

Technical report on the anisotropic cylinder-cylinder interactions at all separations

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I will list all the equations that are needed for numerical implementation of the anisotropic cylinder-cylinder interactions. The code should be centered around the equations enclosed in frames, Eq. 12 and Eq. 29.

I. INTRODUCTION

We will write the complete van der Waals - dispersion interaction free energy between two anisotropic cylinders at all separations, including the retardation effects. We start with the Lifshitz theory of van der Waals interactions between two semiinfinite anisotropic uniaxial dielectric layers across a finite layer of dielectric function ϵ_m and thickness ℓ as worked out by Barash [6] - the result of this calculation is the interaction free energy between the two layers as a function of their separation ℓ and the angle between their principal dielectric anisotropy axes θ : $\mathcal{G}(\ell, \theta)$. The dielectric response of the two dielectrically uniaxial half-spaces is given by the values of their dielectric functions $\overline{\epsilon}_{\parallel}$, parallel and $\overline{\epsilon}_{\perp}$, perpendicular to their respective axes. We shall use $\overline{\epsilon}_{1,\parallel}$ ($\overline{\epsilon}_{1,\perp}$) and $\overline{\epsilon}_{2,\parallel}$ ($\overline{\epsilon}_{2,\perp}$) for the left and right half-spaces, respectively. Note also that in the theory of van der Waals interactions [8, 9] all the dielectric response functions are evaluated at imaginary frequencies, thus $\epsilon_{\parallel,\perp} = \epsilon_{\parallel,\perp}(i\omega)$. $\epsilon_{\parallel,\perp}(i\omega)$ is referred to as the London - van der Waals transform of the response function $\epsilon_{\parallel,\perp}(\omega)$ and is given by the Kramers - Kronig relations. It is strictly a real, monotonically decaying function of ω .

From the interaction free energy between two half-spaces one can extract the interaction between two cylinders (see Fig. 1) by assuming that the two half-spaces are dilute assemblies of anisotropic cylinders. One should keep in mind, however, that the cylinder dielectric response is isotropic in the plane perpendicular to the cylinder axis - we call this the transverse dielectric response. The difference between the transverse response and the response in the direction parallel to the cylinder axis (longitudinal response) constitutes the dielectric anisotropy of the problem. The derivation closely follows the arguments of Pitaevskii for evaluating the interactions between isotropic impurity atoms in a homogeneous fluid [10]. We assume that the two anisotropic half-spaces are composed of anisotropic cylinders of radii R_1 and R_2 at volume fractions v_1 and v_2 , with $\epsilon_{1,\perp}^c$ ($\epsilon_{2,\perp}^c$) and $\epsilon_{1,\parallel}^c$ ($\epsilon_{2,\parallel}^c$) as the transverse and longitudinal dielectric response functions of the cylinder materials. We then expand $\mathcal{G}(\ell, \theta)$ for two half-spaces as a series in v_1 and v_2 and evaluate the coefficient multiplying the $v_1 v_2$ term. The volume fractions v_1 and v_2 scale with the area

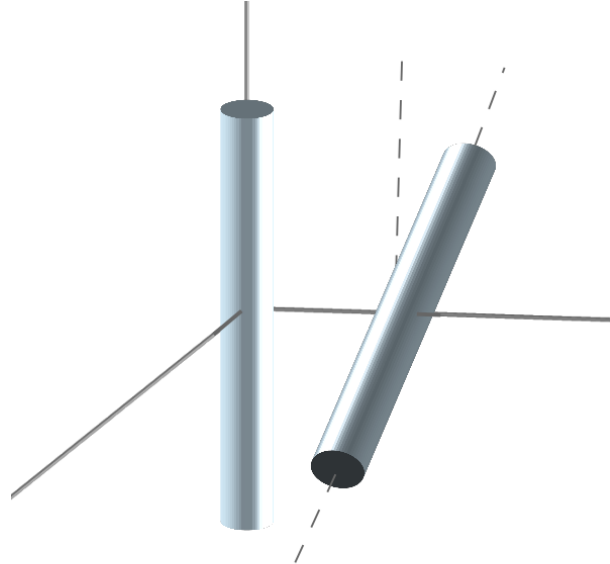


FIG. 1: A sketch of the system of interest (the two cylinders). The quantities describing the geometry of the system are denoted, together with the longitudinal and transverse directions of cylinder in the left half-space (1). The skew angle θ is about an axis normal to the planar boundary defining the limits of each half-space.

density of the cylinders (N_1, N_2) in the direction of their long axes as $v_1 = N_1 \pi R_1^2$ ($v_2 = N_2 \pi R_2^2$). It then follows [8] that the interaction free energy between two cylinders, $G(\ell, \theta)$, whose axes are contained within the two parallel boundaries at a separation ℓ , but skewed at an angle θ (see Fig. 1) is given by

$$\frac{d^2 \mathcal{G}(\ell, \theta)}{d\ell^2} = N_1 N_2 \sin \theta G(\ell, \theta). \quad (1)$$

Conversely, the interaction free energy *per unit length*, $g(\ell)$, between two parallel cylinders is given by the Abel transform (see e.g. Ref. 8, pp 233-235)

$$\frac{d^2 \mathcal{G}(\ell, \theta = 0)}{d\ell^2} = N_1 N_2 \int_{-\infty}^{+\infty} g(\sqrt{\ell^2 + y^2}) dy. \quad (2)$$

In both cases we expand $\mathcal{G}(\ell, \theta)$ to find the coefficient next to $v_1 v_2$ (or equivalently $N_1 N_2$), take the second derivative with respect to ℓ , then use Eqs. 1 and 2 in order to obtain the appropriate pair interaction free energy between cylinders. Note that such an expansion is possible only if the dielectric response at all frequencies is bounded. In the case of an ideal metal Drude-like dielectric response this expansion is not feasible and our method can not be transplanted to that case automatically.

The closest attempt in the literature to evaluate the interaction between two cylinders at all separations comes from Barash and Kyasov [11]. Where this approach can be compared with the one presented here, i.e. for two parallel isotropic cylinders, the results for the interaction free energy between parallel cylinders coincide completely. The results described below were published in A. Šiber, R. F. Rajter, R. H. French, W. Y. Ching, V. A. Parsegian, and R. Podgornik: *Dispersion interactions between optically anisotropic cylinders at all separations: Retardation effects for insulating and semiconducting single-wall carbon nanotubes*, PHYSICAL REVIEW B 80, 165414 (2009).

II. DERIVATION

We use the Pitaevskii *ansatz* in order to extract the interactions between two infinite anisotropic cylinders at all separations and angles from the interaction between two semi-infinite half-spaces of anisotropic uniaxial dielectric material. We start with the fully retarded van der Waals - dispersion interactions between two semiinfinite anisotropic dielectric slabs [6]. The full interaction form is quite involved, but it has a simple limit if the two semiinfinite slabs, \mathcal{L} and \mathcal{R} , separated by an isotropic medium of thickness ℓ , are composed of rarefied material.

