Atoms near magnetodielectric bodies: van-der-Waals energy and Casimir-Polder force

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Abstract:

Based on macroscopic QED in linear, causal media, we present a consistent theory for the Casimir-Polder force acting on an atom positioned near dispersing and absorbing magnetodielectric bodies. The perturbative result for the van-der-Waals energy is shown to exhibit interesting new features in the presence of magnetodielectric bodies. To go beyond perturbation theory, we start with the center-of-mass equation of motion and derive a dynamical expression for the Casimir-Polder force acting on an atom prepared in an arbitrary electronic state. For a non-driven atom in the weak coupling regime, the force as a function of time is shown to be a superposition of force components that are related to the electronic density matrix elements at chosen time. These force components depend on the position-dependent polarizability of the atom that correctly accounts for the body-induced level shifts and broadenings.

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1 Introduction

Being a result of the vacuum fluctuations of the electromagnetic field, Casimir-Polder (CP) forces are experienced by any atomic system in the presence of magnetodielectric bodies. They play an important role in physical chemistry [1], and they hold the key to potential applications in micro- and nanotechnology such as the construction of atomic-force microscopes [2] or reflective atom-optical elements [3].

On short time scales and for weak atom-field coupling, the CP force is commonly derived from the van-der-Waals (vdW) energy calculated by means of time-independent perturbation theory [4-6] or linear response theory [7-9]. Following the former approach, but using a quantization scheme for the electromagnetic field in the presence of dispersing and absorbing magnetodielectric bodies, we derive a general expression for the vdW energy of an atom prepared in an energy eigenstate, where we focus on the influence of the magnetic properties.
The failure of perturbation theory is evident, because it cannot account for the internal dynamics present for an atom initially prepared in an excited state, and because the leading-order atomic polarizability, which essentially determines the CP force acting on an atom in the ground state, does not incorporate the body-induced energy shifts and broadenings, which can become drastic for small atom-surface separations. To overcome this deficiency, we present a dynamical treatment by basing the calculations on the well-known Lorentz force that governs the atomic center-of-mass equation of motion. The CP force is then obtained by taking the average of the Lorentz force with respect to the electronic quantum state of the atom and the electromagnetic vacuum, resulting in a dynamical expression that is valid for both strong and weak atom-field coupling. For the case of weak atom-field coupling, this expression is further evaluated with the aid of the Markov approximation.

2 Basic equations

Let us consider a neutral atomic system (e.g. an atom or a molecule) interacting with the electromagnetic field in the presence of linear, causal, magnetodielectric media and begin with the Hamiltonian

\[ \hat{H} = \hat{H}_A + \hat{H}_F + \hat{H}_{AF}. \]  

(1)

According to the multipolar coupling scheme [10],

\[ \hat{H}_A = \sum_\alpha \frac{\hat{p}_\alpha^2}{2m_\alpha} + \frac{1}{2\varepsilon_0} \int d^3 r \hat{P}_A^2(r) \]  

(2)

is the Hamiltonian for the atomic system consisting of particles \( \alpha \) with charges \( q_\alpha \), masses \( m_\alpha \), positions \( \hat{r}_\alpha \), and canonically conjugated momenta \( \hat{p}_\alpha \), where

\[ \hat{P}_A(r) = \sum_\alpha q_\alpha \hat{r}_\alpha \int_0^1 d\lambda \delta(r - \hat{r}_A - \lambda \hat{r}_\alpha) \]  

(3)

is the atomic polarization relative to the center of mass

\[ \hat{r}_A = \sum_\alpha \frac{m_\alpha}{m_A} \hat{r}_\alpha \]  

(4)

\( (m_A = \sum_\alpha m_\alpha) \),

\[ \hat{r}_\alpha = \hat{r}_\alpha - \hat{r}_A \]  

(5)

denoting shifted particle coordinates. The Hamiltonian \( \hat{H}_F \) characterizing the medium-assisted electromagnetic field is given by [11][12]

\[ \hat{H}_F \equiv \sum_{\lambda=e,m} \int d^3 r \int_0^\infty d\omega \hbar \omega \hat{f}_\lambda^\dagger(r, \omega) \hat{f}_\lambda(r, \omega), \]  

(6)

where the bosonic fields \( \hat{f}_\lambda(r, \omega) \) [and \( \hat{f}_\lambda^\dagger(r, \omega) \)],

\[ \left[ f_{\lambda i}(r, \omega), f_{\lambda i'}(r', \omega') \right] = \delta_{\lambda i} \delta_{ii'} \delta(r - r') \delta(\omega - \omega'), \quad \left[ f_{\lambda i}(r, \omega), f_{\lambda i'}(r', \omega') \right] = 0, \]  

(7)

play the role of the dynamical variables of the electromagnetic field plus the medium, where \( \lambda = e \) and \( \lambda = m \), respectively, refer to the electric and magnetic properties of the medium.
Finally, the multipolar-coupling Hamiltonian describing the interaction between the atomic system and the medium-assisted electromagnetic field in electric dipole approximation reads

\[
\hat{H}_{AF} = -\hat{d}\hat{E}(\hat{r}_A) + \frac{1}{2m_A}[\hat{p}_A, \hat{d} \times \hat{B}(\hat{r}_A)]_+ \tag{8}
\]

[\hat{E}(\hat{r})], electric field; [\hat{B}(\hat{r})], induction field; [\hat{a}, \hat{b}]_+ = \hat{a}\hat{b} + \hat{b}\hat{a}], with

\[
\hat{d} = \sum_\alpha q_\alpha \hat{r}_\alpha = \sum_\alpha q_\alpha \hat{\bar{r}}_\alpha \tag{9}
\]

being the electric dipole moment of the atomic system. Note that the second term on the right-hand side of Eq. (8) describes the Röntgen interaction due to the translational motion of the center of mass \[10\],

\[
\hat{p}_A = \sum_\alpha \hat{p}_\alpha \tag{10}
\]
denoting the total momentum of the atomic system.

