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Palasantzas, G.

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Adhesion stability of rough elastic films in presence of quantum vacuum fluctuations

G. PALASANTZAS *

Department of Applied Physics, Materials Science Center, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands

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Abstract—In this work the influence of vacuum fluctuations through the Casimir effect on the stability of an elastic film conformal onto to a self-affine rough substrate that is brought in close proximity with another parallel plate is studied. By considering the energy balance among adhesion, elastic and Casimir energies, it is shown that beyond certain parameter values (low roughness exponent H and/or high w/ξ ratio with w the rms roughness amplitude and ξ the lateral correlation length) the adhesion energy is counter-balanced by the elastic energy, allowing significant contributions in this regime by the Casimir energy. With increasing lateral correlation length ξ and/or decreasing roughness amplitude w , leading to long wavelength smoothing, the regime of roughness exponents H where the contribution of vacuum fluctuations is significant shifts drastically to a lower value. This occurs so that the short wavelength roughening compensates for the effect of long wavelength smoothing that decreases predominantly the elastic and Casimir energies.

Keywords: Adhesion; stability; elastic thin films; surface roughness; vacuum fluctuations; micro-electromechanical systems.

1. INTRODUCTION

The stability of epitaxial thin films by various factors has been of great interest in the research community due to their technological importance [1–6]. Among the various factors that can influence film stability, the most unusual factor is the Casimir force [7]. This force appears when another plate is brought into close proximity with an elastic film on a solid substrate [6]. Indeed, if the proximity between material objects becomes of the order of nanometers to a few micrometers, forces that are quantum mechanical in nature, namely, van der Waals and Casimir forces, become operative [7–11]. They also may be responsible for stiction, i.e., causing

*To whom correspondence should be addressed. Tel.: (31-50)363-4272. Fax: (31-50)363-4881.
E-mail: G.Palasantzas@rug.nl

adhesion of elements and, as a consequence, may profoundly change the actuation dynamics [12]. Historically, the Casimir effect has been considered to be an exotic quantum phenomenon that results from the perturbation of zero point vacuum fluctuations by conducting plates [7–11]. Because of its relatively short range it starts to make technological impact in the design and operation of MEMS/NEMS [12–24].

Recently, the morphological stability of epitaxial thin elastic films on a substrate has been examined in the presence of the Casimir force between the film surface and another parallel plate [6]. This study showed that when the two parallel plates are sufficiently close the Casimir force may have an important contribution to the surface evolution [6]. Critical undulation wavelengths were derived for two different limiting conditions. In particular it was shown that the Casimir force in both cases decreased the critical wavelength of the surface perturbation [6]. The latter effect can be important only for plate separations d comparable or smaller than the typical plasmon wavelength λ_p (e.g., $\lambda_p \approx 100$ nm for Al) due the fast decay of the Casimir force $\sim d^{-4}$ (for $d > \lambda_p$) [25–28]. The Casimir force or retarded van der Waals (vdW) force operates at separations typically $d \geq 50$ nm [29, 30]. A gradual transition from normal van der Waals forces which are dominant at separations $d < 10$ nm to Casimir forces occurs between these separations [29–31].

Since plates with rough surfaces lead to stronger attractive Casimir forces [20, 21, 25–28], it is natural to assume that they can affect profoundly the stability of thin elastic films adhered on these plates [6]. Indeed, this can be relevant for NEMS and nanometrology studies where, for example, researchers have measured the mechanical properties of ultra thin buckled polymer films with thickness down to 5 nm [32]. In other studies of Si-based nanochannels for biomolecular applications [33], it was observed that the walls of the channel were pierced off due to various reasons including also the Casimir force due to vacuum fluctuations. As a result we cannot exclude the contribution of quantum fluctuations in emerging nanomechanical systems [1–24, 32, 33].

Therefore, in the present study we will extend earlier adhesion stability studies [6] to the case of random self-affine rough surfaces observed in a wide variety of physical systems [34–36], where an elastic film conformal on a rough surface comes in close proximity (at nanoscale separations ≤ 100 nm) with another plate and confined vacuum fluctuations lead to an attractive Casimir force (Fig. 1). More precisely, we will investigate the subtle conditions under which quantum fluctuations together with the stored elastic energy within the film enhance film deadhesion from the substrate against the adhesion energy mediated by attractive forces between the elastic film and the rough substrate plate.

