Long-range atom-surface interactions for cold atoms

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Abstract. Studies of the long-range interactions between cold atoms and surfaces are now of vital interest. The interest is partly driven by nanotechnology applications, partly by the exploding interest in the encompassing superfield of Casimir effects, and partly by the burgeoning overlap between atomic and molecular physics, condensed matter, and quantum optics. This tutorial lecture will address long-range atom-surface interactions for cold atoms, including an overview of Casimir-Polder interactions and their various manifestations. Some previous theoretical studies that are of particular relevance will be reviewed. In addition some different approaches to the problem and corresponding results, especially concerning the effects of substrate composition, geometry, and finite temperature, will be discussed.

1. Introduction
The interactions between atoms and surfaces are important in many areas of physics. Consequently, the literature is extensive and broad. In this tutorial, I will focus primarily on the long-range atom-surface interactions relevant to the atomic, molecular, and optical physics of cold atoms near surfaces. I will introduce the short-range Lennard-Jones potential and the long-range retarded Casimir-Polder potential, discuss the accurate calculation of the interaction coefficients and the calculation of the potentials for distances that are not too close to the surfaces, and the inclusion of real surface properties, such as dielectric response and temperature. The paper will cover theoretical aspects. A number of recent experiments were presented at the Conference on Atoms and Molecules near Surfaces, see the papers in the present volume, and Refs. 1, 2.

The interaction potential between a ground state atom and a perfectly conducting wall is

\[ V(R) = -C_3 R^{-3}, \quad (1) \]

where \( R \) is the distance between the atom and the wall, \( C_3 \) is the Lennard-Jones coefficient, and atomic units are used throughout (except when it is useful to exhibit \( \hbar \) and the speed of light \( c \)).

The interaction between polarizable systems is mediated by the exchange of virtual photons and as the separation of the systems increases there is a characteristic change in the interaction energy between them. In 1948, Casimir and Polder showed with quantum electrodynamics that for an atom interacting with a perfectly conducting wall the potential is

\[ V(R) = \frac{-3}{8\pi} \hbar c \alpha_d(0) R^{-4}, \quad R \to \infty, \quad (2) \]

1 The resonant interaction between a ground state atom and an excited state atom, e.g. a Na(3s) atom and a Na(3p) atom, will not be considered here.
where $\alpha_d(0)$ is the static electric dipole polarizability of the atom. Subsequent to Casimir and Polder, the result (2) has been obtained by many other investigators, see, for example, the comprehensive treatments in Refs. [4, 5, 6, 7, 8, 9, 10].

The appearance of $\hbar$ and $c$ and the weaker interaction (higher inverse power of $R$) are signatures of the Casimir-Polder potential. To understand the interaction potential (2) it is convenient to begin with a simple derivation of the retarded potential between two finite, polarizable systems that gives the correct expressions up to numerical constants. I follow the simple physical approaches presented in Refs. [11, 12, 13]; other approaches can be found in Refs. [14] and [15].

Suppose that the two polarizable systems (either might be one of atom, electron, ion, or wall) are in the presence of a uniform background (vacuum) field $E_b(\omega)$. The interaction energy of the two systems is given by

$$U(\omega, R) \sim \alpha_1(\omega)|E_{2\to1}(\omega, R) + E_b(\omega)|^2 + 1 \leftrightarrow 2,$$  

(3)

where the second term signifies the interchange of system 1 and system 2 in the first term and at large distances the electric field at system 1 coming from system 2 is

$$E_{2\to1}(\omega, R) \sim e^{iR\omega/c}\frac{\omega^2}{c^2R}p_2(\omega),$$  

(4)

where the electric dipole moment is

$$p_2(\omega) = \alpha_2(\omega)E_b(\omega)$$  

(5)

and the dynamic electric dipole polarizability is

$$\alpha(\omega) = \sum_u f_u/[(E_u - E_0)^2 - \omega^2],$$  

(6)

with $f_u$ the oscillator strength of state $u$ and $E_u - E_0$ the transition frequency between the states $u$ and $0$. Note that the “sum” in eq. (6) includes a sum over all discrete transitions and an integration over the continuum.

