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Interaction potential between small neutral particles and spherical or cylindrical surfaces

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Exact expressions for the interaction potential of a neutral particle with the surfaces of a spherical particle or pore, of a cylindrical filament or channel in matter, is obtained on the basis of the general theory of van der Waals forces for arbitrary distances from the particle to the surface. Simple analytic expressions are obtained for the potential near the center of a pore or near the axis of a cylindrical channel, near the surfaces or at large distances from them, in the case when retardation is neglected, or in the opposite limiting case.

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The van der Waals interaction of neutral atoms, molecules, and small particles with condensed-phase surfaces is important for the calculation of many statistical and kinetic characteristics: the adsorption energies of individual atoms, the chemical potential of an adsorbed layer, the thermodynamics and kinetics of a gas in pores and capillaries in solids. For plane surfaces the potential was considered in Refs. 1 and 2. These calculations were used to obtain the adsorption energy of inert-gas atoms on plane metallic surfaces.[3] Since the curvature of the surface is essential in many cases (micropores, microcapillaries, small particles or filaments of radius ~10^{-7}–10^{-5} cm), the dependences of the interaction potentials on the surface geometry were obtained for the particular case when the retardation is neglected.[4,5]

The purpose of the present paper is to construct the van der Waals interaction potential $U(r)$ of neutral molecules with spherical and cylindrical surfaces in the general case. We use here our earlier results on the thermodynamics of thin films on spherical and cylindrical surfaces,[6,7] and the reasoning proposed by Dzyaloshinskiĭ, E. Lifshitz, and Pitaevskiĭ[1] for planar surfaces.

Consider, for example the interaction of molecules with a spherical particles of radius R (see the figure). We imagine that the molecules form a rarefied gas of density n, filling the volume of the space outside the sphere of radius r. The dielectric constant of this gas is $\varepsilon_3=1+4\pi\alpha n$. In a spherical layer of radii R and r we have vacuum ($\varepsilon_2=1$). The sphere of radius R has dielectric constant ε. Consider the work that the gas performs when its boundary is displaced by Δr. On the one hand

$$W=\Delta\sigma(R,r,n)S\Delta r,\quad S=4\pi r^2. \tag{1}$$

Here $\Delta\sigma(R,r,n)$ is the force acting on a unit interface, and is equal to the discontinuity of the normal contention of the stress tensor of the fluctuating electromagnetic field.[1,5] On the other hand, neglecting the interaction between the gas particles

$$W=U(r)nS\Delta r. \tag{2}$$

Comparing (1) and (2) we get

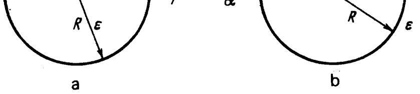

FIG. 1. a) Small particle with polarizability α at a distance r from the center of a sphere (from the axis of a cylinder) of radius R with dielectric constant E. b) The same particle inside a spherical (cylindrical) cavity of radius R in a medium with dielectric constant E.

$$U(r)=\lim_{n\to 0}\frac{\Delta\sigma(R,r,n)}{n}. \tag{3}$$

The reasoning for a spherical cavity or a cylindrical surface are similar to that advanced above, and Eq. (3) remains in force. We can thus use the results of our earlier studies[6,7] to calculate $U(r)$.

1. SPHERICAL CASE

Using the expression for $\Delta\sigma$ from Ref. (5) and Eq. (3), we obtain a general expression for the potential of a neutral molecule located at a distance r from the center of a spherical particle of radius $R(r>R)$:

$$U(r)=-T{\sum_{n=0}^{\infty}}' k_n^3\alpha\sum_{l=1}^{\infty}(2l+1)h_l(k_nr)\Big\{d_l(k_nR)-\Big[\varphi_l^2(k_nr)+\frac{l(l+1)}{k_n^2r^2}\Big]\tilde{d}_l(k_nR)\Big\}. \tag{4}$$

Here

$$d_l(x)=\frac{j_l(x)}{h_l(x)}\frac{\psi_l(x)-\zeta\psi_l(x\zeta)}{\varphi_l(x)-\zeta\psi_l(\zeta x)}$$

$$\tilde{d}_l(x)=\frac{j_l(x)}{h_l(x)}\frac{\zeta\psi_l(x)-\psi_l(\zeta x)}{\zeta\varphi_l(x)-\psi_l(\zeta x)},$$

$$\varphi_l(x)=\frac{d}{dx}\ln(xh_l(x)),\qquad \psi_l(x)=\frac{d}{dx}\ln(xj_l(x)),$$

$$h_l(x)=(\pi/2x)^{1/2}K_{l+1/2}(x),\qquad j_l(x)=(\pi/2x)^{1/2}I_{l+1/2}(x),$$

$$\zeta=\varepsilon^{1/2}(i\xi_n),\qquad \xi_n=2\pi nT/\hbar,\qquad k_n=\xi_n/c,\qquad \alpha=\alpha(i\xi_n);$$

the temperature T is measured in energy unit (with the Boltzmann constant $k_B=1$), c is the speed of light, h is Planck's constant, and I_p and K_p are modified Bessel functions of the first and second kind, respectively, while a primed summation sign means that the term with $n=1$ is taken with a coefficient $\frac{1}{2}$.

