}{ze}\right)^2 e^{-\kappa h}, \quad (17)$$

where  $\varepsilon_0$  is the vacuum permittivity,  $e$  is the elementary charge,  $\varepsilon_r$  the permittivity of the electrolyte,  $z$  is the valence in a symmetrical electrolyte,  $\kappa$  is the inverse Debye length,  $\psi_1$  and  $\psi_2$  are the electrostatic potentials at each plane,  $k$  is Boltzmann constant, and  $T$  the absolute temperature.

Application of equation (13) in this case yields

$$F(D) = 2\pi a \left[ 32\varepsilon_0\varepsilon_r\kappa \tanh\left(\frac{ze\psi_1}{4kT}\right) \tanh\left(\frac{ze\psi_2}{4kT}\right) \left(\frac{kT}{ze}\right)^2 \right] \\ \times \left[ \left(1 - \frac{1}{\kappa a}\right) + \left(1 + \frac{1}{\kappa a}\right) e^{-2\kappa a} \right] e^{-\kappa D}. \quad (18)$$

The Derjaguin approximation is recovered correctly by letting  $\kappa a \rightarrow +\infty$ .

An additional integration on  $D$  provides the particle–plane interaction energy,

$$U(D) = 2\pi a^2 \left[ 32\varepsilon_0\varepsilon_r\kappa \tanh\left(\frac{ze\psi_1}{4kT}\right) \tanh\left(\frac{ze\psi_2}{4kT}\right) \left(\frac{kT}{ze}\right)^2 \right] \\ \times \left[ \left(\frac{1}{\kappa a} - \frac{1}{(\kappa a)^2}\right) + \left(\frac{1}{\kappa a} + \frac{1}{(\kappa a)^2}\right) e^{-2\kappa a} \right] e^{-\kappa D}. \quad (19)$$

In the constant surface potential case, the plane–plane interaction energy is

$$E(h) = \frac{\kappa\varepsilon_0\varepsilon_r}{2} (\psi_1^2 + \psi_2^2) \left[ 1 - \frac{1}{\tanh(\kappa h)} + \frac{2\psi_1\psi_2}{\psi_1^2 + \psi_2^2} \frac{1}{\sinh(\kappa h)} \right]. \quad (20)$$

Substituting this particular form in equation (13) renders

$$F(D) = \pi\varepsilon_0\varepsilon_r(\psi_1^2 + \psi_2^2)\kappa a \left\{ \frac{1}{\tanh(\kappa D + 2\kappa a)} - \frac{1}{\tanh(\kappa D)} + \frac{1}{\kappa a} \log\left(\frac{\sinh(\kappa D + 2\kappa a)}{\sinh(\kappa D)}\right) \right. \\ \left. + \frac{2\psi_1\psi_2}{\psi_1^2 + \psi_2^2} \left[ \frac{1}{\sinh(\kappa D + 2\kappa a)} + \frac{1}{\sinh(\kappa D)} - \frac{1}{\kappa a} \log\left(\frac{\tanh\left(\frac{\kappa D}{2} + \kappa a\right)}{\tanh\left(\frac{\kappa D}{2}\right)}\right) \right] \right\} \quad (21)$$

which, again, provides the appropriate Derjaguin limit when  $\kappa a \rightarrow +\infty$ .

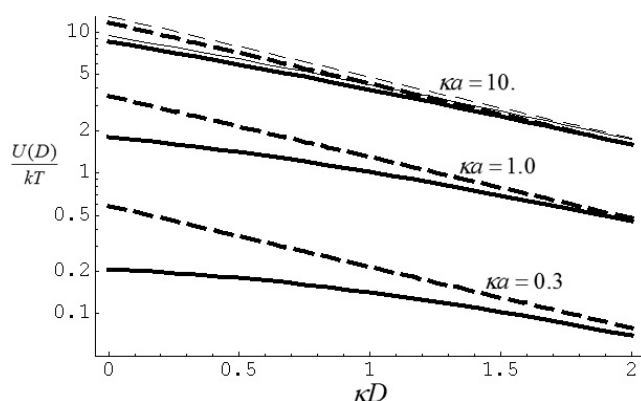
We now integrate equation (21) to obtain an explicit expression for the particle–plane interaction energy,

