

van der Waals energy of an atom in the proximity of thin metal films

M. Boström and Bo E. Sernelius

Department of Physics and Measurement Technology, Linköping University, S-581 83 Linköping, Sweden

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The van der Waals energy of a ground-state atom (or molecule) placed between two metal films is calculated at finite temperature. The attraction between thin metal films and a polarizable object can have half-integer separation dependence. This is in contrast to the usual integer separation dependence, shown for instance in the attraction between an atom and a solid surface. We examine how film thickness, retardation, and temperature influence the interaction. To illustrate the effect of finite thickness of the metal film we calculated the van der Waals energy of ground-state hydrogen and helium atoms, and hydrogen molecules, between thin silver films. We finally, briefly, discuss the possibility to measure this effect.

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I. INTRODUCTION

Recently, there has been considerable interest, both theoretically and experimentally, in fluctuation-induced forces. The fluctuating electromagnetic field in the vicinity of a metal surface is different from that of free space. The gradient of the potential energy, experienced by an atom in the proximity of a metal surface, gives rise to an attractive force. At short separations the optical properties of the metal surface, and the atom, influence the interaction. In this limit the force is referred to as the van der Waals force. At larger separations retardation, or the finite velocity of light, becomes important. In this limit, the interaction depends on the velocity of light, and the static polarizability of the atom. In this limit the force is referred to as the Casimir-Polder force. The purpose of this paper is to show how the van der Waals energy of an atom in between thin-metal films depends on the film thickness. We further examine effects of retardation and temperature on this interaction.

The interaction between a thin metal film with other objects can have fractional separation dependence related to the longitudinal two-dimensional collective excitations. Barash [1,2] calculated the van der Waals (vdW) interaction between a thin metal film with a second thin film. This interaction was found to sometimes have half integer separation dependence. This is one of the rare cases when the separation dependence is determined by collective excitations. This interesting phenomenon has received surprisingly little attention. In the limit when the second film is a very dilute dielectric film Barash obtained the (nonretarded) van der Waals energy of an atom in the proximity of a thin-metal film

$$F^{NR}(d) \approx -15\alpha_0 \hbar \omega_p \sqrt{\pi \delta / 256} d^{7/2}. \quad (1)$$

Here $d, \delta, \alpha_0, \hbar \omega_p$ are the atom-film separation, film thickness, static atomic polarizability, and the plasmon energy of the metal, respectively. At finite temperatures the relevant thermodynamic generalization of the van der Waals energy is the Helmholtz free energy (F). Sernelius and Björk [3] found similar separation dependence for the vdW energy between a pair of quantum wells. The retarded free energy of attraction between a pair of metal films [4] has recently been

calculated. In that particular case a gold film cannot be much thicker than 100 Å to be considered as thin.

The dispersion induced long range forces between solid surfaces and particles have received a lot more attention during the years. When retardation is neglected the long range potential between a surface and an atom takes the form [5] $F \approx -C_3 d^{-3}$. The coefficient C_3 has been calculated for various combinations of surfaces and atoms [6–9]. At larger separations the finite velocity of light becomes important. Casimir and Polder [10] investigated the interaction, between an atom and a conducting plane, in the separation range where retardation is important. At these separations the interaction varies as d^{-4} .

The self-energy shift, due to vdW interactions, of an atom in between metal half spaces has recently been measured. Anderson *et al.* [11] measured the deflection of Rydberg atoms towards a metal surface. Later Sandoghdar *et al.* [12] used laser spectroscopy to measure surface induced self-energy shifts of sodium atoms in excited states. Sukenik *et al.* [13] examined the deflection, due to the Casimir-Polder force, of ground-state sodium atoms. These experiments were in good agreement with theory. The surface induced self-energy shift has been subject to intense theoretical investigations. In particular Panat and Paranjape [14] recently calculated the selfenergy of a hydrogenic atom placed between two metallic slabs. One purpose of that work was to investigate the validity of the dipolar approximation. The origin of the self-energy shift can be considered to be changes in zero-point energies of the surface plasmons due to the presence of the atom. A review on both theoretical and experimental work on Casimir-Polder interactions is given in Ref. [15].