In order to get the interaction free energy between two anisotropic cylinders we assume that both semi-infinite substrates (half-spaces), \mathcal{L} (1) and \mathcal{R} (2), are composite materials made of oriented anisotropic cylinders at volume fractions v_1 and v_2 , with $\epsilon_{1,\perp}^c$ ($\epsilon_{2,\perp}^c$) and $\epsilon_{1,\parallel}^c$ ($\epsilon_{2,\parallel}^c$) as the transverse and longitudinal dielectric response functions of the cylinder materials. For the semi-infinite composite medium of oriented anisotropic cylinders with local hexagonal packing symmetry, so that the corresponding cylinder volume fraction is v , the anisotropic bulk dielectric response function can be derived in the form (see Ref. 8, p.318)

$$\bar{\epsilon}_{\parallel} = \epsilon_m (1 + v \Delta_{\parallel}), \quad \bar{\epsilon}_{\perp} = \epsilon_m \left(1 + \frac{2v \Delta_{\perp}}{1 - v \Delta_{\perp}} \right), \quad (3)$$

where the relative anisotropy measures in the parallel and perpendicular direction are given by

$$\Delta_{\perp} = \frac{\epsilon_{\perp}^c - \epsilon_m}{\epsilon_{\perp}^c + \epsilon_m} \quad \Delta_{\parallel} = \frac{\epsilon_{\parallel}^c - \epsilon_m}{\epsilon_m}. \quad (4)$$

In our case, this holds for both \mathcal{L} and \mathcal{R} half-spaces with the appropriate volume fractions and dielectric responses. ϵ_m is the dielectric function of the isotropic medium between the cylinders as well as between regions \mathcal{L} and \mathcal{R} . We assume in what follows that all the response functions are bounded and finite.

The formulae in Eqs. 1,2 connect the interaction free energy of two semiinfinite half spaces with the interaction free energy between two cylinders either parallel or skewed at a finite angle θ . The Barash result [6] for the complete retarded form of the interactions between two uniaxial media, $\mathcal{G}(\ell, \theta)$, is quite complicated (note also a typo that propagated starting from the original version of the calculation [7] and was first noted in Ref. 12) but can be straightforwardly expanded to second order in N (a term proportional to $v_1 v_2$) for the dielectric response functions of the form Eq. 3, yielding the following result

$$\frac{d^2 \mathcal{G}(\ell, \theta)}{d\ell^2} = \frac{k_B T}{2\pi} \sum_{n=0}^{\infty} \int_0^{\infty} Q dQ \frac{d^2 f(\ell, \theta)}{d\ell^2}. \quad (5)$$

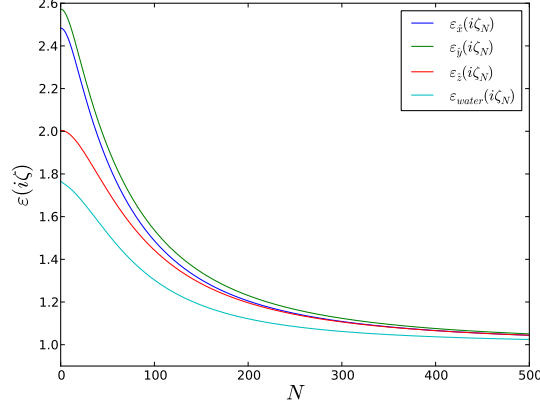


FIG. 2: Anisotropic response functions for CG-10 DNA and water. The DNA response functions in the x and y directions were used as perpendicular and parallel inputs, respectively. CG-10 and water eps2 data was provided by Dan Dryden. CG-10 data scales Wai-Yim's calculations by 4.94 and is assumed to include Na (more info in Dan Dryden email sent to us on Nov. 8, 2013). Water data was built from lorentz oscillators R.H.French,J.Amer.Ceram Soc.,83,9,2117-46(2000), H.D.Ackler, et al,J.Coll.Interface Sci.179,46.

In the above equation, n represent the (thermal) Matsubara indices, the prime on the summation means that the weight of the $n = 0$ term is $1/2$ (see Refs. 8, 11 for details). The second derivative of the function $f(\ell, \theta)$ can be obtained explicitly in terms of the ratios between the relative anisotropy measures (Eq. 4) defined as

$$a = \frac{2\Delta_{\perp}}{\Delta_{\parallel}} = 2 \frac{(\epsilon_{\perp}^c - \epsilon_m)\epsilon_m}{(\epsilon_{\perp}^c + \epsilon_m)(\epsilon_{\parallel}^c - \epsilon_m)} \quad (6)$$

and is obviously frequency dependent. Parameters a_1 and a_2 can be thought of as a specific measure of the anisotropy of the cylinders in the left and right half-spaces when compared with the isotropic bathing medium m . Note that they vanish when the transverse dielectric response of the cylinder material equals the medium response. The explicit form of the second derivative of $f(\ell, \theta)$ now follows as

$$\begin{aligned} \frac{d^2 f(\ell, \theta)}{d\ell^2} = & -\frac{v_1 v_2 \Delta_{1,\parallel} \Delta_{2,\parallel}}{32} \frac{e^{-2\ell \sqrt{Q^2 + \epsilon_m \frac{\omega_n^2}{c^2}}}}{(Q^2 + \epsilon_m \frac{\omega_n^2}{c^2})} \\ & \left\{ 2 \left[(1 + 3a_1)(1 + 3a_2)Q^4 + 2(1 + 2a_1 + 2a_2 + 3a_1 a_2)Q^2 \epsilon_m \frac{\omega_n^2}{c^2} + 2(1 + a_1)(1 + a_2)\epsilon_m^2 \frac{\omega_n^4}{c^4} \right] + \right. \\ & \left. + (1 - a_1)(1 - a_2) \left(Q^2 + 2\epsilon_m \frac{\omega_n^2}{c^2} \right)^2 \cos 2\theta \right\}. \quad (7) \end{aligned}$$

Here R_1 and R_2 are the cylinder radii, assumed to be the smallest lengths in the problem [11]. The frequency dependence of the dielectric functions is in $\epsilon_m(i\omega_n)$, $\epsilon_{\perp}^c(i\omega_n)$ and $\epsilon_{\parallel}^c(i\omega_n)$, and therefore also $a = a(i\omega_n)$. The frequencies in the Matsubara summation are $\omega_n = 2\pi \frac{k_B T}{\hbar} n$. Note that Eq. 7 is symmetric with respect to 1 and 2 indices (left and right half-spaces), as it should be.

This is as far as a general theory can go. We must now deal separately with the cases of skewed and parallel cylinders, since the connection between $\frac{d^2 \mathcal{G}(\ell, \theta)}{d\ell^2}$ and the effective pair interaction between cylinders is different for the two cases, see Eqs. 1,2. We first analyze the case of skewed cylinders.

