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MICROSCOPIC THEORY OF RETARDED VAN DER WAALS FORCES BETWEEN MACROSCOPIC DIELECTRIC BODIES

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Synopsis

In a previous paper we derived an expression for the retarded Van der Waals interaction energy at zero temperature in an arbitrary system of atoms, where an atom was represented by an isotropic harmonic oscillator with one resonance frequency. Starting from this expression we evaluate in this paper the interaction energy between an atom and a semi-infinite dielectric medium, consisting of the same kind of atoms, and that between two dielectric halfspaces. The expressions for the interaction energy can be given in terms of the dielectric constant of the medium, and in this way formulae, earlier derived by Lifshitz from macroscopic considerations are recovered. Limiting formulae for both systems are obtained in the case of small and large separation of the two bodies; furthermore, the first correction (3-particle contribution) to the additive result (2-particle contribution) is given in these limiting cases for both systems.

1. *Introduction.* The presence of attractive Van der Waals forces between neutral atoms suggests that forces of this type do exist between macroscopic bodies too. The attractive force between two semi-infinite dielectric media, separated by a gap, was calculated by Lifshitz[1]) some years ago by means of macroscopic considerations. This calculation started from the idea that in the Maxwell equations for the macroscopic electromagnetic field inside a dielectric nonmagnetic medium a term should be inserted, accounting for a fluctuating electric field, the time average of which vanishes. Due to the special form of the spatial correlation functions for the components of the fluctuating field Lifshitz was able to calculate Maxwell's stress tensor at the boundaries of the bodies and from these he finally arrived at an expression for the attractive force between these bodies. This result could be simplified for the case of small, respectively large separations of the halfspaces (small and large with respect to the principal absorption wavelength of the medium), and in the limit of sufficiently rarefied media the expressions for the force between two individual atoms, as given by Eisenschitz and London[2]) and Casimir and Polder[3]), respectively, were recovered.

In later years some of the results, explicitly or implicitly contained in the original paper by Lifschitz, have been rederived by several authors, who avoided the introduction of a fluctuating field. McLachlan[4]) has shown that Lifshitz's results can be obtained by using the concept of field susceptibility; Mavroyannis[5]) has given approximate expressions (by means of S-matrix perturbation theory) for the interaction of an atom with a conducting wall and with a dielectric halfspace. Dzyaloshinskii *et al.*[6]), applying Green-function techniques, have extended the theory to include the case where the gap is filled by another dielectric medium. This latter problem has also been treated in a very simple way, valid for small distance between the bodies, by Van Kampen *et al.*[7]), by calculating the energy shift of the zero-point vibrations of the electromagnetic field. Very recently Langbein[8]) has given an approximate calculation for the retarded interaction between spheres by means of perturbation theory.

In earlier work[9,10]) we have calculated the non-retarded interaction between an isolated harmonic oscillator and a semi-infinite medium consisting of the same kind of oscillators, starting from a formula for the non-retarded interaction in an arbitrary system of oscillators; this latter formula was obtained by calculating the shift in zero-point energy of the oscillators as a consequence of electrostatic dipole–dipole interaction. Furthermore, in this way Lifshitz's formula for the non-retarded interaction between two semi-infinite dielectric media was rederived from microscopic considerations. In both cases the expression for the interaction energy was expanded in terms of many-particle interactions and the relative contributions of these interactions were discussed.

The present paper is an extension of the work just mentioned to include retarded interactions. Starting from an expression for the retarded interaction energy in a system of identical isotropic harmonic oscillators, which has been derived in a recent paper[11]), we calculate in section 2 the retarded interaction between an isolated oscillator and a semi-infinite dielectric medium† and in section 3 the retarded interaction between two semi-infinite dielectric media. Although the complete results can be given in a microscopic form in terms of the atomic positions, we shall introduce at some point in the calculation the macroscopic concept of the dielectric constant in order to derive the macroscopic formulae of Lifshitz. The three-particle correction to the additive result for the interaction energy will be discussed for the system atom–halfspace as well as for that of two halfspaces.

2. *Interaction of an atom with a semi-infinite dielectric medium.* a. In this section the retarded interaction energy between an isolated atom and a dielectric halfspace consisting of the same kind of atoms will be evaluated

† The interaction of an oscillator with a conducting wall has already been treated in ref. 11.

for arbitrary distance d between the atom and the surface of the medium (the only restriction is that d should be large compared to the interatomic distances in the medium). Some formulae to be derived in this section will prove very useful also in section 3, where the interaction energy between two semi-infinite media will be calculated.