We argue that experiments, similar to those discussed, could be used to observe dimensionality effects due to finite film thickness. In this paper, we assume that the films are either free or deposited on a low energy surface. That is, we assume that the surface is sufficiently transparent so that it does not influence the interaction. In Sec. II we derive the vdW interaction energy of an atom placed between a pair of thin-metal films. We present the basic theory and derive some important asymptotes. In Sec. III the interaction energy of hydrogen molecules and hydrogen and helium atoms placed between two thin silver films is evaluated numeri-

cally. Very similar results would be obtained with, for example, gold films. Using these numerical calculations we investigate how thin the metal films must be to observe dimensionality effects. We further examine the effects of retardation and temperature. The results are discussed in Sec. IV where we further discuss the possibility to measure dimensionality effects.

II. THEORY

Zhou and Spruch [16] used a quantized surface mode technique to determine the van der Waals energy of a polarizable spherical object placed between two thin films. The interaction was derived within the dipole approximation. The approach of Ref. [16] is adopted in this paper. This is expected to be a good approximation, especially for ground state atoms, except at very small separations. Zhou and Spruch examined the zero-temperature interaction energy of atoms with dielectric films, and with perfect metals. In reality, of course, a perfect metal can never be anything but an approximation. In the present work we examine the vdW energy of an atom placed between thin metal films.

We consider two equal films a distance d apart, with a permittivity ϵ_1 and a thickness δ . These films are embedded in a second media, with permittivity ϵ_2 . An atom, with polarizability $\alpha(\omega)$, is separated a distance z from the closest surface. After trivial changes Eq. (4.9) of Ref. [16] can be rewritten for the present system as:

$$F(d) = \frac{\hbar}{2\pi} \int_0^\infty \int_0^\infty d\omega dq q \times \left(\frac{I_{TE}}{1 - G_{TE}^2 e^{-2\gamma_2 d}} + \frac{I_{TM}}{1 - G_{TM}^2 e^{-2\gamma_2 d}} \right), \quad (2)$$

where

$$I_{TE} = G_{TE} \omega^2 [2G_{TE} e^{-2\gamma_2 d} - (e^{2\gamma_2(z-d)} + e^{-2\gamma_2 z})] / \gamma_2 c^2; \quad (3)$$

$$I_{TM} = G_{TM} \omega^2 \left[2G_{TM} e^{-2\gamma_2 d} - \left(1 - \frac{2\gamma_2^2 c^2}{\epsilon_2 \omega^2} \right) \times (e^{2\gamma_2(z-d)} + e^{-2\gamma_2 z}) \right] / \gamma_2 c^2; \quad (4)$$

$$G_{TE/TM} = \frac{1 - e^{-2\gamma_1 \delta}}{1 - (\Delta_{12}^{TE/TM})^2 e^{-2\gamma_1 \delta}} \Delta_{12}^{TE/TM}; \quad (5)$$

$$\Delta_{12}^{TE}(q, i\omega) = \frac{\gamma_1 - \gamma_2}{\gamma_1 + \gamma_2},$$

$$\Delta_{12}^{TM}(q, i\omega) = \frac{\epsilon_2 \gamma_1 - \epsilon_1 \gamma_2}{\epsilon_2 \gamma_1 + \epsilon_1 \gamma_2}, \quad (6)$$

$$\gamma_i = \sqrt{q^2 + \epsilon_i \omega^2 / c^2}.$$

This is the van der Waals energy at zero temperature. There are two contributions to the energy, originating from

transverse magnetic (TM) and transverse electric (TE) modes. Due to the exponential functions, in Eqs. (3) and (4), only wave vectors of the order of $1/d$, or smaller, contribute significantly. We can therefore, approximately, neglect the wave vector dependence of the permittivity. This approximation obviously breaks down at too small separations. However, this is not the only complication in the limit of small separations. This approximation further neglects electron exchange effects and the detailed structure of the surface. The only required experimental quantity is obtained numerically, from the tabulated [17] optical excitation spectrum of silver and a Kramers-Kronig relationship

$$\epsilon(i\omega) = 1 + \frac{2}{\pi} \int_0^\infty dx \frac{x \epsilon''(x)}{x^2 + \omega^2}; \quad \epsilon'' = 2nk. \quad (7)$$