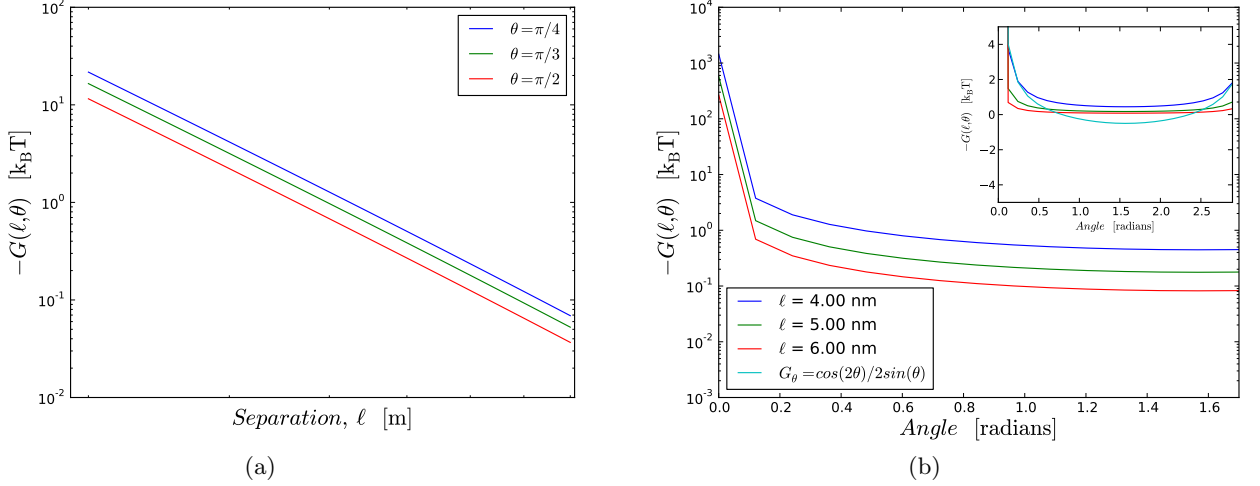


FIG. 3: Full result using Eqs.10,11 (a) Interaction free energy between two cylinders (CG-10 DNA) with radii of 1 nm in water plotted as a function of separation from 2 to 8 nm, for three values of mutual angle. (b) Interaction as a function of mutual angle (0 radians to $\pi/2$ radians) at three values of separation. The inset shows the periodic behavior of $G(\ell, \theta)$ over an interval π for three values of separation. The cyan curve shows the behavior of the theta dependence of G

III. SKEWED CYLINDERS

A. Skewed cylinders - full result

We use Eq. 1 to obtain the interaction free energy between two skewed cylinders:

$$G(\ell, \theta) = -\frac{k_B T}{64\pi} \frac{\pi^2 R_1^2 R_2^2}{\ell^4 \sin \theta} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} \int_0^{\infty} u du \frac{e^{-2\sqrt{u^2 + p_n^2}}}{(u^2 + p_n^2)} g(a_1, a_2, u, p_n, \theta), \quad (8)$$

where $u = Q\ell$,

$$g(a_1, a_2, u, p_n, \theta) = 2 \left[(1 + 3a_1)(1 + 3a_2)u^4 + 2(1 + 2a_1 + 2a_2 + 3a_1 a_2)u^2 p_n^2 + 2(1 + a_1)(1 + a_2)p_n^4 \right] + (1 - a_1)(1 - a_2)(u^2 + 2p_n^2)^2 \cos 2\theta \quad (9)$$

and $p_n^2(\ell) = \epsilon_m(i\omega_n) \frac{\omega_n^2}{c^2} \ell^2$. Another change of variables with $u = p_n t$, yields

$$G(\ell, \theta) = -\frac{k_B T}{64\pi} \frac{\pi^2 R_1^2 R_2^2}{\ell^4 \sin \theta} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} p_n^4 \int_0^{\infty} t dt \frac{e^{-2p_n \sqrt{t^2 + 1}}}{(t^2 + 1)} \tilde{g}(t, a_1(i\omega_n), a_2(i\omega_n), \theta), \quad (10)$$

with

$$\tilde{g}(t, a_1, a_2, \theta) = 2 \left[(1 + 3a_1)(1 + 3a_2)t^4 + 2(1 + 2a_1 + 2a_2 + 3a_1 a_2)t^2 + 2(1 + a_1)(1 + a_2) \right] + (1 - a_1)(1 - a_2)(t^2 + 2)^2 \cos 2\theta. \quad (11)$$

This is the final result for the cylinder-cylinder interaction at all angles when the radii of the cylinders are the smallest lengths in the system. It includes retardation and the full angular dependence. Some simple limits can be obtained from this general expression.

We now transform this result into a form that is suitable for computation and numerical implementation. First we rewrite Eq. 10 as

$$G(\ell, \theta) = -\frac{(\pi R_1^2)(\pi R_2^2)}{2\pi \ell^4 \sin \theta} \left(\mathcal{A}^{(0)}(\ell) + \mathcal{A}^{(2)}(\ell) \cos 2\theta \right), \quad (12)$$

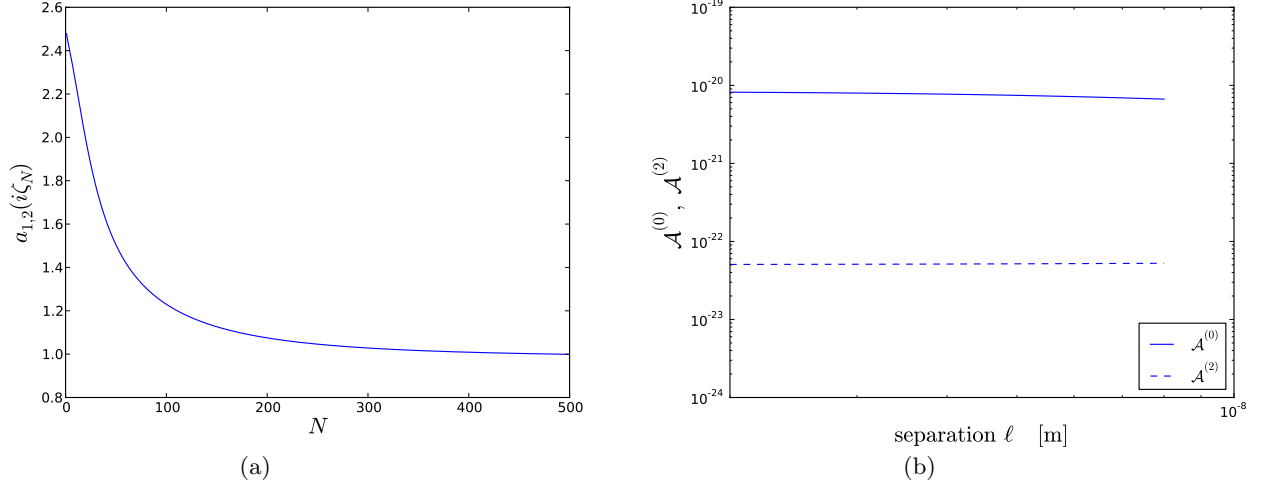


FIG. 4: (a) Anisotropy metric $a_{1,2}(i\zeta_N)$ using Eq.17, compares the anisotropy of the cylinders (DNA) to their intervening material, water for the terms contrubuting to the Matsubara sum. (b) Hamaker coefficients $\mathcal{A}^{(0)}(\ell)$ (solid line) and $\mathcal{A}^{(2)}(\ell)$ (dashed line) calculated using fully retarded formulation, Eqs.13-16.