To obtain the $R$-dependent interaction energy, we retain only the cross terms in eq. (3) giving

$$U(\omega, R) \sim \alpha_1(\omega)E_{2\to1}(\omega, R)E_b(\omega) \sim \alpha_1(\omega)\alpha_2(\omega)E_b^2(\omega)e^{iR\omega/c}(\omega^2/c^2R).$$  

(7)

Summing over modes of the background field, replacing $\sum_k$ with $V \int d\omega \omega^2/c^3$ and $E_b^2(\omega)$ with $\hbar\omega/V$, and cutting off the integration at the highest relevant characteristic frequency, a simple integral expression for the potential is obtained [11]

$$U(R) \sim \frac{\hbar}{c^2R} \int_0^{c/R} d\omega \omega^5 \alpha_1(\omega)\alpha_2(\omega).$$  

(8)

This useful formula, eq. (8), can reproduce the asymptotic Casimir-Polder potentials for various cases such as the interaction between two atoms or between atom and a wall; its range of validity can be extended [16, 17]. In treating the asymptotic potential using eq. (8), we make the replacement $\alpha(\omega) \to \alpha(0)$. For example, the retarded Casimir-Polder potential between two atoms is, according to eq. (8), $U(R) \sim \hbar c\alpha_1(0)\alpha_2(0)R^{-7}$ in agreement with Casimir and Polder [3] who obtained

$$U(R) = -\frac{2\hbar}{4\pi}\hbar c\alpha_1(0)\alpha_2(0)R^{-7}, \quad R \to \infty,$$  

(9)
Spruch and Kelsey [11] showed how eq. (8) can reproduce the atom-wall interaction. Let the wall be approximated by a sphere of radius $CR$, where $C$ is a number less than one, perhaps around $\frac{1}{4}$, so that the systems are separated by a total distance $R + CR = R(1 + C)$. Then the polarizability of the sphere (wall) is $(CR)^3$, which when substituted into eq. (8) with the replacement of $R$ by $R(1 + C)$ (reasonable since $C \ll 1$) yields,

$$V(R) \approx C^3 \alpha_d(0) \frac{\hbar c}{R^4},$$

(10)

in agreement with eq. (2).

Can an expression more precise than eq. (10) for the coefficient in the asymptotic atom-wall interaction potential be obtained simply by integrating the asymptotic atom-atom potential eq. (9) over all the atoms constituting the wall? The calculation was carried out in Ref. [17] with the result

$$V(R) \approx -\frac{69}{160} \frac{\hbar c \alpha(0)}{\pi} R^{-4},$$

(11)

which is about 15% larger than the result of Casimir and Polder, eq. (2). [A similar discrepancy arises when the coefficient $C_3$ appearing in eq. (11) is estimated by integrating the $R^{-6}$ van der Waals interaction pairwise between an atom and each of the atoms in the wall [18].] The overestimation of the actual interaction coefficient is attributed to the non-additivity of long-range dispersion forces—the pairwise treatment does not account for three-body and higher-order interactions. The treatment of walls will be considered in sections 3 and 4 below.

2. Coefficients

We now leave the Casimir-Polder potential until section 4 and consider the accurate evaluation of the Lennard-Jones atom-wall interaction potential eq. (11) for separations sufficiently large that the exchange energy of the overlap between the atomic and surface wave functions (cf. [19]) is not important. For a perfectly conducting wall, the coefficient $C_3$ can be written as

$$C_3 = \frac{1}{4\pi} \int_0^\infty d\omega \alpha_d(i\omega),$$

(12)

or, from direct integration of eq. (12), as

$$C_3 = \frac{1}{12} \left\langle 0 \left| \left( \sum_{i=1}^{N_e} r_i \right)^2 \right| 0 \right\rangle,$$

(13)

where $|0\rangle$ is the wave function of the atom, $N_e$ is the number of electrons, and $r_i$ is the position vector from the nucleus to electron $i$.

The $C_3$ coefficients can be calculated in many ways, including $ab\ initio$ methods [20, 21], density functional theory (DFT) [22], and semiempirical methods [23, 18, 19, 24]. For H, the value of $C_3$ is $\frac{1}{4}$ [25]. Accurate values of $C_3$ have been obtained using $ab\ initio$ non-relativistic methods for Li [21] and He$(^3S)$ [20] and for the heavy alkali-metal atoms (Na, K, Rb, Cs, and Fr) using relativistic many-body perturbation theoretic methods [26, 27]. The most accurate $C_3$ values for some systems of interest for cold atom studies are summarized in Table 1.

Estimates of $C_3$ with eq. (12) using a single oscillator strength in eq. (6) can be inaccurate. For Li the value of $C_3$ is about 1.518 [21], but the value obtained using just the resonance transition is only 1.45, an underestimate of 4.5%. For Cs, the value of $C_3$ is about 4.45 [27], but the value obtained using just the resonance transition is 2.59, an underestimate of 42%. The discrepancy can be traced back to the contribution of the atomic core electrons, see eq. (13) [26]. Another way to see the origin of the discrepancy is to consider eq. (12), which is an integral from $0 < \omega < \infty$. Recall that for high frequencies,

$$\alpha_d(i\omega) \sim \sum_u f_u/\omega^2 = N_e/\omega^2, \quad \omega \sim \infty,$$