Throughout this paper an atom will be represented by an isotropic harmonic oscillator with one resonance frequency ω_0. In a previous paper[11]) we derived an expression for the interaction energy ΔE_0 of an arbitrary number of such oscillators by calculating the difference in zero-point energy of the coupled system of oscillators and radiation field for a given configuration and a configuration where all the oscillators are infinitely far apart. The result could be written as [*cf.* (18) of ref. 11]:

$$\Delta E_0 = \frac{\hbar}{2\pi} \int_0^\infty \mathrm{d}\xi \ln \det[\mathbf{I} + \alpha(\mathrm{i}\xi)\, \mathsf{F}(\mathrm{i}\xi)], \tag{1}$$

where $\mathbf{I}$ is the $3N \times 3N$ unit matrix (N is the number of atoms), $\alpha(\mathrm{i}\xi)$ is the dynamical polarizability of an atom taken at an imaginary frequency:

$$\alpha(z) = \frac{e^2}{M} (\omega_0^2 - z^2 - \mathrm{i}\Gamma z)^{-1}, \qquad \Gamma = \frac{2e^2}{3Mc^3}\, \omega_0^2, \tag{2}$$

and $\mathsf{F}(\mathrm{i}\xi)$ is a $3N \times 3N$ matrix, built up from the 3×3 matrices $\mathsf{F}_{ab}(\mathrm{i}\xi)$, defined by:

$$\mathsf{F}_{ab}(z) = \left(\boldsymbol{\nabla}_a \boldsymbol{\nabla}_b - \frac{z^2}{c^2}\right) R_{ab}^{-1}\, \mathrm{e}^{\mathrm{i}zR_{ab}/c}, \tag{3}$$

where $R_{ab} = |\boldsymbol{R}_a - \boldsymbol{R}_b|$ is the distance between atoms a and b (the result of ref. 11 was originally expressed in terms of a matrix $\mathsf{G}(\mathrm{i}\xi)$; however, $\mathsf{F}(z) = \mathsf{G}(z)$ if z is in the upper half of complex plane [*cf.* (13), ref. 11]).

We now consider the configuration of one atom (labelled 0) at the position $\boldsymbol{R}_0$ outside a semi-infinite medium consisting of the same kind of atoms (at positions $\boldsymbol{R}_a$, $a = 1, \ldots, N$; $N \to \infty$) (see fig. 1) and we apply (1) to this

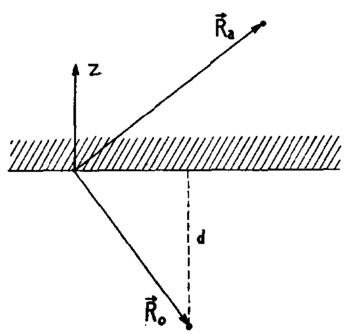

Fig. 1. One atom opposite a dielectric halfspace.

situation and to the situation where the distance d tends to infinity. The difference in interaction energy between these two configurations will then be interpreted as the interaction energy $U_1(d)$ of the isolated atom with the semi-infinite medium:

$$U_1(d) = \Delta E_0(d) - \lim_{d\to\infty} \Delta E_0(d).$$

If we decompose the $3(N+1) \times 3(N+1)$ matrix $\mathsf{F}(i\xi)$ into two parts, $\mathsf{F}(i\xi) = \mathsf{F}^{(1)}(i\xi) + \mathsf{F}^{(2)}(i\xi)$, where $\mathsf{F}^{(1)}(i\xi)$ describes the interaction between the atoms inside the medium only, and $\mathsf{F}^{(2)}(i\xi)$ that between the isolated atom and the atoms in the medium, then it will be clear from the definition (3) of $\mathsf{F}(i\xi)$ that $\mathsf{F}^{(2)}(i\xi)$ goes to zero as d tends to infinity. Therefore, we can write for the interaction energy $U_1(d)$:

$$U_1(d) = \frac{\hbar}{2\pi}\int_0^\infty d\xi\{\ln\det[\mathrm{I} + \alpha(i\xi)\,\mathsf{F}^{(1)}(i\xi) + \alpha(i\xi)\,\mathsf{F}^{(2)}(i\xi)]$$
$$- \ln\det[\mathrm{I} + \alpha(i\xi)\,\mathsf{F}^{(1)}(i\xi)]\}, \tag{4.a}$$

which alternatively may be written as

$$U_1(d) = \frac{\hbar}{2\pi}\int_0^\infty d\xi \ln\det\{\mathrm{I} + \alpha(i\xi)[\mathrm{I} + \alpha(i\xi)\,\mathsf{F}^{(1)}(i\xi)]^{-1}\cdot\mathsf{F}^{(2)}(i\xi)\}. \tag{4.b}$$

We now use (4.a) and the expansion:

$$\ln\det(\mathrm{I} + \mathsf{R}) = -\sum_{n=1}^{\infty} \frac{(-1)^n}{n}\,\mathrm{Tr}\,\mathsf{R}^n, \tag{5}$$

to obtain an expansion of $U_1(d)$ in terms of many-particle interactions:

$$U_1(d) \approx -\frac{\hbar}{2\pi}\sum_{n=2}^{\infty}(-1)^n\int_0^\infty d\xi[\alpha(i\xi)]^n$$
$$\times \sum_{\{a_i\}} \mathrm{Tr}[\mathsf{F}_{a_1a_2}(i\xi)\cdot\mathsf{F}_{a_2a_3}(i\xi)\,\ldots\,\mathsf{F}_{a_{n-2}a_{n-1}}(i\xi)\cdot\mathsf{F}_{a_{n-1}0}(i\xi)\cdot\mathsf{F}_{0a_1}(i\xi)], \tag{6}$$

where the summation over the a_i extends over the atoms in the medium only. The terms omitted in the expansion contain higher powers in $\mathsf{F}^{(2)}(i\xi)$; they may be neglected since they are of higher order in a/d, where a is a characteristic interatomic distance in the medium. (6) should be compared to (14) of ref. 9, which is the corresponding formula for the non-retarded case. (6) can be visualized by means of diagrams in which every factor F_{ab} is represented by a line connecting atoms a and b; the terms written down correspond to diagrams where the isolated atom is connected by two lines with the medium, whereas the terms which have been omitted correspond to diagrams with four and more lines between the isolated atom and the medium.

