Surface enhanced van der Waals force

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Abstract

We consider the van der Waals interaction of an excited atom and a ground-state atom across a vacuum–medium interface under the circumstances of the resonant coupling of the excited atom to a surface polariton mode of the system. We demonstrate that the nonretarded interaction potential between the atoms is, in this case, the same as in an effective medium described by the average dielectric function of the media in contact, except for the different local-field correction factor. The estimate performed for the vacuum–sapphire interface shows that the nonretarded van der Waals (atom–atom) force can be at the surface-mode resonance enhanced by almost three orders of magnitude in comparison to its free-space value. Owing to the local-field effect in the present configuration, this enhancement factor is larger than previously estimated for atoms in front of the same medium by (almost) an order of magnitude.

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1. Introduction

It is well known that the decay rate and energy of an excited atom (atom*) may be significantly affected near a dispersive surface due to the resonant coupling of the atom to the surface polariton mode(s) of the system [1–8]. Owing to the resonant energy shift of the atom, a strong modification of the related atom*–surface force leading even to the atom–surface repulsion has been predicted [1, 4], [6–8] and observed [9]. Based on the Sherkunov formula for the interaction energy between an excited and a ground-state atom [10], we have recently pointed out that the atom*–atom van der Waals potential/force is also strongly enhanced near a dispersive surface whenever the transition frequency of the excited atom coincides with the frequency of a resonant mode of the system [11].

Indeed, consider an excited (e) atom A and a ground-state (g) atom B embedded in an inhomogeneous magnetoelectric system described by the permittivity $\varepsilon(r, \omega)$ and permeability $\mu(r, \omega)$. Assuming, for simplicity, two-level isotropic atoms, the interaction potential between the atoms is, according to [10] [equation (68)], given by

$$U_{AB}^r(r_A, r_B) = \frac{\hbar^3}{c^2} \int_0^{\infty} \frac{d\xi e^{i\xi r}}{\pi^2} \hat{\alpha}_e^A(i\xi) \hat{\alpha}_g^B(i\xi) \times \text{Tr} \left[ \mathbf{G}(r_A, r_B; i\xi) \cdot \mathbf{G}(r_B, r_A; i\xi) \right],$$

where $U_{AB}^r$ and $U_{AB}^o$ are off-resonant and resonant parts of the potential, respectively. In equation (1), $\hat{\alpha}_X^Y(\omega)$ ($X = A, B$) are the atomic polarizabilities whereas $\omega_A = (E_A - E_{gA})/\hbar$ and $d_{eg} = (eA|d^4|eB)$ are, respectively, the transition frequency and the dipole matrix element of atom A. The dyadic $\mathbf{G}(r, r'; \omega)$ is the classical Green’s function for the system satisfying $[\mathbf{I}]$ is unit dyadic

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

with the outgoing wave condition at infinity. Poles of this Green’s function in the complex $\omega$-plane correspond to frequencies of the system resonant (polariton) modes. Accordingly, whenever $\omega_A$ is close to the frequency $\omega_t$ of a resonant mode the Green’s function behaves as

$$|G_{ij}(r_A, r_B; \omega_A)| \sim (\omega_A - \omega_t + i\gamma/2)^{-1}$$

and, owing to the factor $|G_{ij}|^2$, the van der Waals potential is resonantly enhanced with respect to its free-space value.

In order to verify the above considerations and estimate the enhancement of the atom*–atom van der Waals interaction, in [11] we have considered the interaction between the atoms in front of the interface between two...
media under the circumstances of the resonant coupling of the excited atom to the surface polariton mode of the system. Here, we complement this work by considering the atom–atom interaction across the interface between two media, as depicted in figure 1.