The medium-assisted electric field [which in the multipolar coupling scheme has the physical meaning of a displacement field with respect to the atomic polarization \[3\] and the induction field can be related to the fundamental bosonic fields via \[11, 12\]

\[
\hat{E}(\hat{r}) = \int_0^\infty d\omega \hat{E}(\hat{r}, \omega) + \text{H.c.}, \tag{11}
\]

\[
\hat{B}(\hat{r}) = \int_0^\infty d\omega (i\omega)^{-1}\nabla \times \hat{E}(\hat{r}, \omega) + \text{H.c.}, \tag{12}
\]

\[
\hat{E}(\hat{r}, \omega) = \sum_{\lambda=e,m} \int d^3r' G_{\lambda}(\hat{r}, \hat{r}', \omega) \hat{f}_\lambda(\hat{r}', \omega). \tag{13}
\]

The c-number tensors

\[
G_\varepsilon(\hat{r}, \hat{r}', \omega) = i\frac{\omega^2}{c^2} \sqrt{\frac{\hbar}{\pi\varepsilon_0}} \text{Im} \varepsilon(\hat{r}', \omega) G(\hat{r}, \hat{r}', \omega), \tag{14}
\]

\[
G_m(\hat{r}, \hat{r}', \omega) = -i\frac{\omega}{c}\sqrt{-\frac{\hbar}{\pi\varepsilon_0}} \text{Im} \kappa(\hat{r}', \omega) \left[G(\hat{r}, \hat{r}', \omega) \times \nabla_{\hat{r}'} \right], \tag{15}
\]

where \([G(\hat{r}, \hat{r}', \omega) \times \nabla_{\hat{r}'}]_{ij} = \varepsilon_{ijk}\partial_l G_{lk}(\hat{r}, \hat{r}', \omega)\), \(\kappa(\hat{r}, \omega) = \mu^{-1}(\hat{r}, \omega)\), are given in terms of the (classical) Green tensor, which is defined by the differential equation

\[
\left[\nabla \times \kappa(\hat{r}, \omega) \nabla \times -\frac{\omega^2}{c^2} \varepsilon(\hat{r}, \omega)\right] G(\hat{r}, \hat{r}', \omega) = \delta(\hat{r} - \hat{r}') \tag{16}
\]
together with the boundary condition at infinity. Note that the (relative) permittivity \(\varepsilon(\hat{r}, \omega)\) and permeability \(\mu(\hat{r}, \omega)\) of the (inhomogeneous) medium are complex functions of frequency, whose real and imaginary parts satisfy the Kramers-Kronig relations. The Green tensor has the following useful properties \[11\],

\[
G^*(\hat{r}, \hat{r}', \omega) = G(\hat{r}, \hat{r}', -\omega^*), \tag{17}
\]

\[
G(\hat{r}, \hat{r}', \omega) = G^T(\hat{r}', \hat{r}, \omega), \tag{18}
\]

3
\[
\int d^3 s \left\{ \text{Im} \kappa(s, \omega) \left[ G(r, s, \omega) \times \bar{\nabla}_s \right] \left[ \nabla_s \times G^*(s', r', \omega) \right] \\
+ \frac{\omega^2}{c^2} \text{Im} \varepsilon(s, \omega) G(r, s, \omega) G^*(s, r', \omega) \right\} = \text{Im} G(r, r', \omega),
\]

(19)

where the last equation can be combined with the definitions (14) and (15) to yield the useful identity

\[
\sum_{\lambda=e, m} \int d^3 s G_{\lambda ii}(r, s, \omega) G^*_{\lambda ji}(r', s, \omega) = \bar{\hbar} \mu_0 \frac{\pi}{\omega^2} \text{Im} G_{ij}(r, r', \omega).
\]

(20)

3 The van-der-Waals energy

Let us consider an atomic system at rest (\( \hat{r}_A \rightarrow r_A \)) which is prepared in an energy eigenstate |l\rangle, i.e., an eigenstate of the Hamiltonian \( \hat{H}_A \) [Eq. (2)] written in the form

\[
\hat{H}_A = \sum_n E_n |n\rangle \langle n|,
\]

(21)

and calculate, within the framework of Schrödinger’s perturbation theory, the leading-order energy shift

\[
\Delta E_l = \Delta E_l^{(0)} + \Delta E_l^{(1)}(r_A)
\]

(22)

of the state |l\rangle|\{0\}\rangle, where |\{0\}\rangle denotes the ground state of the fundamental fields \( \hat{f}_\lambda(r, \omega) \).

