

Chapter 3

Interactions induced by vacuum fluctuations

Overview

(These chapter notes are based on the papers Henkel & al. (2002); Novotny & Henkel (2008).)

Concepts from fluctuation electrodynamics are used to derive the Casimir-Polder interaction potential between electrically polarizable particles in an elementary way. We recover the exact result, discuss limiting cases and analyze the interaction between a pair of nanoparticles.

The Van der Waals interaction and its macroscopic counterpart, the Casimir interaction, are paradigmatic examples for mechanical forces induced by electromagnetic fluctuations (Craig & Thirunamachandran, 1984; Casimir & Polder, 1948). They are intriguing consequences of quantum mechanics and QED because they persist even at zero temperature where particles and fields are in the ground states. In many respects, however, the zero-point or vacuum fluctuations of the field behave similar to classical fields. For example, their interaction with polarisable objects is governed by the same scattering amplitudes that apply in the classical context (see Barton (1987) for an example). This viewpoint can be formalized further using the fluctuation-dissipation theorem (FDT) for quantum fields, derived by Callen & Welton (1951), as shown in detail by G. S. Agarwal in a series of papers (Agarwal, 1975a,b,c,d). The FDT provides an expres-

sion for the correlations of field fluctuations that is consistent with QED, but requires for their calculation the solution of a classical electrodynamics problem only. Zero-point and thermal noise can thus be handled on the same footing, which is useful for direct applications, as stressed by Henry & Kazarinov (1996).

In this chapter, we take the viewpoint of the FDT and provide a simple derivation of the Casimir-Polder interaction between polarizable particles. The standard QED derivation of the Casimir-Polder potential makes use of fourth-order perturbation theory (Craig & Thirunamachandran, 1984) and the many terms involved easily obscure physical intuition. On the other hand, our approach is physically intuitive because the interaction potential arises naturally from the mutual polarization of two particles in the presence of a fluctuating field. This picture demystifies the concept of quantum zero-point fluctuations. Our approach is also computationally efficient because one only requires the solution for the free space Green tensor without using any mode expansions. Furthermore, charge and field fluctuations enter separately and only the combination of both leads to the full formula for the Casimir-Polder interaction potential. Our calculations generalize those of Boyer (1969) to particles with dispersion and absorption which applies, for example, to metallic nanoparticles. This case is studied in detail for the purpose of illustration, focussing on the contribution of the dipole polarizability. Within this approach, we derive simple expressions for the total interaction energy and estimate its relevance on the nanometer scale for particles in a room-temperature bath.

3.1 Fluctuation induced forces

Electromagnetic fields mediate between charge fluctuations in separate bodies. The resulting charge correlations give rise to a force that is referred to as *dispersion* force. For short distances between the two bodies, the force is called *van der Waals-London* force, whereas at larger separations it is designated as the *Casimir-Polder* force. Although these forces are small on macroscopic scales they cannot be ignored on the scales of nanostructures. In this section we derive the force acting on a small polarizable particle in an arbitrary environment following the steps of Henkel & al. (2002).

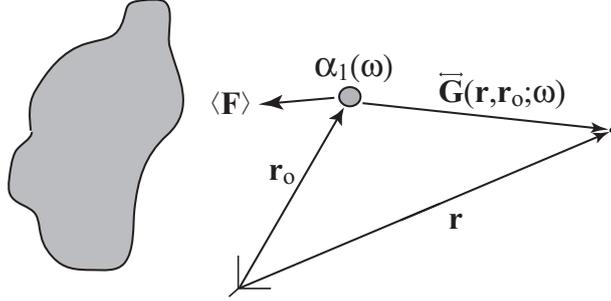


Figure 3.1: Dispersion force acting on a polarizable particle located at $\mathbf{r} = \mathbf{r}_o$. The force originates from correlated charge fluctuations in particle and other bodies in the environment. The latter are accounted for by the Green's function \mathbf{G} evaluated at its origin.