(14)
Table 1. Accurate values of the coefficient $C_3$ for the interaction of an atom in its ground state (except He($^23S$)) with a perfectly conducting wall, eqs. (11) and (12), in atomic units.

| Atom  | H  | He($^23S$) | Li | Na | K  | Rb | Cs | Fr |
|-------|----|------------|----|----|----|----|----|----|
| $C_3$ | 1.901 | 1.518 | 1.89 | 2.97 | 3.53 | 4.5 | 4.71 |
| Ref.  | [25] | [20] | [21] | [24] | [27] | [27] | [27] | [27] |

indicating that a representation of $\alpha(i\omega)$ that only includes the valence electron excitations ($N_e = 1$) will not have the proper high-frequency tail, thereby leading to an inaccurate $C_3$ coefficient from the integral in eq. (12). We ensured that a semi-empirical calculation of $C_3$ for Na [24] included the contribution of the (ionic) core electrons by requiring the oscillator strength distribution to satisfy the Thomas-Reiche-Kuhn sum rule. A related argument concerning inclusion of all virtual excitations was presented by Barton [28], who showed that a two-level atomic model is inadequate to describe energy shifts of an atom near a wall.

3. Ideal walls
For an atom and a perfectly conducting wall, an expression for the potential that is valid from small $R$ to asymptotically large $R$ is available [3, 29, 30],

$$V_{AtM}(R) = -\frac{C_3 f_3(R)}{R^3},$$

(15)

where the dimensionless retardation coefficient is

$$f_3(R) = \frac{1}{8C_3\pi\alpha_{fs}}\int_0^\infty dx \frac{e^{-x}}{\alpha_d(i\pi/2\alpha_{fs}R)}[\frac{1}{2}x^2 + x + 1],$$

(16)

and the subscript AtM denotes the atom-metal wall interaction [30], with $\alpha_{fs}$ the fine structure constant. Eq. (15) has the unretarded result eq. (11) as its limit for small $R$ and the Casimir-Polder result eq. (2) as its limit for large $R$.

For a wall with a dielectric constant $\epsilon$, the potential can be written [29, 30]

$$V_{ALD}(R, \epsilon) = -\frac{\alpha_{fs}^2}{2\pi}\int_0^\infty d\xi \xi^3 \alpha_d(i\xi)\int_1^\infty dp \exp(-2\xi R p \alpha_{fs})H[p, \epsilon(i\xi)],$$

(17)

where

$$H(p, \epsilon) = \frac{s - p}{s + p} + (1 - 2p^2)\frac{s - \epsilon p}{s + \epsilon p}$$

(18)

and

$$s = (\epsilon - 1 + p^2)^{1/2}$$

(19)

and the subscript $D$ denotes the dielectric wall.

Accurate Lennard-Jones coefficients and dynamic dipole polarizabilities for He($^23S$) have been used, for example, to theoretically analyze matter wave interference in an atomic trampoline [31], for a comparison to the experimental results of atomic diffraction from a silicon nitride grating [32], and in analysis of quantum reflection of atoms off of a flat polished Si surface [33].

In fig. 1 plots of accurate values of the atom-wall potentials $V_{ALD}(R, \epsilon)$, eq. (17), for a Li atom and a wall with $\epsilon = 2.123$, a wall with $\epsilon = 2.295$, and for a perfectly conducting wall.
\( V_{ATD}(R, \epsilon) \) for a Li atom and walls with \( \epsilon = 2.123, \epsilon = 2.295, \) and for a perfectly conducting wall \( (\epsilon = \infty) \) \cite{21}, in atomic units.

\( (\epsilon = \infty) \) \cite{21} are presented. Eq. \cite{17} can be readily evaluated for a wall characterized by a dielectric constant once an accurate dynamic electric dipole polarizability function is available. For short range, the effect of the dielectric wall on \( C_3 \) is a reduction by a factor,

\[
V_{ATD}(R, \epsilon) \to -\frac{1}{4\pi R^3} \int_0^\infty d\omega \alpha_d(i\omega) \frac{\epsilon(i\omega) - 1}{\epsilon(i\omega) + 1}, \quad \text{small } R. \tag{20}
\]

For a fixed dielectric constant, \( C_3 \) is reduced by the factor \([ (\epsilon - 1)/(\epsilon + 1) ]\), as is evident in fig. 1. A similar expression describes the reduction of the Casimir-Polder asymptotic potential for a dielectric wall \cite{29}.