The numerical procedure is performed in a standard fashion, described in Ref. [4]. For a real metal the dielectric function at small momenta, and low frequencies, can be modeled with the simple Drude expression

$$\epsilon(i\omega) = 1 + \frac{\omega_p^2}{\omega(\omega + \omega_\tau)}. \quad (8)$$

We will use this model, neglecting damping, to derive asymptotic results. The damping is related to the static conductivity through the expression: $\sigma = \omega_p^2 / 4\pi\omega_\tau$. The London approximation is used to model the atomic polarizability:

$$\alpha(i\omega) = \alpha_0 / (1 + \omega^2 / \omega_a^2). \quad (9)$$

This model was used by Rauber *et al.* [6] to calculate van der Waals coefficients of atoms in the proximity of a substrate. Hydrogen atoms, hydrogen molecules, and helium atoms are taken to have static polarizabilities of 4.50, 5.439, and 1.384 au, respectively. The atomic unit (1 au) corresponds to $1.482 \times 10^{-25} \text{ cm}^3/\text{atom}$. The characteristic absorption frequencies of these particles are taken to be 11.65, 14.09, and 27.65 eV, respectively. These values were obtained in Ref. [6] from a fit to the static polarizability and the pair interaction between equal atoms.

The temperature dependence of the interaction energy is obtained from the Helmholtz free energy. The effect of temperature is taken into account if the frequency integration is replaced with a discrete summation [18].

$$\frac{\hbar}{2\pi} \int_0^\infty d\omega \rightarrow k_B T \sum_{n=0}^\infty ', \quad \omega_n = 2\pi k_B T n / \hbar. \quad (10)$$

The prime on the summation means that the $n=0$ term should be divided by two.

In the remainder of this section we will derive a few important asymptotes. We use the simple models, discussed earlier, for the permittivity and the atomic polarizability. We will consider the simplest possible situation, i.e., when $\epsilon_2 = 1$. Three different limiting asymptotes is discussed: the van der Waals asymptote, the Casimir-Polder asymptote and the high temperature asymptote. When retardation is neglected we can further distinguish between thick-and thin-

metal film. The nonretarded vdW energy of an atom in between a pair of thick metal films is given as

$$\lim_{\delta \rightarrow \infty} F^{NR}(d, z) \approx (\hbar/\pi) \int_0^\infty dq q^2 (e^{2q(z-d)} + e^{-2qz}) \times \int_0^\infty d\omega \alpha(i\omega) \Delta_{NR} / (1 - \Delta_{NR}^2 e^{-2qd}), \quad (11)$$

$$\Delta_{NR} \approx (1 - \epsilon_1)/(1 + \epsilon_1). \quad (12)$$

Although the frequency integration is straightforward, we consider two limiting cases of this expression. Consider first the special case when the atomic resonance occurs at much higher energy than the plasmon resonance. In this case, we can write an approximate expression for the van der Waals energy:

$$\lim_{\delta \rightarrow \infty} \lim_{\omega_a \gg \omega_p} F^{NR}(d, z) \approx \frac{-\hbar \omega_p \alpha_0}{4\sqrt{2}} \int_0^\infty dq q^2 (e^{2q(z-d)} + e^{-2qz}) \frac{2}{1 - e^{-2qd}}, \quad (13)$$

$$\lim_{\delta \rightarrow \infty} \lim_{\omega_a \gg \omega_p} F^{NR}(d, z = d/2) \approx -\hbar \omega_p \alpha_0 7 \zeta(3) / 4 \sqrt{2} d^3, \quad (14)$$

$$\lim_{d \rightarrow \infty} \lim_{\delta \rightarrow \infty} \lim_{\omega_a \gg \omega_p} F^{NR}(z) \approx -\hbar \omega_p \alpha_0 / 8 \sqrt{2} z^3. \quad (15)$$

