Enhanced van der Waals interaction at interfaces

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Received 19 October 2007, in final form 22 January 2008
Published 9 April 2008
Online at stacks.iop.org/JPhysA/41/164020

Abstract
Using a recently obtained (general) formula for the interaction energy between an excited and a ground-state atom (Sherkunov 2007 Phys. Rev. A 75 012705), we consider the interaction energy between two such atoms near the interface between two media. We demonstrate that under the circumstances of the resonant coupling of the excited atom to the surface polariton mode of a vacuum-medium system the nonretarded atom*-atom interaction energy can be enhanced by (several) orders of magnitude in comparison with the van der Waals interaction energy of the two isolated atoms.

PACS numbers: 12.20.–m, 34.20.Cf, 34.50.Dy, 42.50.Nn

1. Introduction
It has been known for quite some time that the decay rate and energy of an excited atom (atom*) may be strongly modified near a dispersive and absorbing surface owing to the resonant coupling of the atom to the surface polariton mode(s) of the system [1–7]. Excitation of surface polaritons by decaying atom (or molecule) and their subsequent conversion into the radiation (photons) by some means is in the core of surface enhanced optical processes such as fluorescence and Raman scattering [8, 9]. Similarly, owing to the resonant energy shift of the atom, a strong modification of the related atom*-surface van der Waals force leading even to the atom-surface repulsion has been predicted [1, 3, 4, 6, 7] and observed [10]. In this work we demonstrate yet another spectacular consequence of the resonant atom*-surface coupling, namely the possibility of a strong enhancement of the van der Waals interaction between an excited and a ground-state atom in vicinity of an interface that supports surface modes.

The atom*-atom interaction has so far been considered only for atoms in free-space [11–16] and lately for atoms embedded in an absorbing medium [17, 18]. A straightforward way to study this interaction in an inhomogeneous system would therefore be to use a macroscopic QED approach appropriate for absorbing systems and derive the interaction potential between the atoms, for example, along the same lines as it was recently done for two ground-state atoms [19]. However, instead of developing the theory from the beginning,
in this work we adopt the Sherkunov formula for the atom–atom interaction potential in an absorbing medium [18]. Indeed, this formula is given in terms of the corresponding Green function and there is nothing in its derivation which crucially depends on the specific system considered in the paper. Accordingly, with the appropriate Green function, it can be applied to an inhomogeneous medium as well.

2. Theory

Consider an excited (e) atom A and a ground-state (g) atom B embedded in an inhomogeneous magnetoelectric system described by the permittivity $\varepsilon(\mathbf{r}, \omega)$ and permeability $\mu(\mathbf{r}, \omega)$. Assuming, for simplicity, two-level isotropic atoms, the vacuum force on the atom A can then be obtained from the potential

$$U_A(\mathbf{r}_A, \mathbf{r}_B) = U_A(\mathbf{r}_A) + U_{AB}(\mathbf{r}_A, \mathbf{r}_B),$$

where

$$U_A(\mathbf{r}_A) = \frac{\hbar}{2\pi c^2} \int_0^\infty d\xi \frac{\xi^2}{\omega_A^3} \frac{\alpha^*_A(\xi)}{\alpha^*_A(\xi)} \text{Tr}[\mathbf{G}^{ie}(\mathbf{r}_A, \mathbf{r}_A; i\xi)] = -\frac{|d_{eg}^A|^2}{3c^2} \text{Re} \text{Tr}[\mathbf{G}^{ie}(\mathbf{r}_A, \mathbf{r}_A; \omega_A)]$$

is the Casimir–Polder potential due to the inhomogeneity of the system [3, 6, 7] and

$$U_{AB}(\mathbf{r}_A, \mathbf{r}_B) = -\frac{\hbar}{2\pi c^2} \int_0^\infty d\xi \frac{\xi^4}{\omega_A^2 \omega_B^2} \frac{\alpha^{*}_A(\xi)}{\alpha^{*}_A(\xi)} \alpha^{*}_B(\xi) \text{Tr}[\mathbf{G}(\mathbf{r}_A, \mathbf{r}_B; i\xi) \cdot \mathbf{G}(\mathbf{r}_B, \mathbf{r}_A; i\xi)]$$