2. Theory

To account for the local-field effects, we adopt the Onsager model and therefore assume small empty spherical cavities around the atoms. Provided that \( \omega_{\text{max}}R_{A(B)}/c \ll 1 \), with \( R_{A(B)} \) being cavity radii and \( \omega_{\text{max}} > \omega_{A(B)} \) an effective cutoff frequency in (1), the Green’s function for this system can to the order of \( \omega R_{A(B)}/c \) be written as [12]

\[
\mathbf{G}(\mathbf{r}_A, \mathbf{r}_B; \omega) = \mathbf{D}(\omega)\tilde{\mathbf{G}}(\mathbf{r}_A, \mathbf{r}_B; \omega)\mathbf{D}_m(\omega),
\]

where

\[
\mathbf{D}(\omega) = \frac{3\varepsilon(\omega)}{2\varepsilon(\omega) + 1},
\]

\[
\mathbf{D}_m(\omega) = \frac{3\varepsilon_m(\omega)}{2\varepsilon_m(\omega) + 1},
\]

and \( (\mathbf{R}_1 = \mathbf{r}_A - \mathbf{r}_B) \)

\[
\tilde{\mathbf{G}}(\mathbf{r}_A, \mathbf{r}_B; \omega) = \int \frac{d^2k}{(2\pi)^2} e^{i\mathbf{k}\cdot\mathbf{R}} \tilde{\mathbf{G}}(\mathbf{z}_A, \mathbf{z}_B; \omega, \mathbf{k}),
\]

\[
\tilde{\mathbf{G}}(\mathbf{z}_A, \mathbf{z}_B; \omega, \mathbf{k}) = 2\pi i \frac{\mu}{\beta} \left[ t_\perp \frac{\hat{\mathbf{k}} - \mathbf{z}}{n_0/c} \frac{\hat{\mathbf{k}} - \mathbf{z}}{n_m/c} + t_\parallel \mathbf{k} \times \mathbf{z} \right] e^{i\beta_\perp \mathbf{z} \cdot \mathbf{z}}
\]

is the Green’s function for the system unperturbed by the Onsager cavities [13]. Here \( \beta = \sqrt{(n_0/c)^2 - k^2} \) and \( \beta_m = \sqrt{(n_m/c)^2 - k^2} \) are the perpendicular wave vectors in the media whereas

\[
t_\parallel(k, \omega) = \frac{\varepsilon \mu_m}{\varepsilon \mu + \varepsilon \mu_m} \frac{2\varepsilon \beta_m}{\varepsilon \beta_m + \varepsilon \beta_m} \text{ and } t_\perp(k, \omega) = \frac{\mu \beta_m}{\mu \beta + \mu \beta_m}
\]

are the Fresnel transmission coefficients for the interface. Considering the situation where the atomic distances from the interface between the media are small compared to \( c/\omega_{\text{max}} \) (but larger than \( R_{A(B)} \)), we can neglect the retardation of the electromagnetic field. The Green’s function in the nonretarded approximation \( \tilde{\mathbf{G}}_{nm}(\mathbf{r}_A, \mathbf{r}_B; \omega) \) is easily obtained using the prescription [13]

\[
\tilde{\mathbf{G}}_{nm}(\mathbf{r}_A, \mathbf{r}_B; \omega) = \frac{c^2}{\varepsilon(\omega)\omega^2} \lim_{c^{-2} \to \infty} \tilde{\mathbf{G}}(\mathbf{r}_A, \mathbf{r}_B; \omega)
\]

and we find

\[
\tilde{\mathbf{G}}_{nm}(\mathbf{r}_A, \mathbf{r}_B; \omega) = \frac{c^2}{\varepsilon(\omega)\omega^2} \int \frac{d^2k}{2\pi} (i\hat{k} - \mathbf{z})(i\hat{k} - \mathbf{z})
\]

\[
\times e^{i\mathbf{k}\cdot(\mathbf{r}_A - \mathbf{r}_B)},
\]