The (ℓ) dependence of the Hamaker coefficients \mathcal{A} is a consequence of (ℓ) dependence of $p_n^2(\ell) = \epsilon_m(i\omega_n) \frac{\omega_n^2}{c^2} \ell^2$. Above we defined

$$\mathcal{A}^{(0)}(\ell) = \frac{k_B T}{32} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} p_n^4(\ell) \int_0^\infty t dt \frac{e^{-2p_n(\ell)\sqrt{t^2+1}}}{(t^2+1)} \tilde{g}^{(0)}(t, a_1(i\omega_n), a_2(i\omega_n)) \quad (13)$$

with

$$\tilde{g}^{(0)}(t, a_1(i\omega_n), a_2(i\omega_n)) = 2 [(1+3a_1)(1+3a_2)t^4 + 2(1+2a_1+2a_2+3a_1a_2)t^2 + 2(1+a_1)(1+a_2)] \quad (14)$$

and

$$\mathcal{A}^{(2)}(\ell) = \frac{k_B T}{32} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} p_n^4(\ell) \int_0^\infty t dt \frac{e^{-2p_n(\ell)\sqrt{t^2+1}}}{(t^2+1)} \tilde{g}^{(2)}(t, a_1(i\omega_n), a_2(i\omega_n), \theta) \quad (15)$$

with

$$< C - LeftRelease > \tilde{g}^{(2)}(t, a_1(i\omega_n), a_2(i\omega_n), \theta) = (1-a_1)(1-a_2)(t^2+2)^2 \quad (16)$$

The numerical implementation should be for Eqs. 12-16. For $a_{1,2}$ one invokes the previous definition Eq. 17

$$a_{1,2}(i\omega_n) = \frac{2\Delta_{\perp}^{(1,2)}(i\omega_n)}{\Delta_{\parallel}^{(1,2)}(i\omega_n)} = 2 \frac{(\epsilon_{\perp}^{c(1,2)}(i\omega_n) - \epsilon_m(i\omega_n))\epsilon_m(i\omega_n)}{(\epsilon_{\perp}^{c(1,2)}(i\omega_n) + \epsilon_m(i\omega_n))(\epsilon_{\parallel}^{c(1,2)}(i\omega_n) - \epsilon_m(i\omega_n))} \quad (17)$$

where $\epsilon_{\perp}^{c(1,2)}$, $\epsilon_{\parallel}^{c(1,2)}$ are the perpendicular, parallel components of the dielectric response functions of the two cylinders and ϵ_m is the same for the medium in between. All these quantities are of course frequency dependent. The n summation is over the Matsubara frequencies, $\zeta_n = 2\pi n k_B T / \hbar$, where n is an integer and the $n=0$ term is counted with a weight $1/2$. At room temperature the Matsubara frequencies are a multiple of $2.4 \times 10^{14} \text{ s}^{-1}$.

B. Skewed cylinders - non-retarded result

The non-retarded limit where $c \rightarrow \infty$, has already been explored in Ref. 5. There $p_n \rightarrow 0$ for all n and we obtain from Eq. 8

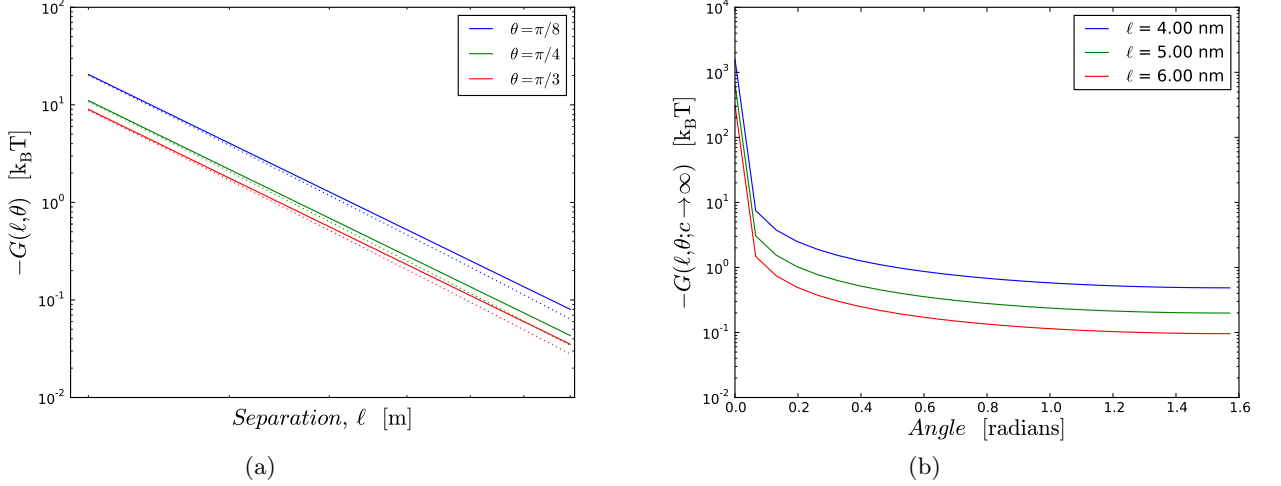


FIG. 5: Non-retarded result using Eq.18 (a) The nonretarded interaction free energy between two cylinders is shown as solid lines as a function of separation (2 nm to 8 nm) for three values of mutual angle. Dashed lines show retarded calculations for the same parameter values. (b) Interaction as a function of mutual angle from 0 to $\pi/2$ for three values of separation.

$$\begin{aligned}
 G(\ell, \theta; c \rightarrow \infty) &= -\frac{k_B T}{64\pi} \frac{\pi^2 R_1^2 R_2^2}{\ell^4 \sin \theta} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} \int_0^\infty u^3 du e^{-2u} [2(1+3a_1)(1+3a_2) + (1-a_1)(1-a_2) \cos 2\theta] = \\
 &= -\frac{k_B T}{64\pi} \frac{\pi^2 R_1^2 R_2^2}{\ell^4 \sin \theta} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} \frac{3}{8} [2(1+3a_1)(1+3a_2) + (1-a_1)(1-a_2) \cos 2\theta]. \quad (18)
 \end{aligned}$$

This formula could also be obtained directly from Eq. 10 taking into account that in the t integration only the terms with large t contribute to the final integral. Expanding the whole integrand for large t returns us to Eq. 18. The $n = 0$ term of this formula for two identical cylinders corresponds to classical dipolar fluctuation forces as analyzed in 13.