The dielectric constant $\varepsilon(i\xi_n)$ is obtained by analytic continuation of the usual dielectric constant $\varepsilon(\omega)$ to the imaginary, and is connected by a dispersion equation with the imaginary part of the dielectric constant, which is responsible for the absorptions:

$$\varepsilon(i\xi_n)=1+\frac{2}{\pi}\int_0^{\infty}\frac{\omega\varepsilon''(\omega)}{\omega^2+\xi_n^2}d\omega. \tag{5}$$

As usual, at temperatures $T\ll\hbar c/\lambda_0$, where λ_0 is the characteristic wavelength in the absorption spectrum, one can go from summation over n to integration, by using the substitution

$$T{\sum_n}'\to\frac{\hbar}{2\pi}\int d\xi.$$

The potential of a neutral molecule located at a distance r from the center of a spherical pore of radius $R(r<R)$ is obtained from (4) by making the substitution

$$j_l(x)\rightleftarrows h_l(x),\qquad \varphi_l(x)\rightleftarrows\psi_l(x). \tag{6}$$

We consider now some important limiting cases.

1. Near the surface of a spherical particle or pore, i.e., at $d=|R-r|\ll R$ the interaction potential is particularly large. In this case the main contribution to the sum over l is made by large l. Using the asymptotic forms of the modified Bessel functions with large indices,[8] we get

$$U(r)=U_0(d)\pm\frac{d}{R}U_1(d). \tag{7}$$

Here $U_0(d)$ is the potential near a plane surface[1,2]; the plus sign corresponds to interaction with a spherical pore, and the minus sign to interaction with a spherical particle. The second term in (7) yields the dimensional dependence of the correction.

In the general case

$$U_0(d)=-T{\sum_{n=0}^{\infty}}'k_n^3\alpha\int_1^{\infty}dp\exp(-2k_npd)\Big[(2p^2-1)\frac{\varepsilon p-s}{\varepsilon p+s}+\frac{s-p}{s+p}\Big], \tag{8}$$

$$U_1(d)=-\frac{T}{d}{\sum_{n=0}^{\infty}}'k_n^2\alpha\int_1^{\infty}\frac{dp}{p}\exp(-2k_npd)$$
$$\times\Big\{(2p^2-1)\frac{s-\varepsilon p}{s+\varepsilon p}\Big[(k_nd)^2(p^2-1)+\frac{p^2-1}{2p^2-1}-\frac{\varepsilon(p^2s^2-\varepsilon)}{s^2(\varepsilon p^2+p^2-1)}$$
$$-2k_nd\frac{\varepsilon(p^2-1)}{s(\varepsilon p^2+p^2-1)}\Big]-\frac{(p^2-1)(s-p)}{s^2(s+p)}(k_nds+1)^2\Big\}, \tag{9}$$
$$\varepsilon=\varepsilon(i\xi_n),\qquad s=(\varepsilon+p^2-1)^{1/2}.$$

In particular, at $d\ll\lambda_0$ we can neglect k_nd compared with l and we have

$$U_0(d)=-\frac{T}{2d^3}{\sum_{n=0}^{\infty}}'\alpha\frac{\varepsilon-1}{\varepsilon+1}, \tag{10}$$

$$U_1(d)=-\frac{T}{2d^3}{\sum_{n=0}^{\infty}}'\alpha\frac{(\varepsilon-1)(\varepsilon+2)}{(\varepsilon+1)^2}. \tag{11}$$

In the opposite limiting case $d\gg\lambda_0$ the main contribution to the sum over n is made by $\xi_n\sim c/d$. If $\varepsilon(i\xi_n)$ has only one characteristic frequency, then we can replace $\varepsilon(i\xi_n)$ in the expression for $U(r)$ by $\varepsilon_0=\varepsilon(0)$ and carry out the integration.