(7)

where

\[
\varepsilon(\omega) = \frac{\varepsilon(\omega) + \varepsilon_m(\omega)}{2}
\]

is the average dielectric function of the two media in contact. As seen, this Green’s function is the same as the nonretarded Green’s function in an infinite medium described by the dielectric function \( \varepsilon(\omega) \). Accordingly, going back to the \((\mathbf{R}, \omega)\)-space recalling that

\[
\frac{1}{\mathbf{R}} = \int \frac{d^2k}{2\pi} e^{i\mathbf{k}\cdot(\mathbf{r}_A - \mathbf{r}_B)}, \quad \mathbf{R} = \mathbf{r}_A - \mathbf{r}_B,
\]

(9)

we finally arrive at

\[
\mathbf{G}_{nm}(\mathbf{r}_A, \mathbf{r}_B; \omega) = \frac{\mathbf{D}(\omega)\mathbf{D}_m(\omega)}{(c^2/\omega^2\varepsilon(\omega))^2} \mathbf{R} + \mathbf{R}^3.
\]

(10)

Inserting the above Green’s function in equation (1) and neglecting the (small) off-resonant part of the potential, we find that \( U_{AB} \) is given by

\[
U_{AB}(\mathbf{r}_A, \mathbf{r}_B) = -\frac{2Re[\sigma_{g}^2(\omega_A)]|\mathbf{d}_{g}|^2|\mathbf{D}(\omega_A)|^2|\mathbf{D}_m(\omega_A)|^2}{|\varepsilon(\omega_A)|^2\mathbf{k}}
\]

(11)

and is enlarged by a factor of

\[
g(\omega_A) = \left| \frac{18\varepsilon(\omega_A)|\varepsilon(\omega_A)|[2\varepsilon(\omega_A) + 1][2\varepsilon_m(\omega_A) + 1]}{[\varepsilon(\omega_A) + \varepsilon_m(\omega_A)][2\varepsilon(\omega_A) + 1][2\varepsilon_m(\omega_A) + 1]} \right|^2
\]

(12)

with respect to its free-space value. Evidently, the potential is resonantly enhanced whenever \( \omega_A \) coincides with the frequency of an interface (surface) mode or with the frequency of a resonant mode of the Onsager cavities. We also observe that in this configuration, as long as the atoms are close to the interface, \( U_{AB}(\mathbf{r}_A, \mathbf{r}_B) \) is isotropic and depends only on the distance between the atoms in contrast to the situation when both atoms are on the same side of the interface [11]. Accordingly, the van der Waals forces on atoms \( \mathbf{F}_X = -\nabla \mathbf{U}_{AB}(\mathbf{r}_A, \mathbf{r}_B) \) obey the third Newton law

\[
\mathbf{F}_A(\mathbf{r}_A, \mathbf{r}_B) = -\frac{12Re[\sigma_{g}^2(\omega_A)]|\mathbf{d}_{g}|^2g(\omega_A)}{R^7}\mathbf{R} = -\mathbf{F}_B(\mathbf{r}_B, \mathbf{r}_A)
\]

(13)
where we therefore can be written as

\[ \varepsilon_m(\omega) = \eta + \frac{(\varepsilon_0 - \eta)\omega^2}{\omega^2 - \omega^2 - i\omega\Gamma}. \]  

(14)

where \( \eta \) and \( \varepsilon_0 \) are background and static dielectric constant, respectively, whereas \( \Gamma \) is the corresponding damping. Quantity \((\varepsilon_0 - \eta)\omega^2\), denoted as \( \eta \omega^2 \) in [11], is a measure of the coupling between the medium polarization and the field. The inverse of the average dielectric function of the system can therefore be written as

\[ \frac{1}{\varepsilon(\omega)} = \frac{2}{\eta + 1} \left( 1 - \frac{\varepsilon_0 - \eta}{\varepsilon_0 + 1} \frac{\omega^2}{\omega^2 - \omega^2 - i\omega\Gamma} \right). \]  

(15)

where

\[ \omega_S = \sqrt{\frac{\varepsilon_0 + 1}{\eta + 1}} \omega_T \]  