C. Skewed cylinders - low temperature result

At low temperatures, when the summation over the Matsubara frequencies can be turned into an integral over n with $dn = \hbar/(2\pi k_B T) d\omega$, the corresponding interaction free energy is

$$G(\ell, \theta) = -\frac{\hbar}{128\pi^2} \frac{\pi^2 R_1^2 R_2^2}{c^4 \sin \theta} \int_0^\infty d\omega \omega^4 \Delta_{1,\parallel}(i\omega) \Delta_{2,\parallel}(i\omega) \epsilon_m(i\omega) \int_0^\infty t dt \frac{e^{-2\sqrt{\epsilon_m(i\omega)} \frac{\omega}{c} \ell \sqrt{t^2+1}}}{(t^2+1)} \tilde{g}(t, a_1(i\omega), a_2(i\omega), \theta). \quad (19)$$

We now rework this equation to obtain the retarded result for the interaction between two semiconducting cylinders. Note here that we can not derive the Casimir limit properly as our formulation is not valid for nominally infinite zero-frequency (Drude-like) dielectric response. For that case see Ref. 11. First instead of variable ω , we introduce $x = \frac{\ell}{c} \sqrt{t^2+1} \omega$. Then, following closely the arguments in Ref. 14 we obtain the interaction free energy in the form

$$G(\ell, \theta) = -\frac{\hbar c}{128\pi^2} \frac{\pi^2 R_1^2 R_2^2}{\ell^5 \sin \theta} \epsilon_m(0)^2 \Delta_{1,\parallel}(0) \Delta_{2,\parallel}(0) \int_0^\infty dx x^4 \int_0^\infty t dt \frac{e^{-2\sqrt{\epsilon_m(0)} x}}{(t^2+1)^{7/2}} \tilde{g}(t, a_1(0), a_2(0), \theta). \quad (20)$$

Here $\epsilon_m(0)$ and $a_1(0), a_2(0)$ denote the static, i.e. zero frequency, values of the corresponding functions. Obviously in this regime the interaction free energy decays faster with separation, being a reflection of the retardation. All the frequency dependence of the material properties is reduced to the static response in this limit, just as in the general Lifshitz analysis [14].

IV. PARALLEL CYLINDERS

A. Parallel cylinders - full result

The analysis here is somewhat more complicated because the pair interaction energy between the cylinders involves the inverse Abel transform [15]. We start with

$$\frac{d^2 \mathcal{G}(\ell, \theta = 0)}{d\ell^2} = \frac{k_B T}{2\pi} \sum_{n=0}^{\infty} \int_0^{\infty} Q dQ \frac{d^2 f(\ell, \theta = 0)}{d\ell^2}, \quad (21)$$

where

$$\begin{aligned} \frac{d^2 f(\ell, \theta = 0)}{d\ell^2} = & -\frac{v_1 v_2 \Delta_{1,\parallel} \Delta_{2,\parallel}}{32} \frac{e^{-2\ell \sqrt{Q^2 + \epsilon_m \frac{\omega_n^2}{c^2}}}}{(Q^2 + \epsilon_m \frac{\omega_n^2}{c^2})} \\ & \left\{ 2 \left[(1 + 3a_1)(1 + 3a_2)Q^4 + 2(1 + 2a_1 + 2a_2 + 3a_1 a_2)Q^2 \epsilon_m \frac{\omega_n^2}{c^2} + 2(1 + a_1)(1 + a_2) \epsilon_m^2 \frac{\omega_n^4}{c^4} \right] + \right. \\ & \left. + (1 - a_1)(1 - a_2)(Q^2 + 2\epsilon_m \frac{\omega_n^2}{c^2})^2 \right\}, \end{aligned} \quad (22)$$

and again $v_1 = N \pi R_1^2$ ($v_2 = N \pi R_2^2$) and $a = \frac{2\Delta_{\perp}}{\Delta_{\parallel}}$. We continue by introducing the Abel transform and its properties. Namely, if we define

$$\int_{-\infty}^{+\infty} g(\sqrt{\ell^2 + y^2}) dy = f(y), \quad (23)$$

then

$$g(\ell) = -\frac{1}{\pi} \int_{\ell}^{+\infty} \frac{f'(y) dy}{\sqrt{y^2 - \ell^2}}. \quad (24)$$

Taking this into account when considering Eqs. 22, we remain with

$$g(\ell) = -\frac{k_B T}{32} R_1^2 R_2^2 \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} \int_{\ell}^{+\infty} \frac{dy}{\sqrt{y^2 - \ell^2}} \int_0^{\infty} Q dQ \frac{e^{-2y \sqrt{Q^2 + \epsilon_m (i\omega_n) \frac{\omega_n^2}{c^2}}}}{(Q^2 + \epsilon_m (i\omega_n) \frac{\omega_n^2}{c^2})^{1/2}} h(a_1(i\omega_n), a_2(i\omega_n), Q, \epsilon_m (i\omega_n) \frac{\omega_n^2}{c^2}), \quad (25)$$

where

$$\begin{aligned} h(a_1, a_2, Q, \epsilon_m \frac{\omega_n^2}{c^2}) = & 2 \left[(1 + 3a_1)(1 + 3a_2)Q^4 + 2(1 + 2a_1 + 2a_2 + 3a_1 a_2)Q^2 \epsilon_m \frac{\omega_n^2}{c^2} + 2(1 + a_1)(1 + a_2) \epsilon_m^2 \frac{\omega_n^4}{c^4} \right] + \\ & (1 - a_1)(1 - a_2)(Q^2 + 2\epsilon_m \frac{\omega_n^2}{c^2})^2. \end{aligned} \quad (26)$$

As before, we introduce $p_n^2 = \epsilon_m (i\omega_n) \frac{\omega_n^2}{c^2} \ell^2$, $u = Q\ell$ and $y \rightarrow y/\ell$. This allows us to rewrite the above integrals as

$$g(\ell) = -\frac{k_B T}{32} \frac{R_1^2 R_2^2}{\ell^5} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} \int_1^{+\infty} \frac{dy}{\sqrt{y^2 - 1}} \int_0^{\infty} u du \frac{e^{-2y \sqrt{u^2 + p_n^2}}}{(u^2 + p_n^2)^{1/2}} h(a_1(i\omega_n), a_2(i\omega_n), u, p_n^2), \quad (27)$$

and

$$\begin{aligned} h(a_1(i\omega_n), a_2(i\omega_n), u, p_n^2) = & 2 \left[(1 + 3a_1)(1 + 3a_2)u^4 + 2(1 + 2a_1 + 2a_2 + 3a_1 a_2)u^2 p_n^2 + 2(1 + a_1)(1 + a_2)p_n^4 \right] + \\ & (1 - a_1)(1 - a_2)(u^2 + 2p_n^2)^2. \end{aligned} \quad (28)$$

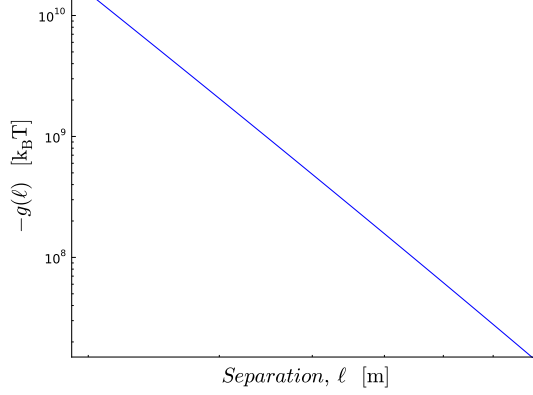


FIG. 6: Using equations 29 and 30, Full result for parallel cylinders as a function of separation

This is the final result for the interaction between two parallel thin cylinders at all separations and contains retardation effects explicitly. In general, the above expression can only be evaluated numerically once the dielectric spectra of component substances are known.