$$-\frac{\text{Re}[\alpha^g_A(\omega_A)]}{3c^4} |d_{eg}^A|^2 \text{Tr}[\mathbf{G}(\mathbf{r}_A, \mathbf{r}_B; \omega_A) \cdot \mathbf{G}^*(\mathbf{r}_B, \mathbf{r}_A; \omega_A)]$$

is the van der Waals (interaction) potential between the atoms A and B [Reference [18], equation (68)]. Here

$$\alpha^{X}_{e(g)}(\omega) = (-\epsilon)^{\frac{3}{2}} \frac{|d^X_{eg}|^2}{3\omega X} \frac{\omega^2}{\omega^2 - \omega^2_i - i\omega \gamma_X}, \quad X = A \text{ or } B,$$

are the atomic polarizabilities, $\omega_X = (E_X^g - E_X^e)/\hbar$ and $d^X_{eg} = \langle e|d^X|g\rangle$ are the transition frequency and the dipole matrix element of atom X, respectively, whereas $\gamma_X$ is the width of its excited state $|\gamma_A| = 0^*$ in (2) and (3) owing to the approaches adopted when deriving these results. The dyadic

$$\mathbf{G}(\mathbf{r}, \mathbf{r}'; \omega; \mathbf{r}; \omega) = \mathbf{G}^0(\mathbf{r}, \mathbf{r}'; \omega) + \mathbf{G}^{ie}(\mathbf{r}, \mathbf{r}'; \omega),$$

with $\mathbf{G}^0(\mathbf{r}, \mathbf{r}'; \omega)$ being the Green function in a homogeneous medium, is the classical Green function for the system satisfying $\mathbf{I} = \mathbf{\hat{x}}x + \mathbf{\hat{y}}y + \mathbf{\hat{z}}z$

$$\left[ \nabla \times \frac{1}{\mu(\mathbf{r}, \omega)} \nabla \times -\varepsilon(\mathbf{r}, \omega) \frac{\omega^2}{c^2} \mathbf{I} \right] \mathbf{G}(\mathbf{r}; \mathbf{r}') = 4\pi \mathbf{\delta}(\mathbf{r} - \mathbf{r}'),$$

with the outgoing wave condition at infinity.

All information about the mode structure of the system are contained in the scattering part of the Green function $\mathbf{G}^{ie}(\mathbf{r}, \mathbf{r}'; \omega)$. Specially, poles of this Green function in the complex $\omega$-plane correspond to frequencies of the system resonant (polariton) modes. Accordingly, whenever $\omega_A$ is close to the frequency $\omega_r$ of a resonant mode $U_A(\mathbf{r}_A, \mathbf{r}_B)$ is strongly modified owing to the second (resonant) terms in (2) and (3). Since these terms are absent if the atom A is in the ground state this effect appears only for excited atoms. Evidently, when $\omega_A \sim \omega_r$, the Casimir–Polder potential $U_A(\mathbf{r}_A)$ exhibits a dispersion due to the atom coupling to the
Two atoms near an interface shown schematically (atom B' is the mirror image of the atom B). Media are described by (complex) refraction indexes $n(\omega) = \sqrt{\varepsilon(\omega)\mu(\omega)}$ and $n_m(\omega) = \sqrt{\varepsilon_m(\omega)\mu_m(\omega)}$.

resonant mode. Simultaneously, in addition of showing the intrinsic (genuine) dispersive type resonance for $\omega_A \sim \omega_B$ governed by $\text{Re}\left[\alpha_B(\omega_A)\right]$, the van der Waals potential is resonantly enhanced when $\omega_A \sim \omega_r$ owing to the factor $|G_{ij}(r_A, r_B; \omega_A)|^2$ in (3). From this factor, we may infer that the resonant enhancement of $U_{AB}(r_A, r_B)$ with respect to its free-space value is due to the exchange of (real) system excitations between the atoms (instead of ordinary photons, as in the free space [14, 15]) and the associated field-intensity enhancement at atomic sites [8, 9].