According to Casimir’s and Polder’s pioneering concept [4], the position-dependent part \( \Delta E_l^{(1)}(r_A) \) of this energy shift can be interpreted as a potential energy

\[
U_l(r_A) = \Delta E_l^{(1)}(r_A),
\]

(23)

commonly called vdW energy, from which the CP force acting on the atom in the state |l\rangle can be derived according to

\[
\mathbf{F}_l(r_A) = -\nabla_A U_l(r_A)
\]

(24)

(\( \nabla_A \equiv \nabla_{r_A} \)). In this approach to the problem, the second term on the right-hand side of Eq. (24) can be disregarded, so that the interaction Hamiltonian that gives rise to the energy shift reduces to

\[
\hat{H}_{AF} = -\hat{d}\hat{E}(\hat{r}_A).
\]

(25)

3.1 General result

The bilinear form of the atom-field coupling Hamiltonian \( \hat{H}_{AF} \) implies that the leading-order energy shift is given by the second-order perturbative correction

\[
\Delta E_l = -\frac{1}{\hbar} \sum_k \sum_{\lambda=e, m} P \int_0^\infty \frac{d\omega}{\omega_{kl} + \omega} \int d^3 r |\langle l|\{0\}| - \hat{d}\hat{E}(\hat{r}_A)|\{1_\lambda(r, \omega)\}\rangle|k\rangle|^2
\]

(26)
\[ \Delta E_l = -\frac{\mu_0}{\pi} \sum_k \mathcal{P} \int_0^\infty d\omega \frac{\omega^2}{\omega_k^2 + \omega} d_l k \text{Im} G(r_A, r_A, \omega) d_k l \]  

(\(d_{lk} = \langle \hat{l}| \hat{d} |k \rangle\)). In order to extract the position-dependent part of the energy shift in accordance with Eq. \((22)\), we note that the atomic system should be located in a free-space region, where the Green tensor can be decomposed into the (translationally invariant) vacuum Green tensor \(G^{(0)}\) and the scattering Green tensor \(G^{(1)}\) that accounts for the presence of magnetodielectric bodies,

\[ G(r, r', \omega) = G^{(0)}(r, r', \omega) + G^{(1)}(r, r', \omega). \]  

The vdW energy \((23)\) can thus be obtained by making the replacement \([G(r_A, r_A, \omega) \rightarrow G^{(1)}(r_A, r_A, \omega)]\) in Eq. \((24)\). The result can be simplified by exploiting the property \((17)\) and the well-known asymptotic properties of the Green tensor for large frequencies (cf. Ref. \[12\]) to transform the integral along the real frequency axis into an integral along the imaginary frequency axis via contour-integral techniques, leading to

\[ U_l(r_A) = U_{l}^{\sigma}(r_A) + U_{l}^{r}(r_A), \]

where

\[ U_{l}^{\sigma}(r_A) = \frac{\mu_0}{\pi} \sum_k \int_0^\infty d\omega \frac{\omega_k u^2}{\omega_k^2 + u^2} d_l k G^{(1)}(r_A, r_A, iu) d_k l \]  

is the off-resonant part of the vdW potential, and

\[ U_{l}^{r}(r_A) = -\mu_0 \sum_k \Theta(\omega_{lk}) \omega_{lk}^2 d_l k \text{Re} G^{(1)}(r_A, r_A, \omega_{lk}) d_k l \]

\([\Theta(z), \text{unit step function}]\) is the resonant part due to the contribution from the residua at the poles at \(\omega = \pm \omega_{lk}\) for \(\omega_{lk} > 0\). Introducing the (lowest-order) atomic polarizability

\[ \alpha_{l}^{(0)}(\omega) = \lim_{\epsilon \to 0} \frac{2}{\hbar} \sum_k \frac{\omega_{lk}}{\omega_{lk}^2 - \omega^2 - i\epsilon} d_l k \otimes d_k l, \]

we may rewrite Eq. \((30)\) in the more compact form

\[ U_{l}^{\sigma}(r_A) = \frac{\hbar \mu_0}{2\pi} \int_0^\infty d\omega u^2 \text{Tr} [\alpha_{l}^{(0)}(iu) G^{(1)}(r_A, r_A, iu)]. \]

Finally, we note that for an atomic system in a spherically symmetric state we have

\[ \alpha_{l}^{(0)}(\omega) = \alpha_{l}^{(0)}(\omega) I = \lim_{\epsilon \to 0} \frac{2}{3\hbar} \sum_k \frac{\omega_{lk}}{\omega_{lk}^2 - \omega^2 - i\epsilon} |d_l k|^2 I \]

\((I, \text{unit tensor})\) and thus Eqs. \((30)\) and \((31)\) simplify to

\[ U_{l}^{\sigma}(r_A) = \frac{\hbar \mu_0}{2\pi} \int_0^\infty d\omega u^2 \alpha_{l}^{(0)}(iu) \text{Tr} G^{(1)}(r_A, r_A, iu), \]

\[ U_{l}^{r}(r_A) = -\frac{\hbar \mu_0}{3} \sum_k \Theta(\omega_{lk}) \omega_{lk}^2 |d_l k|^2 \text{Tr} [\text{Re} G^{(1)}(r_A, r_A, \omega_{lk})]. \]
Equation (29) together with Eqs. (31) and (33) is an extension of previous results to the case of arbitrary causal magnetodielectric bodies. It is worth noting that Eq. (29) also applies to left-handed material [13], for which standard quantization concepts run into difficulties. The calculations presented here can be regarded as the natural foundation for similar results obtained on the basis of (semi-classical) linear response theory for dielectric bodies [8].

Figure 1: The vdW energy $U_0(z_A)12\pi^2c/(\mu_0\omega_T^2 d_A^2)$ of a two-level atom in its ground state situated above a semi-infinite magnetodielectric half-space as a function of the distance between the body and the interface for different values of $\omega_{P,m}$ ($\omega_{P,m}/\omega_{T,m} = 0.75, \omega_{T,e}/\omega_{T,m} = 1.03, \gamma_e/\omega_{T,m} = \gamma_m/\omega_{T,m} = 0.001, \omega_{10}/\omega_{T,m} = 1$).