The ratio of Eqs. (14) and (15), when the distance z is the same, is asymptotically $7\zeta(3)/4 \approx 2.1$, i.e., quite close to 2. Where the Riemann zeta function $\zeta(3) \approx 1.202$. Let us next consider the opposite limit when the atomic resonance energy can be neglected in comparison with the plasmon energy

$$\lim_{\delta \rightarrow \infty} \lim_{\omega_p \gg \omega_a} F^{NR}(d, z) \approx \frac{-\hbar \omega_a \alpha_0}{2} \int_0^\infty dq q^2 (e^{2q(z-d)} + e^{-2qz}) \frac{1 + e^{-2qd}}{(1 - e^{-2qd})^2}, \quad (16)$$

$$\lim_{\delta \rightarrow \infty} \lim_{\omega_p \gg \omega_a} F^{NR}(d, z = d/2) \approx \frac{-\pi^2 \hbar \omega_a \alpha_0}{4d^3}, \quad (17)$$

$$\lim_{d \rightarrow \infty} \lim_{\delta \rightarrow \infty} \lim_{\omega_p \gg \omega_a} F^{NR}(z) \approx \frac{-\hbar \omega_a \alpha_0}{8d^3}. \quad (18)$$

The ratio of Eq. (17) and (18), when the distance z is the same, is asymptotically $\approx \pi^2/4 \approx 2.47$.

To calculate the van der Waals energy of an atom in between thin metal films is slightly more complicated. In this case, there are four coupled surface plasmon modes: two longitudinal and two transverse excitations. In order to ob-

serve dimensionality effects the energy of the atomic resonance should not be negligible small compared to the collective excitations. If we approximate the atomic polarizability with its static value, and take the limit of very thin metal sheets, it becomes straight forward to derive the following asymptotes:

$$\lim_{\delta \rightarrow 0} F^{NR}(d, z) \approx \frac{-\hbar \omega_p \alpha_0 \sqrt{\delta}}{4\sqrt{2}} \int_0^\infty dq q^{2.5} (e^{2q(z-d)} + e^{-2qz}) \times \left(\frac{1}{\sqrt{1 - e^{-qd}}} + \frac{1}{\sqrt{1 + e^{-q}}} d \right), \quad (19)$$

$$\lim_{\delta \rightarrow 0} F^{NR}(d, z = d/2) \approx -6.70938 \hbar \omega_p \alpha_0 \sqrt{\delta} / 2 \sqrt{2} d^{3.5}, \quad (20)$$

$$\lim_{d \rightarrow \infty} \lim_{\delta \rightarrow 0} F^{NR}(z) \approx -15 \hbar \omega_p \alpha_0 \sqrt{\pi \delta} / 256 z^{3.5}. \quad (21)$$

The ratio of Eqs. (20) and (21), when the distance z is the same, is asymptotically 2.02. These asymptotes originate entirely from the two longitudinal, two-dimensional, surface modes. The coupling with the two transverse modes fades away more rapidly. They will only influence the result at very short separations. It is clear from Eqs. (13)–(21) that the vdW-induced self-energy shifts of an atom placed between a pair of metal films, and the corresponding force, depend on the thickness of the metal films. Equally obvious is that the optical properties of both metal and atom can be important for the attraction.

At both very small and very large separations, dimensionality effects vanish. It is well known that the interaction at small separations between objects is strongly dependent on the surface region. The thickness of the films becomes less important. At large separation when retardation becomes important, the thickness once again becomes unimportant. This is so because at large separations the exponential terms in Eq. (2) ensures that only small q and ω values give any significant contributions. In this limit the polarizability of the atom can be replaced with its static value. Furthermore, the permittivity becomes very large, so large that the asymptotic result becomes identical with the Casimir-Polder energy. The Casimir-Polder energy of an atom a distance \tilde{z} from the midpoint between perfect conductors is given as Eq. (4.48) in Ref. [16]:

$$F^{ret}(d, \tilde{z}) \approx \frac{\pi^3 \hbar c \alpha_0}{d^4} \left[\frac{1}{360} - \frac{3 - 2 \cos^2(\pi \tilde{z}/d)}{8 \cos^4(\pi \tilde{z}/d)} \right]. \quad (22)$$

Casimir and Polder derived the long-range retarded energy of an atom in the proximity of a perfect metal

$$F^{ret}(z) \approx -3 \hbar c \alpha_0 / 8 \pi z^4. \quad (23)$$

The ratio of Eqs. (22) and (23), when the distance z is the same, is asymptotically 1.98. The difference of having two surfaces rather than one in the proximity of an atom is an