To illustrate the above considerations and estimate the enhancement of the atom–atom van der Waals interaction, we assume that the atoms A and B are embedded in a system consisting of two semi-infinite media, as depicted in figure 1. To account for the local-field effects, we adopt the Onsager model and therefore also assume small empty spherical cavities around the atoms. Provided that $\omega_{\text{max}} R_X/c \ll 1$, with $R_X$ being cavity radii and $\omega_{\text{max}}$ an effective cutoff frequency in (2) and (3), the Green function for this system can (to the order of $\omega R_X/c$) be written as [20]

$$G^\infty(r_A, r_A; \omega) = \frac{i2\omega}{3c} C(\omega) I + L(\omega) G^\infty(r_A, r_A; \omega),$$

$$G(r_A, r_B; \omega) = L(\omega) G(r_A, r_B; \omega), \quad L(\omega) = \left[\frac{3\varepsilon(\omega)}{2\varepsilon(\omega) + 1}\right]^2.$$ (7a)

Here $C(\omega)$ (given explicitly in [20]) is the reflection coefficient for the field scattered within the cavity and $G(r_A, r_B; \omega)$ is the Green function for the system unperturbed by the Onsager cavities. In order to keep the discussion simple, in this work we consider the situation where the atomic distances from the interface between the media are small compared to $c/\omega_{\text{max}}$ (but larger than $R_X$) so that the retardation of the electromagnetic field can be neglected. The Green function for this system in the nonretarded approximation $G_{\text{nr}}(r_A, r_B; \omega)$ can be found by adding the nonretarded (quasistatic) field $E_{\text{nr}}(r_A, r_B; \omega)$ of an oscillating dipole $d$ at $r_B$ and the corresponding field $E'_{\text{nr}}(r_A, r'_B; \omega)$ of its image

$$d' = r(\omega)(-I_y + \hat{z}z) \cdot d, \quad r(\omega) = \frac{\varepsilon_m(\omega) - \varepsilon(\omega)}{\varepsilon_m(\omega) + \varepsilon(\omega)},$$ (8)

at $r'_B = r_B| - \varepsilon_B \hat{z}$. Here $I_y = \hat{x}x + \hat{y}y$ and $r(\omega)$ is the Fresnel reflection coefficient for the interface in the quasistatic limit. Thus, using [21]

$$E_{\text{tot}}(r_A, r_B; \omega) = \frac{\omega^2}{c^2} G_{\text{nr}}(r_A, r_B; \omega) \cdot d$$ (9)
and equation (7b), we have for the Green function
\[
G_{\mu
u}(\mathbf{r}_A, \mathbf{r}_B; \omega) = \frac{e^2}{\varepsilon(\omega)\omega^2} \left[ \frac{3 \mathbf{R} \cdot \mathbf{R} - \mathbf{R}_A^2}{R^5} + r(\omega) \frac{3 \mathbf{R}' \cdot \mathbf{R}' - \mathbf{R}_B^2}{R^5} \right],
\]
where \( \mathbf{R} = \mathbf{r}_A - \mathbf{r}_B = \mathbf{R}_A + \mathbf{Z} \mathbf{z} \) and \( \mathbf{R}' = \mathbf{r}_A - \mathbf{r}_B' = \mathbf{R}_A + \mathbf{Z}_+ \mathbf{z} \), with \( \mathbf{Z} = \mathbf{z}_A - \mathbf{z}_B \) and \( \mathbf{Z}_+ = \mathbf{z}_A + \mathbf{z}_B \).