3.2 Ground-state atom in front of a magnetodielectric half-space

To illustrate the influence of the magnetic properties on the CP force acting on an atom near a magnetodielectric body, let us apply the theory to a two-level atom $|0\rangle$, ground state; $|1\rangle$, excited state; $\mathbf{d}_A \equiv d_{10} = d_A(\cos \phi \sin \theta \mathbf{e}_x + \sin \phi \sin \theta \mathbf{e}_y + \cos \theta \mathbf{e}_z)$ situated above ($z_A > 0$) a semi-infinite magnetodielectric half-space ($z < 0$). Using the appropriate scattering Green tensor as given, e.g., in Ref. [14], we find that

$$G^{(1)}(\mathbf{r}, \mathbf{r}', \omega) = \frac{i}{8\pi} \int_0^\infty dq \beta_0^2 e^{2i\beta_0 z^2} \left\{ r_s \left( \begin{array}{ccc} 1 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{array} \right) + r_p \frac{\epsilon^2}{\omega^2} \left( \begin{array}{ccc} -\beta_0 & 0 & 0 \\ 0 & -\beta_0 & 0 \\ 0 & 0 & 2q^2 \end{array} \right) \right\},$$

where

$$r_s = \frac{\mu \beta_0 - \beta}{\mu \beta_0 + \beta}, \quad r_p = \frac{\epsilon \beta_0 - \beta}{\epsilon \beta_0 + \beta}.$$
denote the reflection coefficients for s- and p-polarized waves, respectively ($\beta_0^2 = \omega^2/c^2 - q^2$ with \text{Im} \beta_0 > 0, $\beta^2 = \epsilon \mu \omega^2/c^2 - q^2$ with \text{Im} \beta > 0). Substituting this expression into Eq. (35) and recalling Eq. (34) [or equivalently using Eqs. (33) and (34) and averaging over all possible orientations of $d_A$], we derive the following formula for the ground-state vdW energy ($\beta_0 = i \epsilon_0$);

$$U_0(z_A) = \frac{\mu_0 \omega_{10} d_A^2}{12 \pi^2} \int_0^\infty \frac{du}{\omega_{10}^2 + u^2} \int_0^\infty dq \frac{q}{b_0} e^{-2b_0 z_A} \left\{ r_s - r_p \left[ 2 \left( \frac{cp}{\omega} \right)^2 + 1 \right] \right\}. \tag{39}$$

Based upon a single-resonance permittivity and a single-resonance permeability of Drude-Lorentz type,

$$\varepsilon(\omega) = 1 + \frac{\omega_{p,e}^2}{\omega_{T,e}^2 - \omega^2 - i \gamma_e \omega}, \quad \mu(\omega) = 1 + \frac{\omega_{p,m}^2}{\omega_{T,m}^2 - \omega^2 - i \gamma_m \omega}, \tag{40}$$

Fig. displays $U_0(z_A)$ as function of $z_A$ for different values of the magnetic plasma frequency $\omega_{p,m}$. Note that while for short distances the dominant influence of the dielectric properties always leads to the familiar attractive $1/z^3$-potential (cf. Sec. 4.3), strong magnetic properties can give rise a repulsive potential barrier at some intermediate distances.

4 The Casimir-Polder force

Due to the presence of magnetodielectric bodies the structure of the electromagnetic vacuum can drastically change, resulting in a body-induced shifting and broadening of atomic transition lines as the atom comes close the bodies. The perturbative results in Sec. 4 obviously fail to take such effects into account. Further, the enhanced spontaneous decay of an excited atom \cite{15} leads to dynamical effects that cannot be described within the frame of time-independent perturbation theory. In this section, we therefore use a quite distinct, non-perturbative approach to the problem, by basing the calculation of the CP force on the Lorentz force that governs the atomic center-of-mass motion.

4.1 General result

Using the multipolar Hamiltonian \cite{1} together with Eqs. (2), (6) and (8), and recalling the definitions (4) and (10) one can easily verify

$$m_A \ddot{\mathbf{r}}_A = \frac{i}{\hbar} [\hat{H}, m_A \hat{\mathbf{r}}_A] = \hat{\mathbf{p}}_A + \hat{\mathbf{d}} \times \hat{\mathbf{B}}(\mathbf{r}_A), \tag{41}$$

leading to

$$m_A \ddot{\mathbf{r}}_A = \mathbf{F} = \frac{i}{\hbar} [\hat{H}, \hat{\mathbf{p}}_A] + \frac{d}{dt} \left( \hat{\mathbf{d}} \times \hat{\mathbf{B}}(\mathbf{r}_A) \right) = \left\{ \nabla [\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(\mathbf{r})] + \frac{d}{dt} \left[ \hat{\mathbf{d}} \times \hat{\mathbf{B}}(\mathbf{r}) \right] \right\}_{\mathbf{r} = \mathbf{r}_A}, \tag{42}$$

where in the last step magnetic dipole terms have been dropped in consistency with the electric dipole approximation made and terms of the order of $O(v/c)$ ($v$, speed of the atomic center of mass) have been omitted in accordance with the non-relativistic Hamiltonian \cite{11}. Since $\hat{\mathbf{E}}(\mathbf{r})$ and $\hat{\mathbf{B}}(\mathbf{r})$ are defined according to Eqs. (11)–(13), Eq. (12) describes the Lorentz
force acting on an atomic system in the presence of absorbing and dispersing magnetodi-electric bodies and thus generalizes the well-known free-space result [16].