To calculate the force acting on a polarizable particle located at $\mathbf{r} = \mathbf{r}_o$ we use the well-known expression for the gradient force applicable for particles much smaller than the relevant wavelengths in the system (see for example Boyer (1969); Novotny & Hecht (2006)). We have to consider that both field \mathbf{E} and dipole moment \mathbf{D} have fluctuating and induced parts. Therefore,

$$\langle \mathbf{F}(\mathbf{r}_o) \rangle = \sum_i \left[\langle D_i^{(in)}(t) \nabla E_i^{(fl)}(\mathbf{r}_o, t) \rangle + \langle D_i^{(fl)}(t) \nabla E_i^{(in)}(\mathbf{r}_o, t) \rangle \right], \quad (3.1)$$

where $i = \{x, y, z\}$. The first term describes the (spontaneous and thermal) field fluctuations that correlate with the induced dipole according to

$$\hat{\mathbf{D}}^{(in)}(\omega) = \alpha_1(\omega) \hat{\mathbf{E}}^{(fl)}(\mathbf{r}_o, \omega), \quad (3.2)$$

where we assume an isotropic polarizability. For later purposes, we denote the properties of the particle by an index '1'. The second term in Eq. (3.1) originates from the particle's dipole fluctuations and the corresponding induced field according to

$$\hat{\mathbf{E}}^{(in)}(\mathbf{r}, \omega) = \frac{\omega^2}{c^2 \epsilon_0} \mathbf{G}(\mathbf{r}, \mathbf{r}_o; \omega) \cdot \hat{\mathbf{D}}^{(fl)}(\omega). \quad (3.3)$$

Here, \mathbf{G} is the Green's function of the field (calculated including other objects, but without the particle) and \mathbf{r} denotes an arbitrary field point as

visualized in Fig. 3.1. Correlations between fluctuating field and fluctuating dipole are zero because they originate from different physical systems. Likewise, there are no correlations between the induced quantities.

After expressing \mathbf{D} and \mathbf{E} in Eq. (3.1) by their Fourier transforms and making use of the fact that $\mathbf{E}(t) = \mathbf{E}^*(t)$ we obtain

$$\langle \mathbf{F}(\mathbf{r}_o) \rangle = \sum_j \iint_{-\infty}^{\infty} \langle \hat{D}_j^{(in)}(\omega) \nabla \hat{E}_j^{*(fl)}(\mathbf{r}_o, \omega') \rangle e^{i(\omega' - \omega)t} d\omega' d\omega + \quad (3.4)$$

$$\sum_j \iint_{-\infty}^{\infty} \langle \hat{D}_j^{(fl)}(\omega) \nabla \hat{E}_j^{*(in)}(\mathbf{r}_o, \omega') \rangle e^{i(\omega' - \omega)t} d\omega' d\omega . \quad (3.5)$$

Introducing the linear relationships (3.2) and (3.3) and arranging terms allows us to express the first term as a function of $\hat{\mathbf{E}}^{(fl)}$ and the second term as a function of $\hat{\mathbf{D}}^{(fl)}$ as

$$\begin{aligned} \langle \mathbf{F}(\mathbf{r}_o) \rangle &= \sum_j \iint_{-\infty}^{\infty} \alpha_1(\omega) \nabla_2 \langle \hat{E}_j^{(fl)}(\mathbf{r}_o, \omega) \hat{E}_j^{*(fl)}(\mathbf{r}_o, \omega') \rangle e^{i(\omega' - \omega)t} d\omega' d\omega \quad (3.6) \\ &+ \sum_{j,k} \iint_{-\infty}^{\infty} \frac{\omega'^2}{c^2 \epsilon_0} \nabla_1 G_{jk}^*(\mathbf{r}_o, \mathbf{r}_o; \omega') \langle \hat{D}_j^{(fl)}(\omega) \hat{D}_k^{*(fl)}(\omega') \rangle e^{i(\omega' - \omega)t} d\omega' d\omega , \end{aligned}$$

where ∇_n specifies that the gradient has to be taken with respect to the n -th spatial variable in the argument. Using the fluctuation-dissipation theorems for dipole and field [Eqs. (2.47) and (2.52)] and the fact that (the superscript ‘T’ denotes the transposed tensor)

$$\nabla_1 \mathbf{G}(\mathbf{r}, \mathbf{r}_o; \omega) = \nabla_2 \mathbf{G}^T(\mathbf{r}_o, \mathbf{r}; \omega) \quad (3.7)$$

allows us to write the force in the compact form

$$\langle \mathbf{F}(\mathbf{r}_o) \rangle = \sum_j \int_{-\infty}^{\infty} \frac{\omega}{\pi c^2 \epsilon_0} \left[\frac{\hbar \omega}{1 - e^{-\hbar \omega / k_B T}} \right] \text{Im} \left\{ \alpha_1(\omega) \nabla_1 G_{jj}(\mathbf{r}_o, \mathbf{r}_o; \omega) \right\} d\omega. \quad (3.8)$$

Notice, that the force is determined by the properties of the environment which is encoded in the Green’s function \mathbf{G} . The force vanishes in the absence of any objects, i.e. when \mathbf{G} equals the free space Green’s function. Eq. (3.8) allows us to calculate the force acting on a small polarizable

particle in an arbitrary environment. The equation is valid for an isotropic particle but it can be generalized to account for anisotropic polarizabilities such as for molecules with fixed transition dipole moments.