$$U(D) = \pi\varepsilon_0\varepsilon_r(\psi_1^2 + \psi_2^2)a \left\{ \frac{2\psi_1\psi_2}{\psi_1^2 + \psi_2^2} \left[ -\log\left(\frac{\tanh\left(\frac{\kappa D}{2} + \kappa a\right)}{\tanh\left(\frac{\kappa D}{2}\right)}\right) \right. \right. \\ \left. \left. + \frac{2\kappa a + \kappa D}{\kappa a} \log\left(\frac{1 + e^{-\kappa D - 2\kappa a}}{1 - e^{-\kappa D - 2\kappa a}}\right) + \frac{2\kappa a + \kappa D}{\kappa a} \log\left(\tanh\left(\frac{\kappa D}{2} + \kappa a\right)\right) \right] \right. \\ \left. + \frac{\text{Li}_2(e^{-\kappa D - 2\kappa a}) - \text{Li}_2(-e^{-\kappa D - 2\kappa a})}{\kappa a} \right] \\ - \frac{2\psi_1\psi_2}{\psi_1^2 + \psi_2^2} \left[ \frac{\kappa D}{\kappa a} \log\left(\frac{1 + e^{-\kappa D}}{1 - e^{-\kappa D}}\right) + \frac{\kappa D}{\kappa a} \log\left(\tanh\left(\frac{\kappa D}{2}\right)\right) \right] \\ \left. + \frac{\text{Li}_2(e^{-\kappa D}) - \text{Li}_2(-e^{-\kappa D})}{\kappa a} \right] \\ + \log\left(\frac{\sinh(\kappa D + 2\kappa a)}{\sinh(\kappa D)}\right) - \frac{\kappa D}{2\kappa a} [\kappa D + 2\log(1 - e^{-2\kappa D}) \\ - 2\log(\sinh(\kappa D))] + \frac{1}{2\kappa a} \text{Li}_2(e^{-2\kappa D}) \\ + \frac{\kappa D + 2\kappa a}{2\kappa a} [\kappa D + 2\kappa a + 2\log(1 - e^{-2\kappa D - 4\kappa a}) \\ - 2\log(\sinh(\kappa D + 2\kappa a))] - \frac{1}{2\kappa a} \text{Li}_2(e^{-2\kappa D - 4\kappa a}) \left. \right\} \quad (22)$$

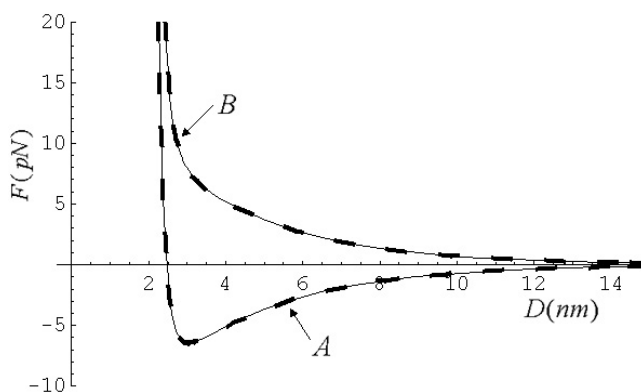
where Li is the logarithm integral [16] function.

To the author's knowledge this is the only instance in which analytical forms for the potential  $U(D)$ , and force  $F(D)$ , have been published in the literature. The lack of such expressions rests on the difficulty of integrating equation (2) directly.

In figure 1 we plot the interaction energies for the linear superposition approximation and the constant surface potential.



**Figure 1.** Plane–particle interaction energies for the linear superposition approximation (dashed) and the constant surface potential (solid). For comparison, the thin lines correspond to the Derjaguin approximation.



**Figure 2.** AFM tip–surface force curve for experimental parameters  $D_0 = 1$  nm,  $a = 7$  nm,  $B = 10^{-72}$  J m<sup>6</sup>,  $\epsilon_r = 78.5$ ,  $1/\kappa = 3$  nm. Line A corresponds to  $\sigma_1\sigma_2 = -20$  pC<sup>2</sup> nm<sup>-2</sup>, and line B to  $\sigma_1\sigma_2 = 20$  pC<sup>2</sup> nm<sup>-2</sup>. The dashed thick lines are those of this work and the thin continuous lines are those from numerical integration in [17].