Next, we determine the temporal evolution of the medium-assisted electromagnetic field with the aid of Hamiltonian [11] together with Eqs. (2), (4), and (8). Recalling the definitions (11) and (13) and exploiting the commutation relations (11), we may write

\[ \hat{E}(r, \omega, t) = \hat{E}_{\text{free}}(r, \omega, t) + \hat{E}_{\text{source}}(r, \omega, t), \]

where we have used similar approximations as in Eq. (42). In accordance with Eqs. (11)–(13), we substitute this result into Eq. (42) and take the expectation with respect to the field state and the internal (electronic) state of the atomic system, leading to

\[ \langle \hat{F}(t) \rangle = \langle \hat{F}_{\text{free}}(t) \rangle + \langle \hat{F}_{\text{source}}(t) \rangle, \]

where we have used similar approximations as in Eq. (42). In accordance with Eqs. (11)–(13), we substitute this result into Eq. (42) and take the expectation with respect to the field state and the internal (electronic) state of the atomic system, leading to

\[ \langle \hat{F}(t) \rangle = \langle \hat{F}_{\text{free}}(t) \rangle + \langle \hat{F}_{\text{source}}(t) \rangle, \]

\[ \langle \hat{F}_{\text{source}}(t) \rangle = \langle \hat{F}_{\text{source}}^{\text{el}}(t) \rangle + \langle \hat{F}_{\text{source}}^{\text{mag}}(t) \rangle, \]

where \( \hat{F} \) and \( \hat{F}_{\text{free}} \) denote the CP force acting on an atomic system prepared in an arbitrary internal quantum state.

4.2 Weak-coupling regime

Representing the electric dipole operator in the form

\[ \hat{d}(t) = \sum_{m,n} d_{mn} \hat{A}_{mn}(t), \]

Equations (46)–(50) apply to both driven and non-driven atomic systems and to both weak and strong atom-field coupling. In particular, when the medium-assisted electromagnetic field is initially in the ground state, then \( \langle \hat{F}_{\text{free}}(t) \rangle = 0 \) is valid and Eqs. (49) and (50), respectively, just determine the electric part and the magnetic part of the time-dependent CP force acting on an atomic system prepared in an arbitrary internal quantum state.
(\hat{A}_{mn} = |m\rangle\langle n|, with |n\rangle, |m\rangle being the internal atomic energy eigenstates), we may express the dipole-dipole correlation function appearing in Eqs. (49) and (50) in terms of correlation functions of the \hat{A}_{mn} (t) as

\langle \hat{d}(t) \otimes \hat{d}(t') \rangle = \sum_{m,n,m',n'} d_{mn} \otimes d_{m'n'} \langle \hat{A}_{mn}(t)\hat{A}_{m'n'}(t') \rangle. \tag{52}

In the weak-coupling regime, the Markov approximation can be exploited, and the correlation functions \langle \hat{A}_{mn}(t)\hat{A}_{m'n'}(t') \rangle can be calculated by means of the quantum regression theorem (see, e.g., Ref. [17]). For a non-degenerate atomic system with sufficiently slow center-of-mass translation frequencies \(\omega \) (cf. Sec. 3.1), resulting in

\[ \langle \hat{d}(t) \rangle = \sum_{m,n,m',n'} d_{mn} \otimes d_{m'n'} \langle \hat{A}_{mn}(t)\hat{A}_{m'n'}(t') \rangle \]

\[ \langle \hat{d}(t) \otimes \hat{d}(t') \rangle = \sum_{m,n,m',n'} d_{mn} \otimes d_{m'n'} \langle \hat{A}_{mn}(t)\hat{A}_{m'n'}(t') \rangle. \tag{52} \]

For \(t \geq t', m \neq n\), \(\hat{r}_A = \hat{r}_A(t)\), where

\[ \tilde{\omega}_{mn}(\hat{r}_A) = \omega_{mn} + \delta \omega_m(\hat{r}_A) - \delta \omega_n(\hat{r}_A), \]

\[ \delta \omega_m(\hat{r}_A) = \sum_k \delta \omega^k_m(\hat{r}_A), \]

\[ \delta \omega^k_m(\hat{r}_A) = \frac{\mu_0}{\pi \hbar} \int_0^\infty d\omega \omega^2 \frac{d_{km} \text{Im} G^{(1)}(\hat{r}_A, \hat{r}_A, \omega) d_{mk}}{\tilde{\omega}_{mk}(\hat{r}_A) - \omega} \tag{56} \]

are the body-induced shifted transition frequencies, and

\[ \Gamma_m(\hat{r}_A) = \sum_k \Gamma^k_m(\hat{r}_A), \]

\[ \Gamma^k_m(\hat{r}_A) = \frac{2\mu_0}{\hbar} \Theta[\tilde{\omega}_{mk}(\hat{r}_A)][\tilde{\omega}_{mk}(\hat{r}_A)]^2 d_{km} \text{Im} G \langle \hat{r}_A, \hat{r}_A, \hat{r}_A, \hat{r}_A, \hat{r}_A \rangle d_{mk} \tag{58} \]

are the position-dependent level widths. Note that the position-independent (infinite) Lamb-shift terms resulting from \(G^{(0)}(\hat{r}_A, \hat{r}_A, \omega)\) [recall Eq. (28)] have been absorbed in the transition frequencies \(\omega_{mn}\). Equation (56) can be rewritten by changing to imaginary frequencies (cf. Sec. 3.1), resulting in

\[ \delta \omega^k_m(\hat{r}_A) = -\frac{\mu_0}{\hbar} \Theta[\tilde{\omega}_{mk}(\hat{r}_A)][\tilde{\omega}_{mk}(\hat{r}_A)]^2 d_{km} \text{Re} G^{(1)}(\hat{r}_A, \hat{r}_A, \tilde{\omega}_{mk}(\hat{r}_A)) d_{mk} \]

\[ + \frac{\mu_0}{\pi \hbar} \int_0^\infty du u^2 \tilde{\omega}_{km}(\hat{r}_A) \frac{d_{km} G^{(1)}(\hat{r}_A, \hat{r}_A, iu) d_{mk}}{[\tilde{\omega}_{km}(\hat{r}_A)]^2 + u^2}. \tag{59} \]