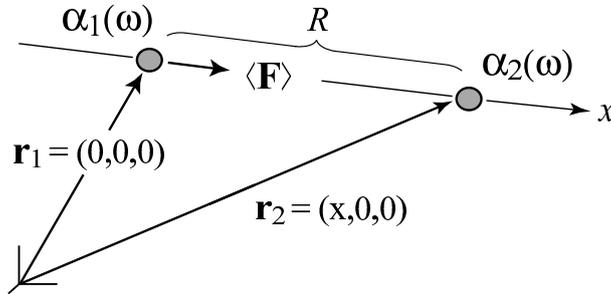


Figure 3.2: Definition of coordinates for the calculation of the dispersion force between two polarizable particles.

3.2 Two atoms: van der Waals–Casimir–Polder interaction

In this section we derive the force acting on a particle with polarizability α_1 due to another particle with polarizability α_2 . As indicated in Fig. 3.2, the two particles are separated by a distance R . For short distances, the force varies as R^{-7} whereas for larger distances the force assumes a R^{-8} dependence. The faster decay with distance at large distances is counter intuitive since electromagnetic fields acquire a longer range when going from the near-field ($1/R^3$) to the far field ($1/R$). It will be shown that for temperatures $T = 0$, the force at all distances can be deduced from a single potential $U(R)$, called the *Casimir-Polder potential*. Finite temperatures affect the force only at large distances where it is already fairly small (Henkel & al., 2002), and hence we will restrict the analysis to the case $T = 0$.

The force in Eq. (3.8) is determined by the Green's function \mathbf{G} . In the presence of a particle with polarizability $\alpha_2(\omega)$ located at \mathbf{r}_2 the Green's

function is calculated to first order as

$$\mathbf{G}(\mathbf{r}, \mathbf{r}_1; \omega) = \mathbf{G}^o(\mathbf{r}, \mathbf{r}_1; \omega) + \frac{\omega^2}{c^2 \varepsilon_0} \mathbf{G}^o(\mathbf{r}, \mathbf{r}_2; \omega) \alpha_2(\omega) \mathbf{G}^o(\mathbf{r}_2, \mathbf{r}_1; \omega), \quad (3.9)$$

where \mathbf{G}^o is the free-space Green's function. The gradient of \mathbf{G} evaluated at its origin $\mathbf{r} = \mathbf{r}_1$ is calculated as

$$\nabla_1 \mathbf{G}(\mathbf{r}_1, \mathbf{r}_1; \omega) = \frac{\omega^2}{c^2 \varepsilon_0} \alpha_2(\omega) [\nabla_1 \mathbf{G}^o(\mathbf{r}_1, \mathbf{r}_2; \omega)] \mathbf{G}^o(\mathbf{r}_2, \mathbf{r}_1; \omega). \quad (3.10)$$

Let us choose the coordinates as $\mathbf{r}_1 = \mathbf{0}$ and $\mathbf{r}_2 = (x, 0, 0) = x \mathbf{e}_x$, with \mathbf{e}_x being the unit vector in x . We then obtain for the sum of the diagonal elements of $\nabla \mathbf{G}$

$$\sum_j \nabla_1 G_{jj}(\mathbf{r}_1, \mathbf{r}_1; \omega) = \frac{\omega^2}{c^2 \varepsilon_0} \alpha_2(\omega) \sum_{j,k} \left[\frac{\partial}{\partial x} G_{jk}^o(x \mathbf{e}_x, 0; \omega) \right] G_{jk}^o(x \mathbf{e}_x, 0; \omega), \quad (3.11)$$

where we made use of the properties of the free-space Green's function \mathbf{G}^o . Using the explicit form of \mathbf{G}^o in the above expression gives

$$\begin{aligned} \sum_i \nabla_1 G_{ii}(\mathbf{r}_1, \mathbf{r}_1; \omega) &= \frac{\omega^2}{c^2 \varepsilon_0} \frac{\exp(2ix\omega/c)}{8\pi^2 x^7} \alpha_2(\omega) \left[-9 + 18i \left(\frac{\omega}{c} x \right) + \right. \\ &\quad \left. 16 \left(\frac{\omega}{c} x \right)^2 - 8i \left(\frac{\omega}{c} x \right)^3 - 3 \left(\frac{\omega}{c} x \right)^4 + i \left(\frac{\omega}{c} x \right)^5 \right] \mathbf{e}_x \end{aligned} \quad (3.12)$$