The expression for U_0 agrees with that previously obtained,[1] and U_1 is similar in form. Since these expressions are very complicated, we present them only for large ε_0, for example, in the important case of a metallic particle or of a pore in a metal ($\varepsilon_0\gg1$):

$$U_0(d)=-\frac{3\hbar c\alpha(0)}{8\pi d^4},\qquad U_1(d)=-\frac{13\hbar c\alpha(0)}{30\pi d^4}. \tag{12}$$

2. If "retardation is neglected," i.e., at $R\ll\lambda_0$ and $r\ll\lambda_0$, the arguments of the functions j_l and h_l are small compared with l and the general expression (4) goes over into the equations obtained by Schmeits and Lucas.[4] For a particle we have

$$U(r)=-\frac{T}{r^3}{\sum_{n=0}^{\infty}}'\alpha(\varepsilon-1)\sum_{l=1}^{\infty}\frac{l(2l+1)(l+1)}{\varepsilon l+l+1}\Big(\frac{R}{r}\Big)^{2l+1}. \tag{13}$$

For a pore1)

$$U(r)=-\frac{T}{r^3}\sum_{n=0}^{\infty}\alpha(\varepsilon-1)\sum_{l=1}^{\infty}\frac{l(2l+1)(l+1)}{\varepsilon l+l+\varepsilon}\Big(\frac{r}{R}\Big)^{2l+1}. \tag{14}$$

3. Far from the particle surface $(r\gg R)$ the potential decreases rapidly. In particular, $\xi_n\sim c/r$ at $r\gg\lambda_0$. Putting $\varepsilon(c/r)=\varepsilon_0$, we get

$$U(r)=-\frac{23\hbar c\alpha(0)(\varepsilon_0-1)}{16\pi R^4(\varepsilon_0+2)}\Big(\frac{R}{r}\Big)^7. \tag{15}$$

At $r\ll\lambda_0$ the potential $U(r)$ is obtained from (13) by retaining in the sum over l the term with $l=1$.

4. We determine the potential near the center of the sphere $(r\ll R)$ by expanding in the general expression for the pore potential (which is obtained from (4) by making the substitution (6) all the functions in powers of k_nr accurate to $(k_nr)^2$:

$$U(r)=U_2(R)+\frac{r^2}{R^2}U_3(R),$$

$$U_2(R)=2T\sum_{n=0}^{\infty}k_n{}^3\alpha\bar{C}_1(k_nR), \tag{16}$$

$$U_3(R)=-\frac{TR^2}{3}\sum_{n=0}^{\infty}{}'k_n{}^5\alpha[C_1(k_nR)-2\bar{C}_1(k_nR)].$$

C_l and $\bar{C}_l$ are obtained here from d_l and $\bar{d}_l$ by using (6). At $R\ll\lambda_0$, in particular, Eq. (16) goes over into (14), in which we must retain the terms with $l=1$ and $l=2$.

At $R\ll\lambda_0$ it is necessary to replace ε in (16) by ε_0. In particular for the potential of a particle near the center of a metallic cavity ($\varepsilon_0\gg1$) we obtain

$$U(r)=\frac{\hbar c\alpha(0)}{R^4}\left[1+5\left(\frac{r}{R}\right)^2\right]. \tag{17}$$

2. CYLINDRICAL CASE

For a cylindrical separation surface, the calculations are much more complicated, but even in this case we were able to obtain for $\Delta\sigma$ equations in closed form.[7] Substituting $\Delta\sigma$ in (3) and taking the limit, we obtain a general expression for the potential of a filament in the general case ($r>R$):

$$U(r)=-\frac{4T}{\pi r^3}\sum_{n=0}^{\infty}{}'\alpha\sum_{m=0}^{\infty}{}'\int_0^{\infty}dx\frac{I_m(z_1)K_m{}^2(z)}{\delta_0{}^0K_m(z_1)}\Big\{q^2(\delta_1{}^0+\delta_0{}^1)$$
$$+\frac{2}{z^2}(x^2\delta_1{}^0-q^2\delta_0{}^1)\left(m^2+z^2-\frac{z}{2}\frac{d}{dz}z\Phi_m(z)\right)$$
$$+4m^2q_1{}^2qx^2z_2{}^2(\varepsilon-1)\Phi_m(z)[\Phi_m(z_1)-\Psi_m(z_1)]\Big\}. \tag{18}$$