The calculation of

\[ \langle \hat{A}_{mn}(t) \rangle = \sigma_{nm}(t) \]

[\sigma_{nm}(0) = \sigma_{nm}] yields (App. A)

\[ \sigma_{nm}(t) = e^{i\tilde{\omega}_{mn}(\hat{r}_A) + \frac{\Gamma_m(\hat{r}_A) + \Gamma_n(\hat{r}_A)}{2} t} \sigma_{nm} \]

for \(m \neq n\), so the remaining task consists in solving the balance equations

\[ \dot{\sigma}_{mm}(t) = -\Gamma_m(\hat{r}_A) \sigma_{mm}(t) + \sum_n \frac{\Gamma^m_n(\hat{r}_A) \sigma_{nm}(t) + \Gamma_n^m(\hat{r}_A) \sigma_{mn}(t)}{2}. \tag{62} \]
Upon using Eqs. (52), (53), and (60), the CP force as defined according to Eqs. (48)–(72) can now be calculated by evaluating the time integrals in the spirit of the Markov approximation, resulting in

\[
\langle \hat{F}(t) \rangle = \sum_{m,n} \sigma_{mn}(t) F_{mn}(r_A), \tag{63}
\]

\[
F_{mn}(r_A) = F_{mn}^{el}(r_A) + F_{mn}^{mag}(r_A), \tag{64}
\]

with

\[
F_{mn}^{el}(r_A) = \left\{ \frac{\mu_0}{\pi} \sum_{k} \int_{0}^{\infty} d\omega \omega^2 \frac{\nabla \otimes d_{mk} \text{Im} G^{(1)}(r, \hat{r}_A, \omega)d_{kn}}{\omega - \omega_{kn}(r_A) - i[\Gamma_k(r_A) + \Gamma_m(r_A)]/2} \right\} + \text{H.c.}, \tag{65}
\]

\[
F_{mn}^{mag}(r_A) = \left\{ \frac{\mu_0}{\pi} \sum_{k} \int_{0}^{\infty} d\omega \omega \omega_{mn}(r_A) \frac{d_{mk} \times \nabla \times \text{Im} G^{(1)}(r, \hat{r}_A, \omega)d_{kn}}{\omega + \omega_{kn}(r_A) - i[\Gamma_k(r_A) + \Gamma_m(r_A)]/2} \right\} + \text{H.c.}, \tag{66}
\]

where we have again made the replacement \(G(r, \hat{r}_A, \omega) \rightarrow G^{(1)}(r, \hat{r}_A, \omega) [\text{cf. Eq. (28)}].\) As \(\hat{r}_A\) effectively enters equations (63)–(66) as a parameter, the caret will be removed in the following \(r_A \rightarrow r_A\). We finally rewrite Eqs. (65) and (66) by going over to imaginary frequencies (cf. Sec. 5.1). Introducing the abbreviating notation

\[
\Omega_{mnk}(r_A) = \hat{\omega}_{mn}(r_A) + i[\Gamma_m(r_A) + \Gamma_k(r_A)]/2 \tag{67}
\]

as well as the generalized atomic polarizability tensor

\[
\alpha_{mn}(r_A, \omega) = \frac{1}{\hbar} \sum_{k} \left[ \frac{d_{mk} \otimes d_{kn}}{\hat{\omega}_{kn}(r_A) - \omega - i[\Gamma_k(r_A) + \Gamma_m(r_A)]/2} + \frac{d_{kn} \otimes d_{mk}}{\hat{\omega}_{km}(r_A) + \omega + i[\Gamma_k(r_A) + \Gamma_m(r_A)]/2} \right], \tag{68}
\]

with \(\alpha_{l}(r_A, \omega) = \alpha_{ll}(r_A, \omega)\) being the ordinary (Kramers-Kronig-consistent) polarizability tensor of an atom in state \(|l\rangle\) (cf. Ref. [18]), we derive

\[
F_{mn}^{el}(r_A) = F_{mn}^{el,or}(r_A) + F_{mn}^{el,r}(r_A), \tag{69}
\]

\[
F_{mn}^{mag}(r_A) = F_{mn}^{mag,or}(r_A) + F_{mn}^{mag,r}(r_A), \tag{70}
\]

where

\[
F_{mn}^{el,or}(r_A) = -\left\{ \frac{\hbar \mu_0}{2\pi} \int_{0}^{\infty} du u^2 \left[ (\alpha_{mn})_{ij}(r_A, iu) + (\alpha_{mn})_{ij}(r_A, -iu) \right] \nabla G^{(1)}_{ij}(r, r_A, iu) \right\}, \tag{71}
\]

\[
F_{mn}^{el,r}(r_A) = \left\{ \mu_0 \sum_{k} \Theta(\hat{\omega}_{nk}) \Omega_{mnk}^2(r_A) \nabla \otimes d_{mk} G^{(1)}(r, r_A, \Omega_{mnk}(r_A))d_{kn} \right\} + \text{H.c.}, \tag{72}
\]

\[10\]
to the short-distance limit

\[ \mathbf{F}_{\text{mag,or}}^{m m}(\mathbf{r}_A) = \left\{ \frac{\hbar \mu_0}{2\pi} \int_0^\infty du \, u^2 \, \text{Tr} \left( \left[ \frac{\tilde{\omega}_{mn}(\mathbf{r}_A)}{iu} \alpha_{mn}^\dagger(\mathbf{r}_A, iu) \right] \times \left[ \mathbf{\nabla} \times \mathbf{G}^{(1)}(\mathbf{r}, \mathbf{r}_A, iu) \right] \right) \right\}_{\mathbf{r}=\mathbf{r}_A}, \]  