The 2-particle contribution $U_1^{(2)}(d)$ to $U_1(d)$ is given by the $n = 2$ term in (6):

$$U_1^{(2)}(d) = -\frac{\hbar}{2\pi}\int_0^\infty d\xi[\alpha(i\xi)]^2 \sum_a \mathrm{Tr}[\mathsf{F}_{a0}(i\xi)\cdot\mathsf{F}_{0a}(i\xi)];$$

it is simply the sum of all pair interactions between the isolated atom and atoms in the medium. If we replace the summation over particles by an integration this contribution can be written as:

$$U_1^{(2)}(d) = -\frac{\hbar\rho}{c^3}\int_0^\infty d\xi\ \xi^3[\alpha(i\xi)]^2\left[\frac{1}{2(\xi d/c)^3} + \frac{1}{(\xi d/c)^2} + 1 - 2\frac{\xi d}{c}\int_0^\infty \frac{e^{-p}}{p + 2(\xi d/c)}\, dp\right] e^{-2(\xi d/c)}, \tag{7}$$

where ρ is the particle density in the medium.

b. In order to be able to express $U_1(d)$ in terms of the dielectric constant ε (or equivalently in terms of the index of refraction $n = \sqrt{\varepsilon}$) we shall now investigate the response of the semi-infinite medium to the electromagnetic field of an oscillating dipole $\boldsymbol{\mu}_0(t)$ at the position $\boldsymbol{R}_0$. If we assume for this dipole a harmonic time dependence $:\boldsymbol{\mu}_0(t) = \boldsymbol{\mu}_0\, e^{-i\omega t}$, the equations for the induced dipole moments of the atoms in the medium read:

$$\boldsymbol{\mu}_a(t) = -\tilde{\alpha}(\omega)\ \mathsf{F}_{a0}(\omega)\cdot\boldsymbol{\mu}_0(t) - \tilde{\alpha}(\omega)\sum_{b\neq a}\mathsf{F}_{ab}\cdot\boldsymbol{\mu}_b(t), \qquad a = 1, \ldots, N, \tag{8}$$

where $\tilde{\alpha}(\omega) = \lim_{\varepsilon\to 0}\tilde{\alpha}(\omega + i\varepsilon)$ has been defined in ref. 11, eq. (16); we notice that $\tilde{\alpha}(\omega)$ can be obtained from (2), the expression for $\alpha(\omega)$, by replacing Γ by $\Gamma(\omega) \equiv (2e^2/3Mc^3)\,\omega^2$. (8) may be solved formally by putting:

$$\boldsymbol{\mu}_a(t) = -\tilde{\alpha}(\omega)\sum_b \{[\mathrm{I} + \tilde{\alpha}(\omega)\ \mathsf{F}(\omega)]^{-1}\}_{ab}\cdot\mathsf{F}_{b0}(\omega)\cdot\boldsymbol{\mu}_0(t). \tag{9}$$

On the other hand the induced atomic dipoles are related to the macroscopic polarization $\boldsymbol{P}(\boldsymbol{R}, t)$ and the macroscopic electric field $\boldsymbol{E}(\boldsymbol{R}, t)$ inside the medium by the equations:

$$\rho\boldsymbol{\mu}_a(t) = \boldsymbol{P}(\boldsymbol{R}_a, t) = \frac{\varepsilon(\omega) - 1}{4\pi}\boldsymbol{E}(\boldsymbol{R}_a, t), \tag{10}$$

where ρ is the particle density and $\varepsilon(\omega)$ the dynamical dielectric constant for the (isotropic) medium. Since the macroscopic electric field is a linear function of the dipole $\boldsymbol{\mu}_0(t)$, we can write:

$$\boldsymbol{E}(\boldsymbol{R}_a, t) = -\,\mathsf{M}_{a0}(\omega)\cdot\boldsymbol{\mu}_0(t), \tag{11}$$

where $\mathsf{M}_{a0}(\omega)$ is a 3×3 matrix to be calculated below. Comparison of (9) with (10) and (11) yields the equality:

$$\tilde{\alpha}(\omega) \sum_b \{[\mathrm{I} + \tilde{\alpha}(\omega)\,\mathsf{F}(\omega)]^{-1}\}_{ab}\cdot\mathsf{F}_{b0}(\omega) = \frac{\varepsilon(\omega) - 1}{4\pi\rho}\,\mathsf{M}_{a0}(\omega). \tag{12}$$

The matrix $\mathsf{M}_{a0}(\omega)$ can be calculated using the macroscopic Maxwell equations and the requirement that near the dipole $\boldsymbol{\mu}_0$ the field behaves as:

$$\boldsymbol{E}(\boldsymbol{R}, t) \approx \left(\boldsymbol{\nabla}\boldsymbol{\nabla} + \frac{\omega^2}{c^2}\right)\frac{\exp\left(\mathrm{i}\dfrac{\omega}{c}|\boldsymbol{R} - \boldsymbol{R}_0|\right)}{|\boldsymbol{R} - \boldsymbol{R}_0|}\cdot\boldsymbol{\mu}_0(t).$$