(16)

is the surface-mode frequency. In conjunction with equation (11), this equation explicitly demonstrates resonant enhancement of \( U_{AB} \) when \( \omega_A \sim \omega_S \). We note, however, that \( D_m(\omega) \) also exhibits a resonant structure. Indeed, with equation (14) this function reads

\[ D_m(\omega) = \frac{3}{2\eta + 1} \left( \eta + \frac{\varepsilon_0 - \eta}{\varepsilon_0 + 1} \frac{\omega_C^2}{\omega_C^2 - \omega^2 - i\omega\Gamma} \right), \]  

(17)

where

\[ \omega_C = \sqrt{\frac{2\varepsilon_0 + 1}{2\eta + 1}} \omega_T \]  

(18)

is the surface-mode frequency of the Onsager cavity. Since \( \omega_C \) is pretty close to \( \omega_S \), the enhancements of \( U_{AB} \) due to two resonances (partially) combine. Indeed, for small damping, we find that \( D_m(\omega_S) \approx 3 \) so that the peak value of the surface enhanced van der Waals potential/force given by

\[ g(\omega_S) \approx \frac{4(\varepsilon_0 - \eta)^2}{(\eta + 1)^2(\varepsilon_0 + 1)^2} \frac{\omega_S^2}{\Gamma^2} |D_m(\omega_S)|^2 \]  

(19)

is enlarged by (almost) nine times due to the local-field effect.

We illustrate these considerations in figure 2 where we have plotted \( U_{AB}^s \) with and without local-field correction factor as a function of \( \omega_A \). Note that the local-field corrected potential (full line) is additionally scaled by a factor of 10. To obtain these curves we have let in equation (11)

\[ a_s^R(\omega) = a_s^R(0) \frac{\omega_B^2}{\omega_B^2 - \omega^2 - i\omega\gamma_B} \]  

(20)

and expressed the potential in units \( U_{AB}^0 = 2|d_{AB}^0|^2a_s^R(0)/\hbar^6 \). Medium parameters are taken the same as in [11] and correspond to sapphire around the surface polariton resonance at \( \lambda_S = 12.21 \mu \text{m} \) [8], whereas parameters of atom \( B \) are chosen quite arbitrarily. We see that, besides a resonant structure at \( \omega_B \) as would exist in free space, the potential also exhibits resonances at \( \omega_S \) and \( \omega_C = 1.04\omega_S \) corresponding to the surface and local-field enhanced interaction between the atoms, respectively. As mentioned, owing to the proximity of these two resonances, each peak is actually due to the combined surface and local-field effects. Thus for the local-field corrected surface enhanced potential \( g(\omega_S) = 2947.6 \) whereas for the potential without the local-field correction the enhancement factor is 8.8 times smaller, \( g(\omega_S) = 336.2 \), which is of the same order as \( g(\omega_S) \) found in [11] for the case when the atoms are in vacuum close to the sapphire surface. Similarly, we find for the total enhancement factor at the Onsager cavity resonance \( g(\omega_C) = 1217.9 \), whereas the enhancement factor without the contribution of the surface-mode resonance at \( \omega_S \) is \( g(\omega_C) = 79.3 \).

4. Summary

In this work, we have considered the local-field corrected van der Waals interaction between an excited and a ground-state atom across the interface between two media. We have demonstrated that the (nonretarded) atom–atom potential is of the same form as in a homogeneous medium described by the average dielectric function of the two media in contact. Accordingly, as a function of the transition frequency of the excited atom, the potential generally exhibits resonances associated with the interface (surface) modes and with modes of small spherical cavities around the atoms figuring in the Onsager model for the local field. Numerical calculation performed for the vacuum–sapphire system reveals that in such systems, owing to the close surface and the Onsager cavity resonances, the van der Waals potential can be at the (vacuum–medium) surface-mode frequency enhanced by over three orders of magnitude, of which (nearly) one order of magnitude is due to the enhancement of the local-field correction factor for the medium.
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