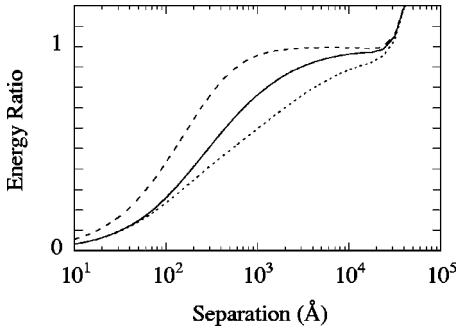


FIG. 1. The ratio between the calculated vdW energy of a ground-state hydrogen atom and the Casimir-Polder asymptote. The dashed line is the result in the proximity of a perfectly conducting surface. The solid (dotted) line is the result in the proximity of a silver half space (50 Å film).

increase in energy. In the mid-point between two films the ratio is the largest. Usually, it is close to two.

At even larger separation another asymptote becomes important: the long-range thermal asymptote. Thermal effects on the interaction between two atoms have received considerable interest lately [19,20]. The long-range temperature-dependent contribution originates from the $n=0$ part of the frequency summation. In the limit of zero frequency the permittivity [Eq. (8)] of a metal approaches infinity as $1/\omega$. If we compare with Eq. (2), we realize that this is too slow to get any contribution from the transverse electric $n=0$ contribution. This is true for the van der Waals energy of a pair of metal half-spaces; for a pair of thin metal films; and for an atom in between any of these systems. Using Eqs. (2) and (10) we obtain the following asymptotes:

$$F^{NR}(d, z, n=0) \approx -k_B T \alpha_0 \int_0^\infty dq q^2 (e^{2q(z-d)} + e^{-2qz}) / (1 - e^{-2qd}), \quad (24)$$

$$F^{NR}(d, z=d/2, n=0) \approx -7\zeta(3) k_B T \alpha_0 / 2d^3, \quad (25)$$

$$\lim_{d \rightarrow \infty} F^{NR}(z, n=0) \approx -k_B T \alpha_0 / 4z^3. \quad (26)$$

The ratio of Eqs. (25) and (26), when the distance z is the same, is asymptotically 2.1. An atom in the proximity of a perfect metal surface will cross over from the Casimir-Polder asymptote [Eq. (23)] to the 300 K thermal asymptote [Eq. (26)] at approximately $3.7 \mu\text{m}$.

III. NUMERICAL RESULTS

We have numerically determined the retarded free energy of an atom in the vicinity of thin metal films. The calculations have, unless otherwise is stated, been performed at a temperature of 300 K. Figure 1 shows the ratio between the van der Waals energy of a ground state hydrogen atom, in the proximity of a surface, and the Casimir-Polder energy [Eq. (23)]. Three surfaces are considered: a 50 Å silver film; a silver half space; a perfect metal surface. There are distinct differences between all these three cases. This figure indi-

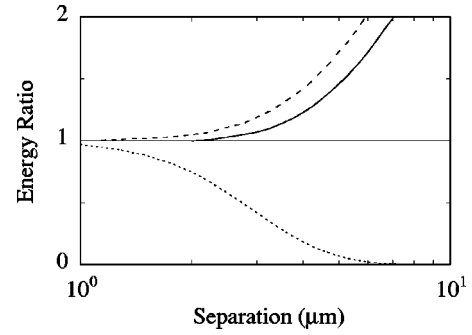


FIG. 2. A hydrogen atom in the proximity of a 50 Å silver film. The ratio between energy contributions calculated at 300 K to the same contribution calculated at 50 K. The dashed, dotted, and solid curves correspond to the TM, TE, and total contribution, respectively.