We describe the electromagnetic field outside the medium by the superposition of an outgoing wave from the dipole $\boldsymbol{\mu}_0$, and a wave reflected by the medium; inside the medium the field will be described by a refracted wave. We then decompose each wave into plane waves, using the representation[12]:

$$\exp\left(\mathrm{i}\frac{\omega}{c}|\boldsymbol{R} - \boldsymbol{R}_0|\right)/|\boldsymbol{R} - \boldsymbol{R}_0|$$

$$= \frac{\mathrm{i}}{\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{\mathrm{d}k_1\,\mathrm{d}k_2}{2k_3}\,\mathrm{e}^{\mathrm{i}k_1(x-x_0)}\,\mathrm{e}^{\mathrm{i}k_2(y-y_0)}\,\mathrm{e}^{\mathrm{i}k_3|z-z_0|},$$

$$k_3 = \left(\frac{\omega^2}{c^2} - k_1^2 - k_2^2\right)^{\frac{1}{2}}, \qquad \operatorname{Im} k_3 \geq 0, \tag{13}$$

which is valid if $\operatorname{Im}\omega > 0$ ($\operatorname{Im}\omega = 0$ is allowed if $\operatorname{Re}\omega \geq 0$). The usual boundary conditions for the electromagnetic field at the surface of a dielectric medium now give a relation for the amplitudes of every plane wave inside and outside the medium. From this relation the following expression for $\mathsf{M}_{a0}(\omega)$ can be derived (the magnetic permeability of the medium has been taken equal to unity):

$$\mathsf{M}_{a0}(\omega) = -\frac{\mathrm{i}}{\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{\mathrm{d}k_1\,\mathrm{d}k_2}{k_3 + k_3'}\,\mathrm{e}^{\mathrm{i}k_1(x_a - x_0)}\,\mathrm{e}^{\mathrm{i}k_2(y_a - y_0)}$$

$$\times\,\mathrm{e}^{\mathrm{i}k_3' z_a}\,\mathrm{e}^{-\mathrm{i}k_3 z_0}\left[\frac{\omega^2}{c^2}\boldsymbol{a}\boldsymbol{a} + \frac{k_3 + k_3'}{\varepsilon(\omega)\,k_3 + k_3'}\,\boldsymbol{b}^{(2)}\,\boldsymbol{b}^{(1)}\right], \tag{14}$$

where

$$\boldsymbol{a} = (k_1^2 + k_2^2)^{-\frac{1}{2}} (k_2, -k_1, 0),$$

$$\boldsymbol{b}^{(1)} = \boldsymbol{k}^{(1)} \times \boldsymbol{a}, \qquad \boldsymbol{k}^{(1)} = (k_1, k_2, k_3),$$

$$\boldsymbol{b}^{(2)} = \boldsymbol{k}^{(2)} \times \boldsymbol{a}, \qquad \boldsymbol{k}^{(2)} = (k_1, k_2, k_3').$$

$$k_3' = \left(\frac{\omega^2}{c^2}\,\varepsilon(\omega) - k_1^2 - k_2^2\right)^{\frac{1}{2}}, \qquad \mathrm{Im}\, k_3' \geq 0,$$

If $\omega = 0$ we find that

$$\mathsf{M}_{a0}(0) = \frac{2}{\varepsilon(0) + 1}\,\mathsf{T}_{a0},$$

where $\mathsf{T}_{a0} = \boldsymbol{\nabla}_a \boldsymbol{\nabla}_0 R_{a0}^{-1}$; this result can be obtained directly by applying the Maxwell equations to the case of an electrostatic dipole, as was done in the calculation of the non-retarded Van der Waals interaction[9,10]). It is easily verified from the properties[13]) of $\varepsilon(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega)$, where $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ are real, that both k_3 and k_3' have no branch points if ω is situated in the first quadrant of the complex plane, and that there they have a positive imaginary part. Furthermore, it can be shown that the integrand has no poles if ω is in the first quadrant, since $k_3 + k_3' \neq 0$ and $\varepsilon(\omega)\, k_3 + k_3' \neq 0$ in this case.

We wish to point out here that $\varepsilon(\omega)$ may have an imaginary part for real ω because of the imaginary part of the polarizability accounting for radiation damping; this is different from the treatment of the electrostatic problem in earlier papers[9,10]), where $\varepsilon(\omega)$ was real for real ω.