We now again transform this result into a form that is suitable for computation and numerical implementation. Rewriting Eq. 27 as

$$g(\ell) = -\frac{3(\pi R_1^2)(\pi R_2^2)}{8\pi \ell^5} \mathcal{A}(\ell), \quad (29)$$

we introduced the Hamaker coefficient

$$\mathcal{A}(\ell) = \frac{k_B T}{12\pi} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} \int_1^{+\infty} \frac{dy}{\sqrt{y^2 - 1}} \int_0^{\infty} u du \frac{e^{-2y\sqrt{u^2 + p_n^2(\ell)}}}{(u^2 + p_n^2(\ell))^{1/2}} h(a_1(i\omega_n), a_2(i\omega_n), u, p_n^2(\ell)) \quad (30)$$

with $h(a_1(i\omega_n), a_2(i\omega_n), u, p_n^2(\ell))$ defined in Eq. 28. This result is simpler than in the skewed case because it does not contain any angle dependence. In general $\mathcal{A}(\ell)$ can not be written in terms of $\mathcal{A}^{(0)}(\ell)$ and $\mathcal{A}^{(2)}(\ell)$ of the skewed cylinders.

B. Parallel cylinders - non-retarded result

In the non-retarded limit, $c \rightarrow \infty$, the above formula reduces to

$$\begin{aligned} g(\ell; c \rightarrow \infty) &= -\frac{k_B T}{32} \frac{R_1^2 R_2^2}{\ell^5} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} (3 + 5(a_1 + a_2) + 19a_1 a_2) \int_1^{+\infty} \frac{dy}{\sqrt{y^2 - 1}} \int_0^{\infty} u^4 du e^{-2yu} = \\ &= -\frac{9 k_B T}{(64 \times 32)\pi} \frac{\pi^2 R_1^2 R_2^2}{\ell^5} \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} \{3 + 5[a_1(i\omega_n) + a_2(i\omega_n)] + 19a_1(i\omega_n)a_2(i\omega_n)\}. \end{aligned} \quad (31)$$

Let us assume that the two cylinders are identical with radius a , so that we can finally write

$$g(\ell; c \rightarrow \infty) = -\frac{3}{(32^2)\pi} \frac{(\pi a^2)^2}{\ell^5} \mathcal{A}(\ell = 0), \quad (32)$$

where

$$\mathcal{A}(\ell = 0) = \frac{3}{2} k_B T \sum_{n=0}^{\infty} \Delta_{1,\parallel} \Delta_{2,\parallel} \left(3 + 5[a_1(i\omega_n) + a_2(i\omega_n)] + 19a_1(i\omega_n)a_2(i\omega_n) \right) \quad (33)$$

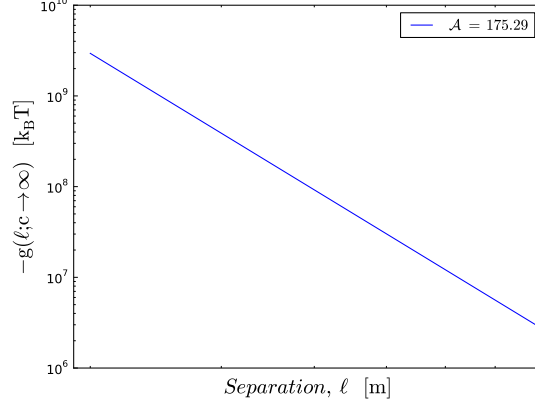


FIG. 7: Using equations 32 and 33, non-retarded result for parallel cylinders as a function of separation, Eq. 33 $\mathcal{A}(\ell = 0) = 175.29$.

For the case where the two interacting cylinders are composed of solid isotropic dielectric materials this form of the interaction free energy can be compared with the result obtained by Barash and Kyasov (Eq. 10 in Ref. 11) and can be reduced to it *exactly*. Again the $n = 0$ term of this formula for two identical cylinders corresponds to classical dipolar fluctuation forces as analyzed in Ref. 13.

With definitions Eq. 17

$$a_{1,2}(i\omega_n) = \frac{2\Delta_{\perp}^{(1,2)}(i\omega_n)}{\Delta_{\parallel}^{(1,2)}(i\omega_n)} = 2 \frac{(\epsilon_{\perp}^{c(1,2)}(i\omega_n) - \epsilon_m(i\omega_n))\epsilon_m(i\omega_n)}{(\epsilon_{\perp}^{c(1,2)}(i\omega_n) + \epsilon_m(i\omega_n))(\epsilon_{\parallel}^{c(1,2)}(i\omega_n) - \epsilon_m(i\omega_n))} \quad (34)$$

we then obtain

$$\mathcal{A}(\ell = 0) = \frac{3}{2} k_B T \sum_{n=0}^{\infty} \left(3 \Delta_{1,\parallel} \Delta_{2,\parallel} + 10 [\Delta_{\perp}^{(1)} \Delta_{2,\parallel} + \Delta_{\perp}^{(2)} \Delta_{1,\parallel}] + 76 \Delta_{\perp}^{(1)} \Delta_{\perp}^{(2)} \right) \quad (35)$$

This is exactly the same as obtained by Parsegian as cited in Ref. [22].

V. PARALLEL CYLINDERS - ZERO TEMPERATURE RESULT

As with skewed cylinders, we can take the zero temperature limit where the summation over the Matsubara frequencies becomes an integral over n with $dn = \hbar/(2\pi k_B T) d\omega$. Again we introduce $x = \frac{\ell}{c} \sqrt{t^2 + 1} \omega$. Then, as for skewed cylinders, we obtain the interaction free energy per unit length of two parallel cylinders,

$$g(\ell) = -\frac{\hbar c}{64\pi^3} \frac{\pi^2 R_1^2 R_2^2}{\ell^6} \epsilon_m(0)^{5/2} \Delta_{1,\parallel}(0) \Delta_{2,\parallel}(0) \int_0^{\infty} dx x^5 \int_1^{+\infty} \frac{dy}{\sqrt{y^2 - 1}} \int_0^{\infty} \frac{tdt e^{-2\sqrt{\epsilon_m(0)} yx}}{(t^2 + 1)^{7/2}} \tilde{h}(t, a_1(0), a_2(0)). \quad (36)$$

Here

$$\tilde{h}(t, a_1, a_2) = 2 [(1 + 3a_1)(1 + 3a_2)t^4 + 2(1 + 2a_1 + 2a_2 + 3a_1a_2)t^2 + 2(1 + a_1)(1 + a_2)] + (1 - a_1)(1 - a_2)(t^2 + 2)^2. \quad (37)$$

The spatial dependence is, again, one power higher in the retarded regime than in the non-retarded regime. All the frequency dependence of the material properties in the retarded limit is again reduced to the static response as in the Lifshitz analysis [14].