We now introduce this Green's function into the force formula (3.8) which, for $T = 0$, reads as

$$\langle \mathbf{F}(x) \rangle = \frac{\hbar}{\pi c^2 \varepsilon_0} \int_0^\infty \omega^2 \operatorname{Im} \left\{ \alpha_1(\omega) \sum_i \nabla_1 G_{ii}(\mathbf{r}_1, \mathbf{r}_1; \omega) \right\} d\omega. \quad (3.13)$$

Here, we made use of the fact that contributions with negative frequencies vanish [c.f. Eq. (2.49)].

It is straightforward to show that $\nabla \times \langle \mathbf{F} \rangle = 0$ and hence the force is conservative. Therefore, $\langle \mathbf{F} \rangle$ is associated with a potential U defined as

$$\begin{aligned} U = - \int \langle F(x) \rangle dx &= \frac{\hbar}{16\pi^3 \varepsilon_0^2 x^6} \operatorname{Im} \int_0^\infty \alpha_1(\omega) \alpha_2(\omega) e^{2ix\omega/c} \times \\ &\quad \left[-3 + 6i \left(\frac{\omega}{c} x \right) + 5 \left(\frac{\omega}{c} x \right)^2 - 2i \left(\frac{\omega}{c} x \right)^3 - \left(\frac{\omega}{c} x \right)^4 \right] d\omega. \end{aligned} \quad (3.14)$$

We now substitute the integration variable as $\tilde{\omega} = \omega c$ and replace the interparticle distance by R . We then realize that the integrand is analytic in the upper half-space of the integration variable and that the integrand goes to zero as $|\tilde{\omega}| \rightarrow \infty$. Therefore, we can integrate along the imaginary axis using

$$\int_0^{\infty} f(\tilde{\omega}) d\tilde{\omega} = i \int_0^{\infty} f(i\eta) d\eta. \quad (3.15)$$

Combining these mathematical tricks we obtain for the interparticle potential

$$U = -\frac{\hbar c}{16\pi^3 \varepsilon_0^2 R^6} \int_0^{\infty} \alpha_1(i\eta) \alpha_2(i\eta) e^{-2\eta R} [3 + 6\eta R + 5(\eta R)^2 + 2(\eta R)^3 + (\eta R)^4] d\eta. \quad (3.16)$$

We made use of the fact that $\alpha_i(\Omega)$ is purely real on the imaginary axis $\Omega = i\eta$. Eq. (3.16) is the celebrated *Casimir-Polder* potential valid for any interparticle separations R . Our result agrees with rigorous calculations based on quantum electrodynamics using fourth-order perturbation theory (Craig & Thirunamachandran, 1984). Our model allows us to incorporate higher order corrections by adding additional interaction terms to the Green's function \mathbf{G} in Eq. (3.9), describing multiple scattering and higher-order multipoles. With spherical particles, for example, the latter can be found from the Mie series for the scattered field. The force can be retrieved from the potential using $\langle \mathbf{F} \rangle = -\nabla U$.

3.2.1 Short distances: van der Waals potential

It is interesting to evaluate the potential for the limiting cases of large and small interparticle distances. For short distances we retain only the first term in the bracket, set $\exp(-2\eta R) = 1$, and obtain

$$U(R \rightarrow 0) = -\frac{6\hbar}{32\pi^3 \varepsilon_0^2} \frac{1}{R^6} \int_0^{\infty} \alpha_1(i\xi) \alpha_2(i\xi) d\xi. \quad (3.17)$$

This is the *van der Waals* potential valid for short interparticle distances R . The potential depends on the dispersive properties of the particle polarizabilities and scales with the inverse sixth power of the particle

separation R .