The above result for $\mathsf{M}_{a0}(\omega)$ allows us to extend (12) to the positive imaginary axis where, however, we have to replace $\tilde{\alpha}$ by α in order to avoid poles of the left-hand side of (12) on the imaginary axis (*cf.* the appendix of ref. 11):

$$\alpha(i\xi) \sum_b \{[\mathrm{I} + \alpha(i\xi)\, \mathsf{F}(i\xi)]^{-1}\}_{ab} \cdot \mathsf{F}_{b0}(i\xi)$$
$$= \frac{\varepsilon(i\xi) - 1}{4\pi\rho}\, \mathsf{M}_{a0}\,(i\xi). \tag{15}$$

Expansion of the matrix between curly brackets yields:

$$\frac{\varepsilon(i\xi) - 1}{4\pi\rho}\, \mathsf{M}_{a_1 0}(i\xi) = \alpha(i\xi)\, \mathsf{F}_{a_1 0}(i\xi) + \sum_{n=2}^{\infty} (-1)^{n-1}[\alpha(i\xi)]^n$$
$$\times \sum_{a_2, \ldots, a_n} \mathsf{F}_{a_1 a_2}(i\xi) \cdot \mathsf{F}_{a_2 a_3}(i\xi) \ldots \mathsf{F}_{a_{n-1} a_n}(i\xi) \cdot \mathsf{F}_{a_n 0}(i\xi). \tag{16}$$

c. The formulae derived in b will now be used to express the interaction energy $U_1(d)$ of the isolated atom and the medium in terms of the dielectric constant (for imaginary frequencies) of the medium. We might start either from (4b) and substitute (15) or, equivalently, from (6) and then substitute (16); to keep a close analogy to the non-retarded case we choose the latter way. Comparison of (6) with (16) shows that $U_1(d)$ can be written as:

$$U_1(d) = -\frac{\hbar}{2\pi}\int_0^\infty \mathrm{d}\xi\, \alpha(\mathrm{i}\xi)\, \frac{\varepsilon(\mathrm{i}\xi)-1}{4\pi\rho} \sum_{a_1} \mathrm{Tr}\, \mathsf{M}_{a_1 0}(\mathrm{i}\xi)\cdot \mathsf{F}_{0 a_1}(\mathrm{i}\xi). \tag{17}$$

It is obvious from the definition (3) of $\mathsf{F}(z)$ that we can use the representation (13) to express $\mathsf{F}_{0a}(\mathrm{i}\xi)$ as:

$$\mathsf{F}_{0a}(\mathrm{i}\xi) = \frac{\mathrm{i}}{\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \frac{\mathrm{d}k_1\,\mathrm{d}k_2}{2k_3} \times \mathrm{e}^{\mathrm{i}k_1(x_a-x_0)}\,\mathrm{e}^{\mathrm{i}k_2(y_a-y_0)}\,\mathrm{e}^{\mathrm{i}k_3(z_a-z_0)}\left(\boldsymbol{k}^{(1)}\boldsymbol{k}^{(1)} + \frac{\xi^2}{c^2}\right). \tag{18}$$

We now introduce in (17) for $\mathsf{M}_{a_1 0}(\mathrm{i}\xi)$ and $\mathsf{F}_{0a_1}(\mathrm{i}\xi)$ the expressions (14) and (18), respectively, and we replace the summation over the atoms in the medium by an integration; this yields:

$$\sum_{a_1} \mathrm{Tr}\, \mathsf{M}_{a_1 0}(\mathrm{i}\xi)\cdot \mathsf{F}_{0a_1}(\mathrm{i}\xi) = -4\pi\mathrm{i}\rho\frac{\xi^2}{c^2}\int_0^\infty \kappa\,\mathrm{d}\kappa\, \frac{\mathrm{e}^{-2\mathrm{i}k_3 z_0}}{k_3(k_3+k_3')^2} \times \left\{\frac{\xi^2}{c^2} - \frac{k_3+k_3'}{\varepsilon(\mathrm{i}\xi)\,k_3+k_3'}\left(\kappa^2 + k_3k_3' + 2\frac{c^2}{\xi^2}\kappa^2 k_3(k_3+k_3')\right)\right\}$$

where $\kappa^2 = k_1^2 + k_2^2$, and where the integration over the direction of (k_1, k_2) has already been performed. The integration over κ will now be transformed into an integration over the variable p where $p^2 = 1 + c^2\kappa^2/\xi^2$; with the abbreviation $s = (\varepsilon(\mathrm{i}\xi) - 1 + p^2)^{\frac{1}{2}}$ the expression (17) for $U_1(d)$ can finally be written as:

$$U_1(d) = -\frac{\hbar}{2\pi c^3}\int_0^\infty \mathrm{d}\xi\, \xi^3\alpha(\mathrm{i}\xi) \times \int_1^\infty \mathrm{d}p\, \mathrm{e}^{-2(\xi p d/c)}\left[\frac{s-p}{s+p} + (2p^2-1)\frac{\varepsilon p - s}{\varepsilon p + s}\right]. \tag{19}$$

This result for the interaction energy has been derived before byMcLachlan [4]) in a completely different way; the expression for the force, which may be found from this result by differentiation with respect to d, is in agreement with the expression for the force as it can be extracted from Lifshitz's formula. It should be noticed that this force is always attractive due to the fact that on the imaginary axis $\varepsilon(i\xi) \geq 1$.