Here

$$\begin{gathered}
z=(x^2+q^2)^{1/2},\quad q=rk_n,\quad q_1=\rho q,\\
\rho=R/r,\quad z_1=\rho z,\quad z_2=\rho(x^2+q^2\varepsilon)^{1/2},\\
\delta_0{}^0=p^2+q_1{}^2z_1{}^3z_2{}^2(\Psi_{12}-\Phi_{21})(\varepsilon\Psi_{12}-\Phi_{21}),\\
\delta_1{}^0=p^2+q_1{}^2z_1{}^2z_2{}^2(\Psi_{12}-\Phi_{21})(\varepsilon\Psi_{12}-\Psi_{21}),\\
\delta_0{}^1=p^2+q_1{}^2z_1{}^2z_2{}^2(\Psi_{12}-\Psi_{21})(\varepsilon\Psi_{12}-\Phi_{21}),\\
\Psi_m(z)=\frac{d}{dz}\ln I_m(z),\quad \Phi_m(z)=\frac{d}{dz}\ln K_m(z),\\
p=\rho mxq_1{}^2(\varepsilon-1),\quad \Psi_{ik}=z_i\Psi_m(z_k),\quad \Phi_{ik}=z_i\Phi_m(z_k).
\end{gathered} \tag{19}$$

The formula for the potential of a particle in a cylindrical channel is obtained from (18) by making in all expressions the substitutions

$$I_m\rightleftarrows K_m,\quad \Phi_m\rightleftarrows\Psi_m. \tag{20}$$

We consider now some important limiting cases.

1. The potential near a cylindrical surface ($d=|R-r|\ll R$) is given by

$$U(r)=U_0(d)\pm\frac{d}{2R}U_1(d), \tag{21}$$

where $U_0(d)$ and $U_1(d)$, just for a sphere, are determined by Eqs. (10)–(12) respectively at $d\ll\lambda_0$ and $d\gg\lambda_0$. The reason why the size correction is half as large is apparently due to replacement of the average sphere curvature by the average cylinder curvature, which is half as large.

2. If retardation is neglected ($R, r\ll\lambda_0$) we obtain for the potential of a cylindrical filament

$$U(r)=-\frac{4T}{\pi r^3}\sum_{n=0}^{\infty}{}'\alpha\sum_{m=0}^{\infty}{}'\int_0^{\infty}dx\frac{I_m(\rho x)K_m{}^2(x)}{K_m(\rho x)}$$
$$\times(m^2+x^2+x^2\Phi_m{}^2(x))\frac{(\varepsilon-1)\Psi_m(\rho x)}{\varepsilon\Psi_m(\rho x)-\Phi_m(\rho x)}. \tag{22}$$

The potential in a cylindrical channel is obtained from (22) by using the substitution (20).

Apart from the symbols, these equations coincide with those of Ref. 5.

At large distances, when $r\gg R$ the potential decreases more slowly than in the spherical case. In particular, at $r\gg\lambda_0$ we get from (18), by retaining the first two terms in the sum over m and putting $\varepsilon(c/r)\approx\varepsilon_0\equiv\varepsilon(0)$,

$$U(r)=-\frac{14\hbar c\alpha(0)(\varepsilon_0-1)(\varepsilon_0+4)}{15\pi R^4(\varepsilon_0+1)}\left(\frac{R}{r}\right)^6. \tag{23}$$

At $R\ll r\ll\lambda_0$ it follows from (23) that

$$U(r)=-\frac{9\hbar}{128R^3}\left(\frac{R}{r}\right)^5\int_0^{\infty}\alpha\frac{(\varepsilon-1)(\varepsilon+7)}{\varepsilon+1}d\xi. \tag{24}$$

4. The potential near the axis of a cylindrical channel ($r\ll R$) can be determined by expanding in powers of r^2/R^2 the general equation for the potential, obtained from (18)–(19) by the substitution (20). In the general case the expression is quite cumbersome; we confine ourselves therefore to a particular case, $R\ll\lambda_0$ and $R\gg\lambda_0$. We assume

$$U(r)=U_4(R)+\frac{r^2}{R^2}U_5(R). \tag{25}$$

We then have at $R\ll\lambda_0$

$$U_4(R)=\frac{\hbar}{\pi^2R^3}\int_0^{\infty}\alpha(\varepsilon-1)d\xi\int_0^{\infty}x^2(F_0+F_1)dx,$$
$$U_5(R)=\frac{\hbar}{\pi^2R^3}\int_0^{\infty}\alpha(\varepsilon-1)d\xi\int_0^{\infty}x^4\left(\frac{3F_0}{4}+F_1+\frac{F_2}{4}\right)dx, \tag{26}$$
$$F_i=\frac{K_i(x)}{I_i(x)}\frac{\Phi_i(x)}{\Psi_i(x)-\varepsilon\Phi_i(x)}.$$

At $R\gg\lambda_0$ we must replace ε by ε_0. In particular for metals ($\varepsilon_0\gg1$)

$$U(r)=-\frac{3\hbar c\alpha(0)}{4R^4}\left[1+\frac{9}{2}\left(\frac{r}{R}\right)^2\right]. \tag{27}$$