VI. SCREENED ZERO FREQUENCY TERM

Because of the presence of salt the zero frequency (classical) contribution to the Hamaker coefficients is screened. This means that instead of the Laplace equation one should take into account the linearized Debye-Huckel equation. This is of course approximate and more sophisticated statistical mechanical theories could be taken into account. Nevertheless we remain within the framework of the linearized Debye-Huckel theory.

Instead of going through the derivation once again for this case we note that within the DH approximation formally Eq. 7 would remain the same if we make the substitution $\epsilon_m \frac{\omega_n^2}{c^2} \rightarrow \kappa^2$, where κ^2 is the inverse Debye screening length. This means that the $n = 0$ term of Eq. 10 could be written as

$$G^{(0)}(\ell, \theta) = -\frac{k_B T}{64 \times 2\pi} \frac{(\pi R_1^2)(\pi R_2^2)}{\ell^4 \sin \theta} \Delta_{1,\parallel} \Delta_{2,\parallel} (\kappa \ell)^4 \int_0^\infty dt \frac{e^{-2(\kappa \ell) \sqrt{t^2+1}}}{(t^2+1)} \tilde{g}(t, a_1(0), a_2(0), \theta), \quad (38)$$

where we took into account that on the DH level $p_n^2(\ell) = \epsilon_m(i\omega_n) \frac{\omega_n^2}{c^2} \ell^2 \rightarrow \kappa^2 \ell^2$ and everything else remains unchanged.

Analogously the zero frequency term for the parallel cylinder case Eq. 27 would be modified to

$$g^{(0)}(\ell) = -\frac{k_B T}{32 \times 2\pi^2} \frac{(\pi R_1^2)(\pi R_2^2)}{\ell^5} \Delta_{1,\parallel} \Delta_{2,\parallel} \int_1^{+\infty} \frac{dy}{\sqrt{y^2-1}} \int_0^\infty u du \frac{e^{-2y\sqrt{u^2+(\kappa \ell)^2}}}{(u^2+(\kappa \ell)^2)^{1/2}} h(a_1(0), a_2(0), u, (\kappa \ell)^2), \quad (39)$$

and

$$h(a_1(0), a_2(0), u, (\kappa \ell)^2) = 2 \left[(1+3a_1)(1+3a_2)u^4 + 2(1+2a_1+2a_2+3a_1a_2)u^2(\kappa \ell)^2 + 2(1+a_1)(1+a_2)(\kappa \ell)^4 \right] + (1-a_1)(1-a_2)(u^2+2(\kappa \ell)^2)^2. \quad (40)$$

This completes the derivation of the van der Waals interactions between two anisotropic cylinders at all separations. On the lowest level one needs not take into account the above derivation for the zero frequency term but just skip it in the frequency summation. This would give correctly the vdW interactions at separations larger than the Debye length.

VII. INTERACTIONS AT SMALL SPACINGS

We take the main results from Ref. [21]. In the limit of small separations between the two cylinders $\ell/a \rightarrow 0$, we reformulate the approach based on the *Derjaguin method* and introduced for a single cylinder and a substrate. For closely opposed curved surfaces where c_1^1, c_1^2 are the principal curvatures of the surface 1 and c_2^1, c_2^2 are the principal curvatures of the surface 2, the *Derjaguin method* leads to the interaction energy of the form

$$G(\ell, \theta; a_1, a_2) = \int \int_{-\infty}^{+\infty} \mathcal{G}(\ell + \frac{1}{2}c_1x^2 + \frac{1}{2}c_2y^2) dx dy, \quad (41)$$

where c_1 and c_2 are defined as

$$c_1 c_2 = (c_1^1 c_2^1 + c_1^2 c_2^2) + (c_1^1 c_2^1 + c_1^2 c_2^2) \sin^2 \theta + (c_1^1 c_2^2 + c_1^2 c_2^1) \cos^2 \theta. \quad (42)$$

With polar variables the integral Eq. 41 can be rewritten as

$$G(\ell, \theta; a_1, a_2) = \int_0^{2\pi} \int_0^{+\infty} \mathcal{G}(\ell + \frac{1}{2}\rho^2) \frac{\rho d\rho d\phi}{\sqrt{c_1 c_2}}. \quad (43)$$

For two cylinders with radii a_1 and a_2 at an angle θ the above equations can be cast in the form

$$G(\ell, \theta; a_1, a_2) = \frac{2\pi \sqrt{a_1 a_2}}{\sin \theta} \int_\ell^\infty \mathcal{G}(h, \theta) dh. \quad (44)$$

This gives to the lowest order in the Δ 's

$$G(\ell, \theta; a_1, a_2) = -\frac{\sqrt{a_1 a_2} k_B T}{8\pi \ell \sin \theta} \sum_{n=0}^\infty \int_0^{2\pi} d\psi \Delta_{\mathcal{L}m}(\psi) \Delta_{\mathcal{R}m}(\theta - \psi). \quad (45)$$

The angular integral is again analytically solvable for any anisotropy and leads to the following result for the interaction free energy of two cylinders of equal radii $a_1 = a_2 = a$

$$G(\ell, \theta; a) = -\frac{a}{4} \frac{k_B T}{\ell \sin \theta} \left(\mathcal{H}^{(0)} + \mathcal{H}^{(2)} \cos^2 \theta \right), \quad (46)$$

where $\mathcal{H}^{(0)}$ and $\mathcal{H}^{(2)}$ are obtained from

$$\mathcal{H}^{(0)} + \mathcal{H}^{(2)} = \frac{1}{2\pi} \sum_{n=0}^{\infty} \int_0^{2\pi} d\psi \left(\frac{\epsilon_{\perp}^c(\mathcal{R}) \sqrt{1 + \gamma^c(\mathcal{R}) \cos^2 \psi} - \epsilon_m}{\epsilon_{\perp}^c(\mathcal{R}) \sqrt{1 + \gamma^c(\mathcal{R}) \cos^2 \psi} + \epsilon_m} \right) \left(\frac{\epsilon_{\perp}^c(\mathcal{L}) \sqrt{1 + \gamma^c(\mathcal{L}) \cos^2 \psi} - \epsilon_m}{\epsilon_{\perp}^c(\mathcal{L}) \sqrt{1 + \gamma^c(\mathcal{L}) \cos^2 \psi} + \epsilon_m} \right) \quad (47)$$

and

$$\mathcal{H}^{(0)} = \frac{1}{2\pi} \sum_{n=0}^{\infty} \int_0^{2\pi} d\psi \left(\frac{\epsilon_{\perp}^c(\mathcal{R}) \sqrt{1 + \gamma^c(\mathcal{R}) \cos^2 \psi} - \epsilon_m}{\epsilon_{\perp}^c(\mathcal{R}) \sqrt{1 + \gamma^c(\mathcal{R}) \cos^2 \psi} + \epsilon_m} \right) \left(\frac{\epsilon_{\perp}^c(\mathcal{L}) \sqrt{1 + \gamma^c(\mathcal{L}) \sin^2 \psi} - \epsilon_m}{\epsilon_{\perp}^c(\mathcal{L}) \sqrt{1 + \gamma^c(\mathcal{L}) \sin^2 \psi} + \epsilon_m} \right). \quad (48)$$

For two identical cylinders the \mathcal{L} and \mathcal{R} values are the same. Again we omit writing the explicit frequency dependence of all the dielectric functions. This dependence should be entered when numerical calculations are performed. Here again

$$\gamma^c = \frac{\epsilon_{\parallel}^c - \epsilon_{\perp}^c}{\epsilon_{\perp}^c}, \quad (49)$$

and ϵ_{\perp}^c and ϵ_{\parallel}^c are the transverse and longitudinal dielectric responses of the cylinder and ϵ_m that of the solution medium.