In the following we shall investigate the properties of (19) in somewhat more detail. Let us first consider the case $\omega_0 d/c \ll 1$, where ω_0 is the resonance frequency of the atom. If ξ is replaced by $\omega_0 x$, we see that the exponential function in (19) decreases slowly with increasing p for not too large x, while on the other hand the term within square brackets behaves as $2p^2(\varepsilon-1)/(\varepsilon+1)$ for large p. Therefore, we may approximate the integrand for the integration over p by

$$2p^2 \frac{\varepsilon-1}{\varepsilon+1} \exp\left(-2\frac{\omega_0 d}{c} xp\right),$$

which yields for the interaction energy the expression for the non-retarded interaction obtained before [9]:

$$U_1(d) \approx -\frac{\hbar}{4\pi d^3} \int_0^\infty d\xi\, \alpha(i\xi) \frac{\varepsilon(i\xi)-1}{\varepsilon(i\xi)+1}, \qquad \omega_0 d/c \ll 1.$$

In ref. 10 the expansion of $U_1(d)$ in 2-, 3-, *etc.* particle contributions has been given; here we only quote the result for the first two contributions under the simplifying assumption that the relation between α and ε is given by the relation of Clausius–Mossotti: $(\varepsilon-1)/(\varepsilon+2) = \frac{4}{3}\pi\rho\alpha$:

$$U_1(d) \approx -\frac{\pi\hbar\omega_0\rho[\alpha(0)]^2}{8d^3}(1 - \tfrac{1}{2}\pi\rho\alpha(0) + \ldots), \qquad \omega_0 d/c \ll 1.$$

(In taking over this result from ref. 10 we have put $\Gamma = 0$). In the opposite case where $\omega_0 d/c \gg 1$ the exponential decreases very rapidly with increasing x, so that we may replace the arguments of α and ε by 0. The expression for the interaction energy can then be written as

$$U_1(d) \approx -\frac{3\hbar c\alpha(0)}{16\pi d^4}$$

$$\times \int_1^\infty \frac{dp}{p^4}\left[\frac{s(0)-p}{s(0)+p} + (2p^2-1)\frac{\varepsilon(0)\,p - s(0)}{\varepsilon(0)\,p + s(0)}\right], \qquad \omega_0 d/c \gg 1.$$

Performing the integration over p, we obtain $U_1(d)$ in the form given by Dzyaloshinskii *et al.* [(4.38) of ref. 6]. If again we assume the validity of the relation of Clausius–Mossotti the expansion of $U_1(d)$ in terms of many-particle contributions is found to be:

$$U_1(d) \approx -\frac{23\hbar c\rho[\alpha(0)]^2}{40d^4}\left(1 - \frac{370}{483}\pi\rho\alpha(0) + \ldots\right), \qquad \frac{\omega_0 d}{c} \gg 1.$$

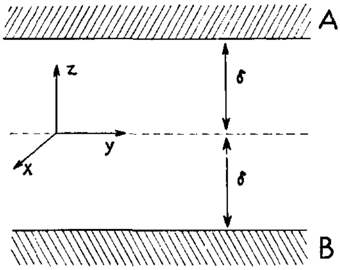

Fig. 2. Configuration of two halfspaces.

3. *Interaction between two semi-infinite dielectric media.* We shall use some of the formulae of section 2 to calculate the interaction energy of two identical dielectric halfspaces A and B, separated by a gap of width d. To keep the calculations symmetric with respect to A and B we choose the origin of the coordinate system halfway in the gap (see fig. 2). The matrix specifying the interaction between the atoms in A will be denoted by $\mathsf{F}_{AA}(i\xi)$, *etc.* The interaction energy per unit area, $U_2(d)$, is then given by (Ω is the surface area):

$$\Omega U_2(d) = \Delta E_0(d) - \lim_{d\to\infty} \Delta E_0(d)$$

$$= \frac{\hbar}{2\pi}\int_0^\infty d\xi \ln\det\begin{pmatrix} I + \alpha(i\xi)\,\mathsf{F}_{AA}(i\xi) & \alpha(i\xi)\,\mathsf{F}_{AB}(i\xi) \\ \alpha(i\xi)\,\mathsf{F}_{BA}(i\xi) & I + \alpha(i\xi)\,\mathsf{F}_{BB}(i\xi)\end{pmatrix}$$

$$- \frac{\hbar}{2\pi}\int_0^\infty d\xi \ln\det\begin{pmatrix} I + \alpha(i\xi)\,\mathsf{F}_{AA}(i\xi) & 0 \\ 0 & I + \alpha(i\xi)\,\mathsf{F}_{BB}(i\xi)\end{pmatrix}$$

$$= \frac{\hbar}{2\pi}\int_0^\infty d\xi \ln\det\begin{pmatrix} I & \alpha(i\xi)[I+\alpha(i\xi)\,\mathsf{F}_{AA}(i\xi)]^{-1}\cdot\mathsf{F}_{AB}(i\xi) \\ \alpha(i\xi)[I+\alpha(i\xi)\,\mathsf{F}_{BB}(i\xi)]^{-1}\cdot\mathsf{F}_{BA}(i\xi) & I\end{pmatrix}. \tag{20}$$

With the aid of (14) and (15) we arrive at the following equalities:

$$\alpha(i\xi)[I + \alpha(i\xi)\,\mathsf{F}_{AA}(i\xi)]^{-1}\cdot\mathsf{F}_{AB}(i\xi) = \frac{\varepsilon(i\xi) - 1}{4\pi\rho}\,\mathsf{M}_{AB}(i\xi),$$

$$\alpha(i\xi)[I + \alpha(i\xi)\,\mathsf{F}_{BB}(i\xi)]^{-1}\cdot\mathsf{F}_{BA}(i\xi) = \frac{\varepsilon(i\xi) - 1}{4\pi\rho}\,\mathsf{M}_{BA}(i\xi). \tag{21}$$