3. DISCUSSION OF RESULTS

The equations obtained by us permit a numerical or analytic calculation of $U(r)$ in the case when $\alpha(i\xi_n)$, $\varepsilon(i\xi_n)$ are known. It may turn out that in those cases when the effects are considerable to any degree, $U(r)$ can be calculated with retardation neglected, i.e., by using Eqs. (13), (14), and (23) obtained in Refs. 4 and 5. Frequently, in fact, one uses for example (at sufficiently high frequencies) formulas of the plasma type for $\alpha(i\xi_n), \varepsilon(i\xi_n)$, which contain one characteristic frequency:

$$\alpha(i\xi_n)\approx\frac{\alpha_0}{1+\xi_n{}^2/\omega_0{}^2},\quad \varepsilon(i\xi_n)\approx1+\frac{\omega_p{}^2}{\xi_n{}^2}, \tag{28}$$

where ω_p is the plasma frequency and ω_0 is the characteristic frequency. If ω_p and ω_0 are properly chosen, such formulas lead to fair agreement with experiment, for example for the adsorption energy (if reasonable account is taken of the repulsion forces).[3] Since $\omega_0 \sim 10$ eV and $\omega_p \sim 10$ eV, which corresponds to $\lambda_0 \sim 10^3$ Å, we can neglect retardation at least for distances ~10–100 Å.

It must be remembered, however, that the spectrum of complex atoms has in fact, besides the main contribution of the outer electrons with low ionization energy, also deep lying levels with energies 100–1000 eV, corresponding to $\lambda_0 \sim 10$–100 Å. The total contribution of such levels to the potential $U(r)$ is estimated at several per cent to several dozen per cent. A manifestation of such a contribution is apparently the deviation from the d^{-3} law for the chemical potential of helium films on fluorite crystals, starting with film thicknesses ~100 Å.[9] We suggest therefore that for an accurate calculation of the interaction potential of molecules with spherical and cylindrical particles and with cavities of diameter 10–100 Å it is necessary in the general case to use our formulas.

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[1] Equation (14) of Schmeits and Lucas is in error: a factor 2 was left out in front of the expression for $2l+1$ under the summation sign [see Eq. (47) of Ref. 4].

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Relaxation processes in ferromagnetic metals

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The effect of the s–d exchange interaction on magnon and conduction-electron relaxation in ferromagnetic metals is investigated. It is shown that magnon scattering by electrons is the dominant relaxation process for long-wave magnons with wave vectors $k < k_0$ (k_0 is the threshold wave-vector value in the single-magnon interaction process). The expression obtained for the magnon damping allows the elucidation of the temperature-independent contribution to the ferromagnetic-resonance (FMR) line width at relatively high temperatures and the increase of the line width with decreasing temperature. The conductivity of a ferromagnetic metal at low temperatures, when electron scattering by two magnons is the dominant electron-relaxation process, is considered. It is also shown that the nonequilibrium long-wave magnons that arise under FMR conditions make an additional contribution to the electron and magnon relaxation that can be experimentally detected at attainable resonance-field amplitudes.

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1. INTRODUCTION

In ferromagnetic metals, the spin waves corresponding to the spatial oscillations of the d-electron magnetization interact most strongly with the conduction electrons (the s electrons). This interaction leads to relaxation in both the electron and magnon systems. In experiment, the relaxation in the electron system is reflected in the temperature dependence of the conductivity and other kinetic characteristics of the metal. The relaxation of the long-wave magnons is investigated in experiments by the method of ferromagnetic resonance (FMR), while the relaxation of the short-wave magnons are investigated in experiments with the aid of neutron scattering. The relaxation of the electron subsystem in ferromagnetic metals has been fairly fully investigated both theoretically and experimentally,[1] something which cannot be said about the relaxation of the long-wave magnons.

Extensive experimental data have thus far been accumulated on the FMR line width in different metals.[2-8] It has been found that the line width is determined by two main factors: inhomogeneous line broadening, which arises as a result of the finite depth of penetration of the high-frequency field into the metal (the "inhomogeneous exchange" mechanism)[9,10] and the relaxation contribution.[2,6] Experimentally, these two contributions reliably split up, the relaxation contribution predominating at relatively high frequencies. But the attempts that have been made to theoretically interpret the relaxation contribution cannot be considered to be satisfactory.[11-13] The relaxation channels that have been considered either have an incorrect temperature dependence, or yield for the FMR line width a value that is much smaller than the experimentally observed value.

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