cates that there is a possibility to observe dimensionality effects. Due to retardation all three ratios approach 1. At large enough separations thermal effects become important. The thermal effects are examined in more detail in Fig. 2. We consider the van der Waals energy of a hydrogen atom in the proximity of a 50 Å-thick silver film. The results are presented as the ratio between three different energies calculated at 300 K to the corresponding energy calculated at 50 K. The energies considered are: the total free energy; the contribution from TM modes; and the contribution from the TE modes. At 300 K, thermal effects become important at approximately $1.2 \mu\text{m}$. The contribution from transverse magnetic (electric) modes increases (decreases) with temperature. Due to cancellations between these two contributions thermal effects do not become important until approximately $2 \mu\text{m}$. It is interesting to note that the entropy corresponding to the different energy contributions behaves very differently. The entropy associated with the transverse magnetic modes, approaches a positive, temperature independent, constant. On the contrary the entropy associated with the transverse electric modes has a negative value. At high-enough temperatures this entropy approaches zero. At low temperatures the sum of these two contributions can become negative. The different behaviors are simply related to whether or not the $n=0$ part gives any contribution.

In Fig. 3, we once again examine the vdW energy of a hydrogen atom in the proximity of a 50 Å-thick silver film. As a comparison we have included the vdW energy of an atom in the proximity of a silver half space. We have further added two more curves; the non-retarded vdW energy of an atom in the proximity of a 50 Å-thick silver film; and the Casimir-Polder asymptote. It is obvious that the non-retarded results cannot be applied at any separation of interest. Despite profound effects of retardation, dimensionality effects are still present. Retardation influences the dependence of basically every possible parameter. In the nonretarded situation considered in Sec. II we found an asymptotic square root thickness dependence. We examined this for a hydrogen atom a distance 800 Å away from a 20 Å thick silver film. We found 6% (28%) deviations from this power-law when retardation was neglected (included). The ratio between the vdW energy, of a hydrogen atom, in the middle of two silver

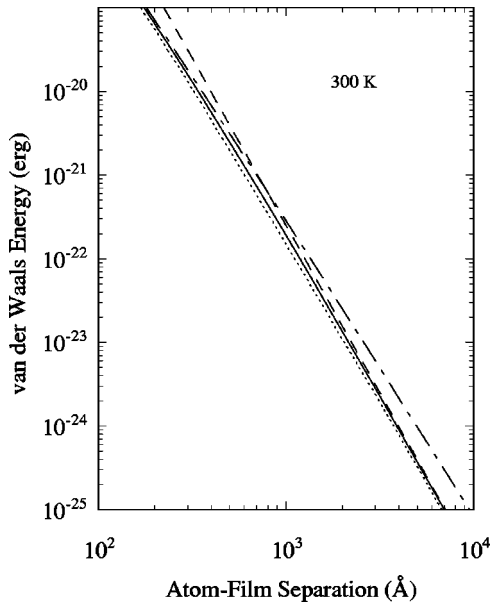


FIG. 3. The vdW energy of a hydrogen atom in the proximity of a silver surface. The solid (dotted) line is the result in the proximity of a half space (50 Å film). The dashed-dotted line is the non-retarded result in the proximity of a 50 Å film. The dashed line represents the Casimir-Polder asymptote.

films and the corresponding energy in between metal half spaces is shown in Fig. 4. From this figure follows that a 200 Å-thick film can almost be considered as thick. Guided by Fig. 3 and Fig. 4 we suggest that in order to observe dimensionality effects the film separation should not be larger than half a micrometer. We finally investigate dimensionality effects on the energy of an atom in a 4000 Å-wide cavity. In Fig. 5 we present the ratios between the vdW energies, of different atoms, in between thin and thick silver films. The

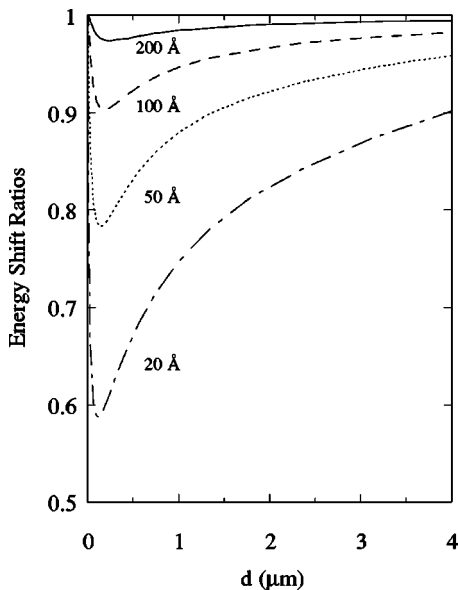


FIG. 4. The ratio between the vdW energy of a hydrogen atom in the midpoint between silver films and the corresponding energy between silver half spaces.