Here the $3N \times 3N$ matrix $\mathsf{M}_{AB}(i\xi)$ is built up from the 3×3 matrices $\mathsf{M}_{ab}(i\xi)(a \in A,\ b \in B)$, defined by [*cf.* (14)]:

$$\mathsf{M}_{ab}(\mathrm{i}\xi) = \frac{\mathrm{i}}{\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \frac{\mathrm{d}k_1\,\mathrm{d}k_2}{k_3 + k_3'}\, \mathrm{e}^{\mathrm{i}(k_3 - k'_3)\delta}\, \mathrm{e}^{\mathrm{i}k_1(x_a - x_b)}\, \mathrm{e}^{\mathrm{i}k_2(y_a - y_b)}$$

$$\times\, \mathrm{e}^{\mathrm{i}k'_3 z_a}\, \mathrm{e}^{-\mathrm{i}k_3 z_b}\left[\frac{\xi^2}{c^2}\,\boldsymbol{aa} - \frac{k_3 + k_3'}{\varepsilon(\mathrm{i}\xi)\,k_3 + k_3'}\,\boldsymbol{b}^{(2)}\,\boldsymbol{b}^{(1)}\right],$$

and the $3N \times 3N$ matrix $\mathsf{M}_{\mathrm{BA}}(\mathrm{i}\xi)$ from the 3×3 matrices $\mathsf{M}_{ba}(\mathrm{i}\xi)$ ($b \in \mathrm{B}$, $a \in \mathrm{A}$):

$$\mathsf{M}_{ba}(\mathrm{i}\xi) = \frac{\mathrm{i}}{\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \frac{\mathrm{d}k_1\,\mathrm{d}k_2}{k_3 + k_3'}\, \mathrm{e}^{\mathrm{i}(k_3 - k'_3)\delta}\, \mathrm{e}^{\mathrm{i}k_1(x_b - x_a)}\, \mathrm{e}^{\mathrm{i}k_2(y_b - y_a)}$$

$$\times\, \mathrm{e}^{-\mathrm{i}k_3' z_b}\, \mathrm{e}^{\mathrm{i}k_3 z_a}\left[\frac{\xi^2}{c^2}\,\boldsymbol{aa} - \frac{k_3 + k_3'}{\varepsilon(\mathrm{i}\xi)\,k_3 + k_3'}\,\boldsymbol{b}^{(4)}\boldsymbol{b}^{(3)}\right],$$

where

$$\boldsymbol{b}^{(3)} = \boldsymbol{k}^{(3)} \times \boldsymbol{a}, \qquad \boldsymbol{k}^{(3)} = (k_1, k_2, -k_3)$$

and

$$\boldsymbol{b}^{(4)} = \boldsymbol{k}^{(4)} \times \boldsymbol{a}, \qquad \boldsymbol{k}^{(4)} = (k_1, k_2, -k_3').$$

On substitution of the equalities (21) in (20) this expression becomes:

$$\Omega U_2(d) = \frac{\hbar}{2\pi}\int_0^{\infty} \mathrm{d}\xi \ln\det\begin{pmatrix} \mathbf{I} & \dfrac{\varepsilon(\mathrm{i}\xi) - 1}{4\pi\rho}\,\mathsf{M}_{\mathrm{AB}}(\mathrm{i}\xi) \\ \dfrac{\varepsilon(\mathrm{i}\xi) - 1}{4\pi\rho}\,\mathsf{M}_{\mathrm{BA}}(\mathrm{i}\xi) & \mathbf{I} \end{pmatrix}$$

$$\equiv \frac{\hbar}{2\pi}\int_0^{\infty} \mathrm{d}\xi\, \ln\det(\mathbf{I} + \mathsf{R}).$$

We now use again the expansion (5); since the trace of an odd power of R equals zero we only have to calculate the trace of the even powers. The first step in the calculation is then the evaluation of the matrix elements of $\mathsf{M}_{\mathrm{AB}}\cdot\mathsf{M}_{\mathrm{BA}}$ and of $\mathsf{M}_{\mathrm{BA}}\cdot\mathsf{M}_{\mathrm{AB}}$. We shall only mention the following result which can be obtained by replacing the summation over particles by an integration:

$$\mathrm{Tr}[\mathsf{M}_{\mathrm{AB}}(\mathrm{i}\xi)\cdot\mathsf{M}_{\mathrm{BA}}(\mathrm{i}\xi)]^n = \mathrm{Tr}[\mathsf{M}_{\mathrm{BA}}(\mathrm{i}\xi)\cdot\mathsf{M}_{\mathrm{AB}}(\mathrm{i}\xi)]^n$$

$$= \frac{\Omega}{4\pi^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathrm{d}k_1\,\mathrm{d}k_2 \left\{\left(\frac{4\pi\rho\xi^2}{c^2(k_3 + k_3')^2}\,\mathrm{e}^{2\mathrm{i}k_3\delta}\right)^{2n}\right.$$