We compare these plots with those obtained by numerical integration [10] and find perfect agreement to the extent that the corresponding curves are so close that no visual difference can be detected when plotted in figure 2.

In addition we show, in figure 1, curves corresponding to Derjaguin approximation for  $\kappa a = 10$ , that is for large sphere radius. For the cases  $\kappa a = 1$  and 0.3, the disagreement is much beyond 50%.

## 5. Analysis of AFM experiments

Forces between an AFM tip and a plane in aqueous solutions have been investigated as a function of pH and ionic concentration [17]. Given that the tip, at its closest approach to the surface, appears spherical, it is reasonable to conjecture that the tip–plane forces can be derived from equation (13). This is purposively so in colloid probe microscopy, in which a colloidal sphere is attached at the end of the tip. Thus, in this section, the tip plays the role of

a large colloidal particle. In addition, once the tip–surface interaction is found, it can also be used to gain understanding of a colloidal system, in which the colloidal particles interact with the plane.

The plane–plane energy is [17]

$$E(h) = \frac{2\pi}{\varepsilon_r \varepsilon_0 \kappa} \sigma_1 \sigma_2 e^{-\kappa(h-D_0)} + \frac{B}{72\pi(h-D_0)^8} \quad (23)$$

where  $\sigma_1$  and  $\sigma_2$  are the surface charge densities of plane and tip, and  $B$  and  $D_0$  are parameters, usually determined experimentally [9]. In particular, the parameter  $D_0$  is introduced in AFM analysis, because it is not possible to know *a priori* where the contact between the tip and the sample is.

In order to compare the corresponding tip–surface force with the experimental force–distance curve, the derivative of equation (2) with respect to distance was integrated numerically in [17].

Given the relevance of equation (23) to AFM measurements in liquids, we now use equation (13) to produce a closed-form expression for the force-separation,

$$\begin{aligned} F(D) = 2\pi a \left[ \frac{2\pi\sigma_1\sigma_2}{\varepsilon_r\varepsilon_0\kappa} (e^{-\kappa(D-D_0)} + e^{-\kappa(D+2a-D_0)}) \right. \\ \left. + \frac{B}{72\pi} \left( \frac{1}{(D-D_0)^8} + \frac{1}{(D+2a-D_0)^8} \right) \right] \\ + 2\pi \left[ \frac{2\pi\sigma_1\sigma_2}{\varepsilon_r\varepsilon_0\kappa^2} (e^{-\kappa(D+2a-D_0)} - e^{-\kappa(D-D_0)}) \right. \\ \left. + \frac{1}{7} \frac{B}{72\pi} \left( \frac{1}{(D+2a-D_0)^7} - \frac{1}{(D-D_0)^7} \right) \right]. \quad (24) \end{aligned}$$

Figure 2 shows two sets of curves for experimentally relevant situations. The continuous lines were obtained by numerical integration in [17]. The dashed lines correspond to plots of equation (24).

In addition, by having available the expression (24) for the force, we can answer theoretical questions. For example, the zero of  $F(D)$  in equation (24) (when  $\sigma_1\sigma_2 < 0$ ) corresponds to an accretion region for colloidal particles near the surface. That is, if the solution contained colloids, that region is where a large concentration of the particles would be found. Since no zeros exist for  $\sigma_1\sigma_2 > 0$ , one expects a homogeneous distribution of the colloidal particles, with the exception of a small depletion layer near the surface due to the dominant repulsion there. This problem is of particular current relevance to understand adsorption and self-assembly of colloidal nanoparticles on surfaces and has been studied both experimentally [18] as well as computationally [19]. In addition, from the closed-form expressions reported in this paper we can also evaluate the gradient of the force at equilibrium, which is related to the effective spring constant of oscillation of particles around equilibrium. This parameter, in conjunction with the knowledge of the viscosity of the liquid, is related to the timescales necessary for the colloidal layer to achieve equilibrium, thus providing a useful tool for colloidal adlayer formation.