Combining the nonretarded limit [21] of (5) and (7a) with (10), we find that
\[
\text{Tr} \left[ G_{\mu
u}^\infty(\mathbf{r}_A, \mathbf{r}_A; \omega) \right] = \frac{6e^2}{\omega^2 R_A} \frac{\varepsilon(\omega) - 1}{2\varepsilon(\omega) + 1} + \frac{2e^2}{\omega^2 R_A^2} \frac{L(\omega)}{\varepsilon(\omega)}.
\]

In conjunction with (2), this leads to (we omit the position-independent part of the potential due to the nearby medium)
\[
U_A(\mathbf{r}_A) = -\frac{\hbar}{4\pi\varepsilon_A^3} \int_0^\infty d\xi \alpha_s^A(\xi) \frac{L(\xi)}{\varepsilon(\xi)} \frac{L(\xi)}{\varepsilon(\xi)} W(R_{ij}, Z, Z_+; \xi)
- \frac{2|\mathbf{d}_{eg}|^2 |L(\omega_A)|^2}{3|\varepsilon(\omega_A)|^2} \text{Re}[\alpha_s^B(\omega_A)] W(R_{ij}, Z, Z_+; \omega_A),
\]
\[
W(R_{ij}, Z, Z_+; \omega) = \frac{3}{R^6} + |r(\omega)|^2 \frac{3}{R^6} - \text{Re}[r(\omega)] \frac{3(R_1^4 - Z^2 Z_+^2) + R^2 R^2}{R^3 R^5}.
\]

Here, the first two terms come from the direct interaction between the atoms and the interaction of atom A with the image of atom B, respectively, whereas the third one is an interference term.

According to (4), the off-resonant (first) term in (13) is of the same form as the van der Waals potential between two ground-state atoms. Therefore, referring the reader to [19] for a detailed discussion of this term, here we pay attention only to the (usually much larger) resonant part of the atom–atom potential \( U_{AB}^{\text{res}} \). Using (4), we rewrite \( U_{AB}^{\text{res}} \) as
\[
U_{AB}^{\text{res}}(\mathbf{r}_A, \mathbf{r}_B) = -\frac{2|\mathbf{d}_{eg}|^2 |L(\omega_A)|^2}{|\varepsilon(\omega_A)|^2} \frac{\omega_B^2(\omega_B - \omega_A)}{R^6} \frac{\omega_A^2 - \omega_A^2}{\omega_B^2 - \omega_A^2} \left[ 1 + \frac{3|\varepsilon(\omega_A)|^2}{R^6} - \frac{3}{R} \frac{3(R_1^4 - Z^2 Z_+^2) + R^2 R^2}{R^3 R^5} \right].
\]

Evidently, the last factor here describes the modification of the resonant atom–atom potential in an infinite medium owing to the presence of the nearby interface. As seen, it is highly anisotropic and depends not only on the distance of the molecules from the interface but also on their mutual orientation with respect to it.

3. Discussion

In order to keep the discussion simple, we assume that the atoms are embedded in the vacuum \( \varepsilon(\omega) = 1 \) in front of a dielectric medium that is around a resonant at \( \omega_T \) described by the
Relative interaction potential for parallel (solid line) and perpendicular (dashed line) orientation of the atoms with respect to the surface at the distance $z_A = 0.1R$ from the atom A as a function of the transition frequency of atom A. Relevant medium parameters are $\eta = 2.71, \epsilon(0) = 6.57, \Gamma = 0.015\omega_S$ and $\omega_S = 1.54 \times 10^{14}\text{s}^{-1}$ [7]. The transition frequency and linewidth of atom B are $\omega_B = 0.9\omega_S$ and $\gamma_B = 10^{-3}\omega_S$, respectively.

dielectric function [5]

$$\varepsilon_m(\omega) = \eta \left(1 + \frac{\omega_p^2}{\omega^2 - \omega^2 - i\omega\Gamma}\right),$$

where $\eta$ is a background dielectric constant, $\eta\omega_p^2$ is (essentially) the coupling between the medium polarization and the field and $\Gamma$ is the corresponding damping constant. We note that this dielectric function is physically unacceptable generally since it does not tend to unity for large frequencies. However, it can safely be employed as an effective permittivity in a (finite) frequency interval around $\omega_T$. The reflection coefficient to be used in (14) therefore reads