(73)

\[ \mathbf{F}_{\text{mag,or}}^{m n}(\mathbf{r}_A) = \left\{ \frac{\mu_0}{2} \sum_k \Theta(\tilde{\omega}_{mk}) \tilde{\omega}_{mn}(\mathbf{r}_A) \Omega_{mnk}(\mathbf{r}_A) \mathbf{d}_{mk} \times \left( \mathbf{\nabla} \times \mathbf{G}^{(1)}(\mathbf{r}, \mathbf{r}_A, \Omega_{mnk}(\mathbf{r}_A)) \right) \right\}_{\mathbf{r}=\mathbf{r}_A} + \text{H.c.} \]  

(74)



Equation (51) together with Eq. (53) and Eqs. (69)–(74) is the natural generalization of the perturbative result of Sec. 3.1. In particular, an atom initially prepared in an energy eigenstate \( |l\rangle \) experiences for \( \Gamma_C t \ll 1 \) (\( \Gamma_C \), characteristic atomic decay rate) the CP force \( \langle \hat{F}(t) \rangle \approx \langle \hat{F}(0) \rangle = \mathbf{F}_{\text{el,or}}^{50}(\mathbf{r}_A) = \mathbf{F}_{\text{el,or}}(\mathbf{r}_A) + \mathbf{F}_{\text{el,or}}^{\ell r}(\mathbf{r}_A) \) with

\[ \mathbf{F}_{\text{el,or}}^{50}(\mathbf{r}_A) = -\frac{\hbar \mu_0}{4\pi} \int_0^\infty du \, u^2 \left[ (\alpha_i)_{ij}(\mathbf{r}_A, iu) + (\alpha_i)_{ji}(\mathbf{r}_A, -iu) \right] \mathbf{\nabla}_A \mathbf{G}^{(1)}(\mathbf{r}, \mathbf{r}_A, iu), \]  

(75)

\[ \mathbf{F}_{\text{el,or}}^{\ell r}(\mathbf{r}_A) = \frac{\mu_0}{2} \sum_k \Theta(\tilde{\omega}_{lk}) \Omega_{lk}(\mathbf{r}_A) \left\{ \mathbf{\nabla} \otimes \mathbf{d}_{lk} \mathbf{G}^{(1)}(\mathbf{r}, \mathbf{r}, \Omega_{lk}(\mathbf{r}_A)) \mathbf{d}_{lk} \right\}_{\mathbf{r}=\mathbf{r}_A} + \text{H.c.} \]  

(76)

[\Omega_{lk}(\mathbf{r}_A) \equiv \Omega_{lk}(\mathbf{r}_A) ; \mathbf{d}_{lk} \text{ real}]. Note that due to the position dependence of the atomic polarizability even the ground-state CP-force \( \mathbf{F}_{\text{el,or}}^{50}(\mathbf{r}_A) = \mathbf{F}_{\text{el,or}}(\mathbf{r}_A) \) cannot be derived from a potential in the usual way. It is worth noting that when the atom is initially prepared in a coherent superposition of states, then the corresponding off-diagonal force components \( \sigma_{nm}(t) \mathbf{F}_{mn}(\mathbf{r}_A) \) \( (n \neq m) \) can also contribute to the total CP force as given by Eq. (53). These components contain contributions not only from the electric part of the Lorentz force but also from the magnetic part, as can be easily seen from inspection of Eqs. (76) and (74).

4.3 Excited atom in front of a magnetodielectric half-space

To illustrate the effects of body-induced level shifting and broadening, let us again consider a two-level atom in front of a magnetodielectric half-space (cf. Sec. 3.2) and calculate the CP force \( \mathbf{F}_{11}(\mathbf{z}_A) \) acting on the atom in the upper state. For simplicity, we restrict our attention to the short-distance limit \( z_A \sqrt{\varepsilon / \mu} / c \ll 1 \), where the approximations \( \beta_0 \approx i \delta, \beta \approx i \delta \) can be applied to Eq. (57). In this limit, which is governed by the dielectric properties of the material, Eq. (51) [together with Eqs. (52) and (53)] and Eq. (57) [together with Eq. (58)] approximate to

\[ \tilde{\omega}_{10}(\mathbf{z}_A) = \omega_{10} + \delta \omega_{1}(\mathbf{z}_A) - \delta \omega_{0}(\mathbf{z}_A) = \omega_{10} - \frac{d_A^2 (1 + \cos^2 \theta)}{32 \pi \varepsilon_0 \hbar z_A^3} \frac{\left| \varepsilon \tilde{\omega}_{10}(\mathbf{z}_A) \right| ^2 - 1}{\left| \varepsilon \tilde{\omega}_{10}(\mathbf{z}_A) \right| + 1}, \]  