As above we now introduce the Hamaker coefficients according to the definitions

$$\mathcal{A}^{(0)} = \frac{3}{2} k_B T \mathcal{H}^{(0)} \quad \text{and} \quad \mathcal{A}^{(2)} = \frac{3}{2} k_B T \mathcal{H}^{(2)}, \quad (50)$$

and thus obtain for the interaction free energy of the two cylinders of equal radii $a_1 = a_2 = a$

$$\boxed{G(\ell, \theta; a) = -\frac{a}{6(\ell - 2a)} \frac{1}{\sin \theta} \left(\mathcal{A}^{(0)} + \mathcal{A}^{(2)} \cos^2 \theta \right).} \quad (51)$$

This is the final expression for the interaction free energy between two cylinders at a general angle θ and separation ℓ in the proximal limit.

Examine now the interaction free energy of two identical anisotropic cylinders of radius a at zero mutual angle. In this case the interaction free energy **per unit length** can be obtained in the form ([8], p.172)

$$g(\ell, \theta; a) = -\frac{k_B T \sqrt{a}}{16 \ell^{3/2}} \left(\mathcal{H}^{(0)} + \mathcal{H}^{(2)} \right), \quad (52)$$

where $\mathcal{H}^{(0)}$ and $\mathcal{H}^{(2)}$ are obtained in complete analogy to Eq. 47 and 48 from

$$\mathcal{H}^{(0)} + \mathcal{H}^{(2)} = \frac{1}{2\pi} \sum_{n=0}^{\infty} \int_0^{2\pi} d\psi \left(\frac{\epsilon_{\perp}^c(\mathcal{R}) \sqrt{1 + \gamma^c(\mathcal{R}) \cos^2 \psi} - \epsilon_m}{\epsilon_{\perp}^c(\mathcal{R}) \sqrt{1 + \gamma^c(\mathcal{R}) \cos^2 \psi} + \epsilon_m} \right) \left(\frac{\epsilon_{\perp}^c(\mathcal{L}) \sqrt{1 + \gamma^c(\mathcal{L}) \cos^2 \psi} - \epsilon_m}{\epsilon_{\perp}^c(\mathcal{L}) \sqrt{1 + \gamma^c(\mathcal{L}) \cos^2 \psi} + \epsilon_m} \right). \quad (53)$$

Of course, for two identical cylinders, the ϵ values for \mathcal{L} and \mathcal{R} are the same. Introducing again the Hamaker coefficient as before

$$\left(\mathcal{A}^{(0)} + \mathcal{A}^{(2)} \right) = \frac{3}{2} k_B T \left(\mathcal{H}^{(0)} + \mathcal{H}^{(2)} \right), \quad (54)$$

we get the interaction free energy per unit length of the parallel cylinders expressed through the surface to surface separation

$$\boxed{g(\ell, a) = -\frac{\sqrt{a}}{24(\ell - 2a)^{3/2}} \left(\mathcal{A}^{(0)} + \mathcal{A}^{(2)} \right).} \quad (55)$$

This is now the sixth and last result that we will use to quantify the van der Waals - London dispersion interaction between two parallel cylindrical CNTs at small separations.

A. Grosberg's interpolation formula

Let us now compare Eq. 32 and Eq. 55 for two equal cylinders of radius a . We have

$$g(\ell) = -\frac{3}{(32^2)\pi} \frac{(\pi a^2)^2}{\ell^5} \mathcal{A}(\ell=0), \quad g(\ell) = -\frac{\sqrt{a}}{24 (\ell - 2a)^{3/2}} \left(\mathcal{A}^{(0)} + \mathcal{A}^{(2)} \right). \quad (56)$$

Grosberg [23] showed that there is an interpolation formula for

$$\frac{g(\ell, a)\ell}{\beta^2} = \begin{cases} \left(\frac{\beta\pi}{4} \right)^2 & ; \beta \ll 1 \\ \left(\frac{\pi}{576} \right) (1 - 2\beta)^{-3/2} & ; 1 - 2\beta \ll 1 \end{cases} \quad (57)$$

with $\beta = a/\ell$, of the form

$$\frac{g(\ell, a)\ell}{\beta^2} = \left(\frac{\beta\pi}{4} \right)^2 (1 - 2\beta)^{-3/2} \left(1 - 2\beta + \frac{2\beta}{9\pi} \right) \quad (58)$$

Our formulas are however not of the Hamaker-type that Grosberg was using, so the Hamaker coefficient for the small and large separation limit are not the same. In our case we have rather

$$\frac{g(\ell, a)\ell}{\beta^2} = \begin{cases} \frac{3\pi}{32^2} \mathcal{A}(\ell=0) \beta^2 & ; \beta \ll 1 \\ \frac{1}{6\sqrt{2}} (\mathcal{A}^{(0)} + \mathcal{A}^{(2)}) (1 - 2\beta)^{-3/2} & ; 1 - 2\beta \ll 1 \end{cases} \quad (59)$$

Nevertheless one can still use the same philosophy to derive

$$\frac{g(\ell, a)\ell}{\beta^2} = \frac{3\pi}{32^2} \mathcal{A}(\ell=0) \beta^2 (1 - 2\beta)^{-3/2} \left(1 - 2\beta + \mathcal{R} \frac{32^2}{18\sqrt{2}\pi} (2\beta) \right) \quad (60)$$

where \mathcal{R} is the ratio

$$\mathcal{R} = \frac{\mathcal{A}^{(0)} + \mathcal{A}^{(2)}}{\mathcal{A}(\ell=0)}. \quad (61)$$

This should be used as an interpolation formula between the small and the large separation regimes. This can be written also as

$$g(\ell, a) = g_{>}(\ell, a) (1 - 2\beta)^{-1/2} + 32 \beta^5 g_{<}(\ell, a), \quad (62)$$

where $g_{>}(\ell, a)$ is the interaction free energy per unit length at large separations, $\beta \ll 1$, that goes into the non-retarded form still at large separations and $g_{<}(\ell, a)$ is the non-retarded (of course) interaction free energy at small separations $1 - 2\beta \ll 1$. This seems to complete the analysis.

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