$$r(\omega) = \frac{\eta - 1}{\eta + 1} + \frac{\sigma^2}{\omega_S^2 - \omega^2 - i\omega\Gamma}, \quad \sigma^2 = \frac{\varepsilon(0) - 1}{\varepsilon(0) + 1} = \frac{\eta - 1}{\eta + 1},$$

where $\omega_S = \sqrt{\eta\omega_p^2/(\eta + 1) + \omega^2}$ is the surface-mode frequency. Equations (14) and (16) explicitly demonstrate the intrinsic dispersion of $U_{AB}$ when $\omega_A \sim \omega_B$ and enhancement when $\omega_A \sim \omega_S$. Note that for $\omega_B > \omega_A \sim \omega_S$ ($\omega_B < \omega_A \sim \omega_S$) this surface enhanced potential is attractive (repulsive). It is also immediately seen that when $\omega_A = \omega_S$ the potential $U_{AB}^r$ is enhanced with respect to its free-space value by a factor of

$$g(R_1, z_A, z_B; \omega_S) \simeq \frac{\sigma^4\omega_S^2}{\Gamma^2} \left(1 + \frac{4z_A z_B}{R^2}\right)^{-3}.$$

Since for insulators typically $\Gamma/\omega_S \sim 10^{-2}$ and for (noble) metals ($\eta = 1$ and $\omega_T = 0$) typically $\Gamma/\omega_S \sim 10^{-3}$, this implies that under the circumstances of resonant coupling of atom A to the surface polariton mode at nearby surface $U_{AB}^r$ could be enhanced by several orders of magnitudes.

We illustrate the above considerations in figure 2 where we have plotted $U_{AB}^r$ in units $U_{AB}^0 R = 2|\alpha^A|^2|\alpha^B(0)|/R^6$ as a function of $\omega_A$ for parallel and perpendicular orientation of the atoms with respect to the surface. For these orientations of the atoms $U_{AB}^r$ is a (monotonically decreasing) function of $z_A/R$ only and displayed curves correspond to $z_A = 0.1R$. Medium
parameters are chosen from [7] and correspond to sapphire around the surface polariton resonance at $\lambda_S = 12.21 \, \mu m$ whereas parameters of the atom B are chosen quite arbitrarily. We see that, besides a resonant structure at $\omega_B$ as would exist in the free-space, the potential exhibits also a resonance at the surface mode frequency $\omega_S$ implying a strongly surface enhanced interaction between the atoms. For example, we find that $U_{AB}$ in the parallel geometry is at the surface mode resonance 298.5 times larger than in the free space. We note that this is in a very good agreement with $g(R_{\parallel}, z_A, z_A; \omega_S)$ calculated from (17) for $z_A = 0.1 R$.

Of course, for $\omega_B > \omega_S \sim \omega_A$, the surface enhanced potential is attractive, as illustrated in figure 3 where we have plotted $U_{AB}'$ in the parallel geometry for two symmetric values of $\omega_B$ with respect to $\omega_S$. The largest enhancement is, however, obtained when $\omega_A \sim \omega_B \sim \omega_S$. This ‘double resonance’ case is illustrated in figure 4, where we have plotted $U_{AB}'$ in the parallel (solid line) and perpendicular (dashed line) geometry for $\omega_A$ around $\omega_B = \omega_S$. We see that just below (above) the double resonance the attractive (repulsive) part of the potential is additionally considerably enhanced (note the change of scale on the ordinate axes).
Figure 5. Relative perpendicular force on atom A for parallel orientation of atoms A and B with respect to the sapphire surface. The distance of atom A from the surface is $z_A = 3[\alpha_B^4(0)]^{1/3}$ and the distance between the atoms is $R = z_A$ (solid line) and $R = 5z_A$ (dashed line). Other parameters are the same as in figure 2.