$$\left. + \left(\frac{4\pi\rho(k_1^2 + k_2^2 - k_3k_3')}{(k_3 + k_3')[\varepsilon(\mathrm{i}\xi)\,k_3 + k_3']}\,\mathrm{e}^{2\mathrm{i}k_3\delta}\right)^{2n}\right\}$$

$$= \frac{\Omega}{2\pi}\,\frac{\xi^2}{c^2}\int_1^\infty \mathrm{d}p\,p\left\{\left(\frac{4\pi\rho}{(s+p)^2}\,\mathrm{e}^{-(\xi p d/c)}\right)^{2n} + \left(\frac{4\pi\rho(sp+p^2-1)}{(s+p)(s+\varepsilon p)}\,\mathrm{e}^{-(\xi p d/c)}\right)^{2n}\right\},$$

where the same substitutions have been made as in section 2.c:

$$p^2 = 1 + c^2\kappa^2/\xi^2 \qquad \text{and} \qquad s = (\varepsilon(\mathrm{i}\xi) - 1 + p^2)^{\frac{1}{2}}.$$

The calculation of $U_2(d)$ is now straightforward:

$$U_2(d) = -\frac{\hbar}{\pi\Omega}\int_0^\infty \mathrm{d}\xi \sum_{n=1}^\infty \frac{1}{2n}\left[\frac{\varepsilon(\mathrm{i}\xi)-1}{4\pi\rho}\right]^{2n} \times \mathrm{Tr}[\mathbf{M}_{\mathrm{AB}}(\mathrm{i}\xi)\cdot\mathbf{M}_{\mathrm{BA}}(\mathrm{i}\xi)]^n$$

$$= \frac{\hbar}{4\pi^2c^2}\int_0^\infty \mathrm{d}\xi\,\xi^2\int_1^\infty \mathrm{d}p\,p\left\{\ln\left[1-\left(\frac{s-p}{s+p}\right)^2\mathrm{e}^{-(2\xi p d/c)}\right] + \ln\left[1-\left(\frac{s-\varepsilon p}{s+\varepsilon p}\right)^2\mathrm{e}^{-(2\xi p d/c)}\right]\right\}. \tag{22}$$

Differentiation with respect to d yields an expression for the force per unit area, which agrees with Lifshitz's result.

We shall now consider the two limiting cases of small and large separation of the two bodies, and we shall evaluate the first correction to the additive interaction. For small distance, $\omega_0 d/c \ll 1$, we first integrate (22) by parts with respect to the p variable. There is one term in the expression, obtained in this way, which gives a dominant contribution for small distance. Retaining this term only and introducing a new variable $x = 2\xi p d/c$, we may write for $U_2(d)$:

$$U_2(d) \approx -\frac{\hbar}{32\pi^2 d^2}\int_0^\infty \mathrm{d}\xi\int_0^\infty \mathrm{d}x\,x^2\left\{\left[\frac{\varepsilon(\mathrm{i}\xi)+1}{\varepsilon(\mathrm{i}\xi)-1}\right]^2\mathrm{e}^x - 1\right\}^{-1}, \qquad \frac{\omega_0 d}{c} \ll 1.$$

This result was obtained in ref. 10, where also the expansion of $U_2(d)$ in terms of many-particle contributions has been given. In the case that the relation of Clausius–Mossotti holds the first two terms of this expansion read:

$$U_2(d) \approx -\frac{\pi\hbar\omega_0\rho^2[\alpha(0)]^2}{16d^2}\,(1 - \pi\rho\alpha(0) + \ldots), \qquad \frac{\omega_0 d}{c} \ll 1.$$

We observe that for large distance, $\omega_0 d/c \gg 1$, only small ξ values give an appreciable contribution to (22); therefore, we replace $\varepsilon(\mathrm{i}\xi)$ by $\varepsilon(0)$ and integrate the resulting expression by parts with respect to the ξ variable.

(22) can then be written in the following form:

$$U_2(d) \approx -\frac{\hbar c}{96\pi^2 d^3}\int_1^\infty \mathrm{d}p\int_0^\infty \mathrm{d}x\,\frac{x^3}{p^2}\left\{\left[\left(\frac{s(0)+p}{s(0)-p}\right)^2 \mathrm{e}^x - 1\right]^{-1} + \left[\left(\frac{s(0)+\varepsilon(0)\,p}{s(0)-\varepsilon(0)\,p}\right)^2 \mathrm{e}^x - 1\right]^{-1}\right\}, \qquad \frac{\omega_0 d}{c} \gg 1.$$

If we now expand $U_2(d)$ in many-particle interactions we find:

$$U_2(d) \approx -\frac{23\hbar c\rho^2[\alpha(0)]^2}{120\, d^3}\left(1 - \frac{740}{483}\,\pi\rho\alpha(0) + \ldots\right), \qquad \frac{\omega_0 d}{c} \gg 1,$$

where again the validity of Clausius–Mossotti has been assumed. One notices that both for small and large distance the relative 3-particle contribution to the interaction energy of two half-spaces is twice the relative 3-particle contribution in the case of an atom opposite a semi-infinite medium (*cf.* section 2.c).

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