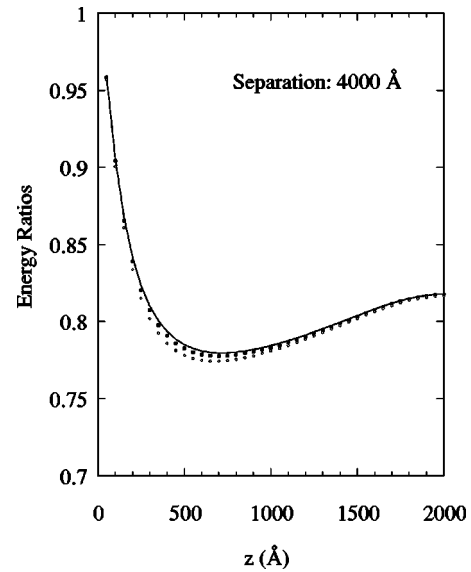


FIG. 5. The ratio between the vdW energy of an atom in between 50 Å silver films and the corresponding energy in between silver half spaces. The solid line corresponds to hydrogen atoms. The filled squares (open circles) correspond to hydrogen molecules (helium atoms).

energy ratio is found to be quite insensitive to the choice of atom. The atom with the highest characteristic absorption frequency is slightly more sensitive to dimensionality.

IV. DISCUSSION

In conclusion, we have demonstrated dimensionality effects on the van der Waals energy of an atom in the proximity of thin-metal films. When retardation is neglected film thickness influences the interaction. Although retardation is found to profoundly affect the interaction, dimensionality effects are still present. In order to observe dimensionality effects a film thickness of 100 Å can be taken as an approximate upper limit. We can distinguish between different separation ranges. At really short separations neither film thickness nor retardation matter. At separations exceeding 100 Å retardation strongly effects the interactions. In the situations considered in this paper the strictly nonretarded result cannot be applied at any separations of interest. It is suggested that numerical methods should be used in any quantitative evaluation of the van der Waals energy. In the “weakly retarded limit” the vdW energy of an atom, in the proximity of a thin film, depends on the film thickness. The differences between a thin and thick metal is found to be of equal magnitude, as the differences between a thick metal and a perfect metal. At even larger separation the interaction approaches the Casimir-Polder result. In this limit the metal surfaces can approximately be replaced with artificial, perfect metal surfaces. At even larger separations the retarded free energy follows the thermal asymptote. This asymptote has the same separation dependence as the non-retarded result between thick metal films. Let us briefly discuss the possibility to measure this dimensionality effect. When an atom moves between two metal surfaces the vdW force will

deflect its path. As discussed in Ref. [11] the energy conservation equation can be used to derive an approximate classical equation of motion. For an atom with constant velocity (v_x) parallel to the metal surfaces, and initially a distance z_0 from the closest surface, we have

$$x = v_x t \approx \sqrt{E_{k_x}} \int_{z_0}^z d\eta / \sqrt{F(z_0) + E_{k_z} - F(\eta)}. \quad (27)$$

The van der Waals energies presented in Fig. 5 are roughly 20% smaller in the proximity of the 50-Å silver film than in the proximity of a silver surface. It is clear that the probability of atoms hitting the surface decreases with decreasing film thickness. As the accuracy of experiments increases quite rapidly we believe that it is, or at least will be, possible to measure dimensionality effects on the van der Waals energy. Monte Carlo calculations, in which the atoms have a Maxwell-Boltzmann velocity distribution, can be used

to quantitatively investigate the opacity of atoms for a particular system. In Ref. [13] the result of such calculations were found to be in good agreement with experimental result.

We would finally like to point at a situation of great technological interest where these dimensionality effects could be relevant. Thin metal films has been used in gas sensors, e.g., a 50-Å silver film [21], deposited on silicon, has been used to detect hydrogen gas. The result presented in this work is not applicable to that particular case. However, it is possible to imagine situations when it would be relevant. At sufficiently low gas temperature it becomes important not to neglect vdW forces.

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