We end this discussion by a short comment on the force on the atom A

$$ F_A(\mathbf{r}_A, \mathbf{r}_B) = -\nabla_A U_A(\mathbf{r}_A, \mathbf{r}_B) $$

(18)

around the surface mode resonance. Keeping only the (most) resonant terms in (12) and (13), we find for $F_A$ [$\varepsilon(\omega) = 1$]

$$ F_A(\mathbf{r}_A, \mathbf{r}_B) \simeq -2|d_{eg}^A|^2 - R \frac{12|d_{eg}^A|^2}{R^8} \text{Re}[e^A(\omega_A)]\varepsilon(\omega_A)^2, $$

(19)

where $\varepsilon(\omega)$ is given by the resonant term in (16). Introducing the function

$$ L(x, y, z) = x^4[(x^2 - y^2)^2 + (yz)^2]^{-1}, $$

(20)

we rewrite $F_A$ at $\omega_A \simeq \omega_S$ in components as

$$ F_{A\parallel}(\mathbf{r}_A, \mathbf{r}_B) \simeq -R \frac{12|d_{eg}^A|^2 \alpha_B^4(0)}{(R^2 + 4z_Az_B)^2} \left(1 - \frac{\omega_A^2}{\omega_B^2}\right) L(\omega_B, \omega_A, \gamma_B) \sigma^4 L(\omega_S, \omega_A, \gamma), $$

(21a)

$$ F_{A\perp}(\mathbf{r}_A, \mathbf{r}_B) \simeq -2|d_{eg}^A|^2 - \frac{2z_A^4(z_A + z_B)\alpha_B^4(0)}{(R^2 + 4z_Az_B)^2} \left(1 - \frac{\omega_A^2}{\omega_B^2}\right) L(\omega_B, \omega_A, \gamma_B) \sigma^2 \right]. $$

(21b)

As seen, because of the presence of the atom B, $F_A$ acquires a parallel component and its perpendicular component is diminished or enlarged owing to the relative positions of the atomic and surface resonances.

The above considerations are illustrated in figure 5 where we have plotted $F_{A\parallel}(\omega_A)$ in units $F_0 = |d_{eg}^A|^2 / 2z_A^4$ for parallel orientation of atoms A and B with respect to the surface and for two values of the parameter $R/z_A$. The distance of atom A from the surface is fixed letting $z_A = 3[\alpha_B^4(0)]^{1/3}$; for $\alpha_B^4(0)$ in the range of polarizabilities of the alkali-metal atoms [≃ (2−6) $\times$ 10$^{-23}$ cm$^3$ [22]], for example, this corresponds to $z_A \sim 1$ nm. As follows from (21b), under these circumstances the atom-atom component of the force decreases rapidly.
with the relative distance between the atoms \( R/z_A \) so that the dashed line practically coincides with the purely Casimir–Polder (atom-surface) force given by the first term in (21b). When the atoms are closer (solid line), owing to its strong repulsion from the atom B for \( \omega_A \sim \omega_S \), the force on atom A is diminished/enhanced in the attractive \( (\omega_A < \omega_S) \)/repulsive \( (\omega_A > \omega_S) \) atom-surface force region. Of course, for \( \omega_B > \omega_S \) the situation is reversed: due to the attraction between the atoms around the surface resonance \( F_{Az}(\omega_A) \) is enhanced/diminished in the attractive/repulsive atom-surface force region. The same effect is also observed for the perpendicular orientation of atoms with respect to the surface, however, owing to the attenuation of the surface-mode field away from the surface, in this case it is much weaker. Evidently, this effect is reinforced when the atoms are closer to the surface and to each other and disappears for large \( z_A \) and/or \( R \).

4. Conclusions

In conclusion, we have demonstrated that the (generalized) Sherkunov formula implies a strong enhancement of the van der Waals atom–atom interaction near a vacuum-medium interface under the circumstances of the resonant coupling of the excited atom with the surface mode of the system. The enhancement is due to the exchange of (real) surface excitations between the atoms (instead of photons) and the accompanying enhancement of the electromagnetic field intensity at atomic sites. This promotes the van der Waals interaction as another example of the surface enhanced phenomena.

Acknowledgments

The author thanks Yuri Sherkunov for discussions and useful comments. This work was supported by the Ministry of Science, Education and Sport of the Republic of Croatia under contract No. 098-1191458-2870.

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