(77)

\[ \Gamma_{1}(\mathbf{z}_A) = \frac{d_A^2 (1 + \cos^2 \theta)}{8 \pi \varepsilon_0 \hbar z_A^3} \frac{\text{Im} \varepsilon \tilde{\omega}_{10}(\mathbf{z}_A)}{\left| \varepsilon \tilde{\omega}_{10}(\mathbf{z}_A) \right| + 1}, \]  

(78)

where we have neglected the off-resonant second term in Eq. (59) as well as the small free-space decay rate. Taking into account that \( \mathbf{F}_{11}(\mathbf{z}_A) = \mathbf{F}_{\text{el,or}}^{50}(\mathbf{z}_A) \approx \mathbf{F}_{\text{el,or}}^{\ell r}(\mathbf{z}_A) \) is valid, from
Eq. (76) [together with Eq. (37) (in the short-distance limit) and Eqs. (77) and (78)] we finally obtain

$$F_{11}(z_A) = F_{11}(z_A)e_z = -\frac{3d_A^2(1 + \cos^2 \theta)}{32\pi\varepsilon_0 z_A^4} \frac{|\varepsilon[\Omega_{10}(z_A)]|^2}{|\varepsilon[\Omega_{10}(z_A)]| + 1} e_z,$$

(79)

where, according to Eq. (67), $\Omega_{10}(z_A) = \tilde{\omega}_{10}(z_A) + i\Gamma_1(z_A)/2$.

Figure 2: The resonant part of the CP force $F_{11}(z_A)$ as a function of the bare transition frequency $\omega_{10}$. It is seen that the typical dispersion profile of the attractive/repulsive CP force around the (surface-plasmon induced) resonance frequency $\omega_S = \sqrt{\omega_{T,e}^2 + \omega_{P,e}^2}/2$, which is already known from perturbation theory, is noticeably reduced by the decay-induced damping while retaining its width, because the competing effects of decay-induced broadening and narrowing due to the frequency shifts almost cancel.

5 Summary

Within the frame of electromagnetic-field quantization in the presence of dispersing and absorbing linear media, we have presented a consistent theory for calculating the CP force
experienced by an atomic system in the presence of an arbitrary arrangement of magnetodielectric bodies. Extending previous results \[6\] to the case of magnetodielectric media, we have derived the vdW energy of an atomic system prepared in an energy eigenstate using perturbative methods and demonstrated that magnetic properties of the media can give rise to interesting effects such as the formation of a potential barrier for ground-state atoms above a magnetodielectric half-space. In an alternative approach based on the Lorentz force that governs the atomic center-of mass motion, we have derived an expression for the CP force that applies to arbitrary atomic states and both strong and weak atom-field coupling. Restricting our attention to a non-driven atom in the weak-coupling regime, we have found that the CP force can be written as a superposition of force components weighted by the time-dependent intra-atomic density matrix elements that solve the intra-atomic master equation. Each force component can be written in terms of the Green tensor for the electromagnetic field and appropriate atomic quantities such as the polarizability. In contrast to the perturbative result, the atomic quantities exhibit body-induced shifts and broadenings of the atomic transition lines which can noticeably influence the CP force when the atomic system comes close to a body.

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A Intra-atomic equations of motion

Upon using the Hamiltonian (1) together with Eqs. (6), (8), and (21), and exploiting the decomposition (51) the following equations of motion can be derived:

\[
\dot{\hat{A}}_{mn} = \frac{i}{\hbar} [\hat{H}, \hat{A}_{mn}] = \frac{i}{\hbar} \sum_k \left[ (d_{nk} \hat{A}_{mk} - d_{km} \hat{A}_{kn}) \int_0^\infty d\omega \hat{E}(\hat{r}_A, \omega) \right]
\]

\[+ \frac{1}{\hbar} \int_0^\infty d\omega \hat{E}_s^\dagger(\hat{r}_A, \omega) (d_{nk} \hat{A}_{mk} - d_{km} \hat{A}_{kn}) \right],
\]

(80)

where a similar approximation as in Eq. (42) has been made. For weak atom-field coupling and sufficiently slow center-of-mass motion, the source-field dynamics as given by Eqs. (45) can be approximated by carrying out the time integral in the Markov approximation, resulting in

\[\int_0^\infty d\omega \hat{E}_{source}(\hat{r}_A, \omega) = \sum_{m,n} g_{mn}(\hat{r}_A) \hat{A}_{mn},\]

(81)

\[g_{mn}(\hat{r}_A) = \frac{i \mu_0}{\pi} \int_0^\infty d\omega \omega^2 \text{Im} G(\hat{r}_A, \hat{r}_A, \omega) d_{mn} \zeta(\tilde{\omega}_{nm}(\hat{r}_A) - \omega)\]

(82)

\[\zeta(x) = \pi \delta(x) + iP/x,\]

with \(\tilde{\omega}_{nm}(\hat{r}_A)\) according to Eq. (54).

We substitute Eqs. (43), (44), and (81) into Eq. (80) and take expectation values with respect to the internal atomic state and the vacuum field state, noting that in this case the contributions from the free-field part (44) vanish. Upon applying the decomposition

\[\frac{i}{\hbar} d_{nk} g_{kn}(\hat{r}_A) = -i \delta \omega_n^k(\hat{r}_A) - \frac{1}{2} \Gamma_n^k(\hat{r}_A),\]

(83)
and using the fact that for a non-degenerate atom off-diagonal density matrix elements decouple from each other as well as from the diagonal ones, one can derive Eqs. (53), (61), and (62).

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