4. Beyond ideal

Thus far, the consideration of the atom has been limited to the electric dipole polarizability and the surface has been considered to be either a perfect conductor or a material with a fixed dielectric constant. Actual surfaces could consist of real metals or dielectrics with frequency dependent properties, possess a nonzero surface temperature, have geometries deviating from a plane, and consist of layers of substrates. Also of interest is the consideration of higher electric multipoles or magnetic interactions and the treatment of an atom in an excited state or of a molecule. We now address these issues.

4.1. Finite temperature and frequency-dependent dielectric constant

Expressions for the atom-wall interaction, for a wall at finite temperature, valid at all but very short distances are available, cf. \cite{34, 5, 35, 36, 10, 37}. In considering finite temperature, a new distance scale appears, the thermal de Broglie wavelength \( \hbar c/k_B T \) of the photons. More formally, the energy in each photon field mode is replaced by the energy including the thermal photons

\[
\frac{1}{2} \hbar c k \rightarrow \frac{1}{2} \hbar c k + \hbar c [\exp(\hbar c k / k_B T) - 1]^{-1}. \tag{21}
\]

The expression for the atom-wall potential at finite temperature accordingly involves a Matsubara summation over frequencies, cf. \cite{38}. In the classical limit of high temperature, where the real photons dominate the virtual photons, the atom-wall potential becomes

\[
V(R, T) \sim -\frac{1}{4} k_B T \alpha_d(0)/R^3, \tag{22}
\]
and note the absence of $\hbar$. Spruch [39] has shown that the classical limit $RT \sim \infty$ simply arises from the replacement of $\frac{1}{2}\hbar kc$ by $k_B T$. This replacement combined with arguments similar to those leading to eq. (3), where we replaced $E_b^2(\omega)$ with $h\omega/V$ to account for virtual photons, would lead to the essential properties of eq. (22), see also Ref. [40]. For a dielectric material eq. (22) is diminished by the factor $[(\epsilon - 1)/(\epsilon + 1)]$ [10].

For surface temperatures of 300 K, evaluations of the interaction potentials for small distances are available for He($2^3S$) or Na atoms and an Au, Si, or SiO$_2$ wall [37] and evaluations for large distances are available for He($2^3S$), Na, or Cs atoms and an Au wall [36], and Rb atoms and a sapphire surface [10]. In addition, there is study of the interaction potential of H atoms near an Ag surface [35].

One ingredient of these calculations that I have not covered in this tutorial is the frequency dependent dielectric constant $\epsilon(i\omega)$—another topic with a vast literature. The reader is referred to Refs. [10] and [37], respectively, for example treatments of the $\epsilon(i\omega)$ function of sapphire and Au.

![Figure 2.](image)

The evaluation of the atom-wall potential at finite temperature for a real wall is illustrated in fig. 2 where the dimensionless multiplicative correction factor to the Casimir-Polder potential is plotted for a He($2^3S$) atom and a Au wall at 300 K [36]. The factor is the ratio of the atom-wall potential to the asymptotic Casimir-Polder potential of eq. (2), $-\frac{3}{8\pi}h\alpha_d(0)R^{-4}$. The linear dependence of the correction factor at small $R$ indicates the Lennard-Jones potential eq. (1) is a good approximation and the linear dependence of the correction factor at large $R$ indicates that the classical potential tail eq. (22) is a good approximation. The roughly flat behavior for separations between 1 and 3 microns indicates the applicability of the Casimir-Polder potential, eq. (2).

4.2. Surface roughness and layers
Surface roughness effects on the interaction of an atom and a wall have been considered, cf. Refs. [5, 41, 42]. The interaction of an atom with a substrate consisting of multiple layers was investigated in Ref. [8]. For thin layers, the power law describing the potential is predicted to be non-integer for certain cases, cf. Refs. [43] and [35].
4.3. Higher multipoles
In addition to the long-range potential arising from the induced electric dipole moment eq. (1), there is an induced quadrupole moment. The interaction potential will be weaker and is expected to drop off as the inverse fifth power. Some expressions and evaluations of coefficients are available in the literature, cf. Ref. [44].

4.4. Molecules
Studies of the interactions of diatomic molecules with surfaces along the lines of recent work with atoms is of interest, as advances in ultra-cold molecule science are continuing [45]. There will be two independent components for a diatomic molecule, similarly to a $P$-state atom interacting with a surface [2]. Theoretical expressions and evaluations of molecule-surface interaction coefficients treating the asymmetric part were given in Refs. [46, 47, 48, 22].

5. Other aspects
In the study of atom-wall interactions the surfaces are usually empirically described. Further developments might lead to ab initio calculation of surface material properties and atomic properties simultaneously, perhaps with density functional theory [22] or path-integral methods [49]. Another intriguing area of research is the repulsive Casimir force, which occurs in the interaction between a fluctuating electric dipole moment and a fluctuating magnetic moment [50].

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