3.2.2 Large distances: Casimir-Polder potential

To obtain the limit for large R , we make the substitution $u = \eta R$ in Eq. (3.16) which leads to the following expression for the interparticle potential

$$U = -\frac{\hbar c}{16\pi^3 \varepsilon_0^2 R^7} \int_0^\infty \alpha_1(icu/R) \alpha_2(icu/R) e^{-2u} [3 + 6u + 5u^2 + 2u^3 + u^4] du. \quad (3.18)$$

Then, in the large distance limit ($R \rightarrow \infty$), one can replace the polarizabilities by their static values $\alpha_i(0)$. After moving them out of the integral one obtains

$$U(R \rightarrow \infty) = -\frac{\hbar c}{16\pi^3 \varepsilon_0^2} \frac{\alpha_1(0) \alpha_2(0)}{R^7} \int_0^\infty e^{-2u} [3 + 6u + 5u^2 + 2u^3 + u^4] du. \quad (3.19)$$

Finally, using the equality

$$\int_0^\infty u^n e^{-2u} du = \frac{n!}{2^{n+1}} \quad \forall n \geq 0, \quad (3.20)$$

one can analytically perform the integration in Eq. (3.19). We then obtain the Casimir-Polder interparticle potential in the limit of large distances as

$$U(R \rightarrow \infty) = -\frac{23\hbar c}{64\pi^3 \varepsilon_0^2} \frac{\alpha_1(0) \alpha_2(0)}{R^7}. \quad (3.21)$$

This result is a pure manifestation of vacuum fluctuations and was first derived by Casimir & Polder (1948). It is remarkable that the potential scales with the inverse seventh power of the interparticle distance R . Thus, for large distances the force decays more rapidly than for short distances. This behavior differs from the distance dependence of the electromagnetic energy density whose fastest decay (R^{-6}) is close to the source. The Casimir potential depends only on the static ($\omega = 0$) polarizabilities of the particles and hence it does not matter what their spectral properties are.

It has to be emphasized that the Casimir-Polder potential (3.16) originates solely from zero-point fluctuations and it does not account

for thermal fluctuations. At finite, but not too high temperature, the potential (3.21) is no longer valid for distances larger than the thermal wavelength $\lambda_T \sim \hbar c/k_B T$ where it shows a cross over back to a R^{-6} power law. At room temperature and $R \ll \lambda_T \approx 10 \mu\text{m}$, thermal corrections to the force are usually more than one order of magnitude weaker than the forces associated with vacuum fluctuations (Henkel & al., 2002).

3.3 Interaction between nanoparticles

(This section was not discussed in the SS 2010 lecture. It based on Novotny & Henkel (2008).) As an example, let us consider the Casimir-Polder potential between two identical nanoparticles with radii a . The particles are made of many atoms. Therefore, as long as the size of the particles is larger than the electron mean-free path their linear response can be represented by a local, but dispersive, dielectric constant $\varepsilon(\omega)$. We first assume that ε is given by a general single Lorentzian function as

$$\varepsilon(\omega) = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\gamma\omega}, \quad (3.22)$$

where ω_p is the plasma frequency, ω_0 a resonance (transition) frequency, and γ a damping constant (decay rate).

Assuming that each particle is much smaller than the relevant wavelengths and that each particle responds to a homogeneous external field (due to the other particle), the polarizability can be represented in the quasi-static limit as

$$\alpha(\omega) = 4\pi\varepsilon_0 a^3 \frac{\varepsilon(\omega) - 1}{\varepsilon(\omega) + 2}. \quad (3.23)$$

Here, we assumed that the particle is surrounded by vacuum ($\varepsilon = 1$). To evaluate the Casimir-Polder potential according to Eq. (3.16) we need to express α in terms of the imaginary frequency $i c \eta$. Combining the expressions for ε and α , and defining $\tilde{\alpha} = \alpha(4\pi\varepsilon_0 a^3)^{-1}$ we obtain

$$\tilde{\alpha}(i c \eta) = \frac{\omega_p^2}{\omega_p^2 + [3\omega_0^2 + 3c\eta(c\eta + \gamma)]}. \quad (3.24)$$

Obviously, for any value of η in the range $[0.. \infty]$ this expression is smaller than one. For $\eta \rightarrow 0$, the maximum value is obtained for $\omega_0 = 0$, i.e. a Drude metal. Hence, we can expect that the Casimir-Polder potential is stronger for metal particles than for dielectric particles.