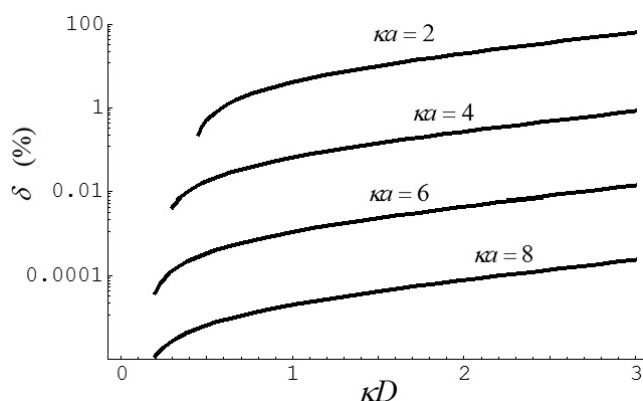
## 6. Generalization of Derjaguin approximation

The Derjaguin approximation has been used extensively to estimate colloidal interactions. But its usefulness relies on our understanding of its limitations. It is thus appropriate, in the light of equation (13), to evaluate the next order of approximation, that is, beyond Derjaguin's.

In the Derjaguin approximation, the force for a colloidal plane–sphere system is

$$F(D) = 2\pi a E(D). \quad (25)$$





**Figure 3.** Percentage difference between exact force, and approximate force from equation (27). As expected, as  $\kappa a$  decreases, the agreement between the exact and the approximate force deteriorates. Moreover, for  $\kappa a < 2$  equation (26) cannot be used in the range of interest. On the other hand, already for  $\kappa a > 6$  the error is below 0.01% and thus can be of use to check trends beyond Derjaguin's range of applicability. For comparison, for  $\kappa a = 6$ , the corresponding difference for the Derjaguin approximation is about 10%.

This result is recovered from equation (13) by noticing that when  $a \rightarrow \infty$ ,  $E(D + 2a) \rightarrow 0$ . In addition, the integral in equation (13) also becomes negligible. Indeed, the function  $E(x)$  decreases as  $x$  becomes large. Call  $\ell$  the typical decay length—for example, for the electric double layer,  $\ell \approx 1/\kappa$ . The integral can be estimated to be  $\approx \ell E(D)$ . Finally, since  $\ell \ll a$ , this last term is much smaller than the first term  $2\pi a E(D)$ . Thus we recover Derjaguin's result.

When  $a$  is large, but not infinite, we must keep all terms in equation (13): the first term remains as before; the second term  $2\pi a E(D + 2a) \approx 2\pi a E(2a)$  since  $a \gg D$ ; the third term becomes  $\approx 2\pi E(D)\ell$ . Thus,

$$F(D) = 2\pi a E(D) \left\{ 1 - \frac{\ell}{a} + \frac{E(2a)}{E(D)} \right\}. \quad (26)$$

This expression is also simple and can be used for estimation purposes. We can test the strength of this statement with a specific quantitative example. Consider the linear superposition approximation for the electric double layer interaction. Equation (18) provides the exact interaction force,  $F_{18}(D)$ . Using equation (17) in (26) provides the approximate force  $F_{26}(D)$ . Then we construct the percentage relative error,

$$\delta = 100 \frac{|F_{18} - F_{26}|}{F_{18}} \quad (27)$$

and plot it in figure 3.

## 7. Conclusions

The central result of this paper is equation (13). It relates the plane–plane energy per unit area with the corresponding sphere–plane force. Similar relationships already exist in the literature [10]. The advantage of our equation (13) is its practicality, in the sense that it can be readily integrated for various cases of interest. Indeed, we were able to integrate equation (13) to provide closed-form expressions for electric double layer interactions in the linear superposition approximation and the constant surface potential. These expressions,

being given in analytical form, are valuable, for input in molecular dynamics simulations, for example. For comparison with Hamaker theory, we also evaluated van der Waals interactions using equation (13). We then obtained an analytical expression for force–distance curves as a function of pH and ionic concentration based on AFM experiments. Finally, we used equation (13) to find the first order correction beyond Derjaguin approximation. This is done in the spirit of preserving a simple form, practical for prompt estimations.

We would like to stress that the evaluation of the potential for the electrostatic double layer was done in the past numerically. Although closed form expressions for the electrostatic double layer potential in non-planar geometries are difficult to obtain, we have presented expressions for the corresponding potential and forces in the sphere–plane geometry. In atomic force microscopy, this is the relevant geometry for tip–sample interactions and a solution of this type can be used, among other things, for calibrations procedures. For atomic force microscopy, forces, as opposed to potentials, are the main quantities; thus the interest of reporting expressions for force in the paper.

### Acknowledgment

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