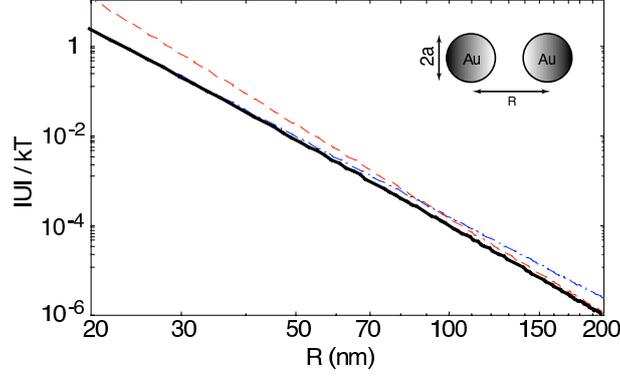


Figure 3.3: Casimir-Polder potential $U(R)$ between two gold nanoparticles with radii $a = 10\text{nm}$. The potential is normalized with the thermal energy kT ($T = 300\text{K}$). The particles' polarizabilities are given by Eqs.(3.22, 3.23) with $\omega_0 = 0$, $\omega_p = 1.38 \cdot 10^{16} \text{ rad/s}$ (9.1 eV). The maximum interaction energy depends on the plasma frequency ω_p . The dashed lines indicate the van der Waals potential ($\propto R^{-6}$) and the Casimir potential ($\propto R^{-7}$), respectively. The two lines intersect at $R \approx 90\text{nm}$.

Let us rewrite Eq. (3.16) in terms of $\tilde{\alpha}$ as

$$U = -\frac{\hbar c}{\pi} \frac{a_1^3 a_2^3}{R^6} \int_0^\infty \tilde{\alpha}_1(i\eta) \tilde{\alpha}_2(i\eta) e^{-2\eta R} [3 + 6\eta R + 5(\eta R)^2 + 2(\eta R)^3 + (\eta R)^4] d\eta. \quad (3.25)$$

At first sight it appears that U scales with the volumes of the particles. However this is not so because the interparticle distance $d = R - (a_1 + a_2)$ also scales with the particle sizes. The combination of these two effects yields a much weaker particle size dependence. For small interparticle distances, U reduces to the van der Waals potential (3.17). Assuming $a_1 = a_2 = a$, setting $R = d + 2a$ and expanding a^6/R^6 for small d yields

$$\max[U] = -\frac{3\hbar}{64\pi} \int_0^\infty \alpha_1(i\eta) \alpha_2(i\eta) d\eta \approx -\frac{\sqrt{3}}{256} \hbar \omega_p, \quad (3.26)$$

where, in the last step, we assumed that $\gamma \ll \omega_p$. This is a very interesting result as it predicts that the maximum value of the interaction potential between two nanoparticles is solely defined by the plasma frequency. The latter is a function of electron density n and effective electron mass m_e according to $\omega_p = \sqrt{ne^2/(m_e\epsilon_0)}$. For semiconductors, ω_p is typically in the infrared whereas for noble metals it is in the visible spectrum and in the UV. For example, for gold and silver the plasma frequency has the values $\omega_p = 1.38 \cdot 10^{16}$ rad/s (9.1 eV) and $\omega_p = 5.8 \cdot 10^{15}$ rad/s (3.8 eV), respectively. As evidenced by its small skin depth, aluminum has a particularly high plasma frequency of $\omega_p = 2.4 \cdot 10^{16}$ rad/s (15.6 eV). With these values the maximum interaction potential for metal nanoparticles is in the range of $\max[U] = -[1 \dots 4]k_B T$ at $T = 300\text{K}$. This is comparable to the energy associated with hydrogen bonds, slightly stronger than molecular van der Waals bonds ($\approx 0.5k_B T$) and much weaker than typical covalent bonds ($\approx 20k_B T$).

As an example, Fig. 3.3 shows the interaction potential between two gold particles with radii $a = 10\text{ nm}$. The curves are normalized by the thermal energy $k_B T$. As indicated by the dashed curves, at a distance $R \approx 70\text{ nm}$ the van der Waals potential and the Casimir potential are equal in strength. Close to contact ($R = 2a$) the interaction energy is comparable to the thermal energy but it drops rapidly with distance. Because each particle is excited locally by the other particle, the dipole-dipole theory applied here has limited validity in the range $R \leq 4a$. More accurate estimates in this regime need to consider higher multipole terms.

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