2. THEORETICAL DESCRIPTION

We can understand the role of random surface roughness on the adhesion of an elastic film as follows [37]. If h and λ represent the perpendicular and parallel roughness length scales, then an elastic energy $U_{el} \approx E\lambda h^2$ is stored within the film

Figure 1. Schematic of the film–plate system under consideration.

of Young’s modulus E , assuming that the film conforms to the substrate roughness, while the gain in adhesion energy is $U_{\text{ad}} \approx -\Delta\gamma\lambda^2$, where $-\Delta\gamma$ is the change in the local surface energy upon contact due to elastic film/substrate interaction (which is of vdW type). If $U_{\text{el}} < U_{\text{ad}}$, the elastic film will remain adhered to the substrate. The condition $U_{\text{el}} \approx -U_{\text{ad}}$ yields $\lambda \approx Eh^2/\Delta\gamma$ [38] providing a measure for the physical length scale over which spontaneous adhesion occurs.

Furthermore, we assume that the substrate surface roughness is described by the single valued random roughness fluctuation function $h(\vec{r})$ with $\vec{r}$ the in-plane position vector $\vec{r} = (x, y)$ so that $\langle h(\vec{r}) \rangle = 0$. Assuming Gaussian random roughness fluctuations and ensemble averaging over possible random roughness configurations, the adhesion energy of the elastic film to the rough substrate is given by [37, 38]

$$U_{\text{ad}} = -\Delta\gamma A_{\text{flat}} \int_0^{+\infty} (\sqrt{1 + \rho^2 u}) e^{-u} du, \quad \rho = \sqrt{\int_0^{Q_c} q^2 C(q) d^2 q}, \quad (1)$$

with A_{flat} the average macroscopic flat contact area, $Q_c = \pi/a_0$ with a_0 a lower roughness cut-off of atomic dimensions, and $\rho = \langle \sqrt{\langle (\nabla h)^2 \rangle} \rangle$ the average local surface slope of the substrate rough surface [39]. $C(q) = \langle |h(q)|^2 \rangle$ is the Fourier transform of the correlation function $C(r) = \langle h(\vec{r})h(0) \rangle$. The elastic energy stored in the film of elastic modulus E and Poisson’s ratio ν is given by [37, 38]

$$U_{\text{el}} = E\tilde{U}_{\text{el}} \quad \text{with} \quad \tilde{U}_{\text{el}} = \frac{A_{\text{flat}}}{4(1 - \nu^2)} \int_0^{Q_c} q C(q) d^2 q \quad (2)$$

assuming complete attachment to the surface over lateral length $L \gg \xi$ with ξ the lateral roughness correlation length.

Next, we consider the Casimir energy. For a rough plate the Casimir energy is given by [25–28]

$$U_{\text{Cas}} = U_{\text{cflat}} + \frac{1}{2} \left(\frac{\partial^2 U_{\text{cflat}}}{\partial d^2} \right) \left[\int P(q) C(q) \frac{d^2 q}{(2\pi)^2} \right], \quad (3)$$

with $U_{\text{cflat}} = (\pi^2 \hbar c / 720 d^3) A_{\text{flat}}$ the Casimir energy for flat perfectly conducting plates, d is the average distance between the plates, c the velocity of light and $\hbar$ the Planck constant. The scattering function $P(q)$ for finite plasmon wavelength λ_p is given by the power-law expressions [25–27]:

$$P(q) = \begin{cases} \text{if } d < \lambda_p: 0.4492dq & \text{for } q \gg 2\pi/d, q \gg 2\pi/\lambda_p \\ (1/3)dq & \text{for } 2\pi/d \ll q \ll 2\pi/\lambda_p \\ \text{if } d > \lambda_p: (7\lambda_p/15\pi)q & \text{for } q \gg 2\pi/d, q \gg 2/\lambda_p, \end{cases} \quad (4)$$

which take into account corrections due to finite conductivity for plate separations $d < \lambda_p$ [25–27]. The optical response of the plates is assumed to be described by the dielectric function $\varepsilon(\omega) = 1 - (\omega_p/\omega)^2$, with ω_p the plasma frequency. Substitution of equation (4) in (3) yields

$$U_{\text{Cas}} = U_{\text{cflat}} + \frac{1}{2} \left(\frac{\partial^2 U_{\text{cflat}}}{\partial d^2} \right) [Gd],$$

$$G = \begin{cases} 0.4492 \int_{Q_d}^{Q_c} q C(q) \frac{d^2 q}{(2\pi)^2} & \text{if } d < \lambda_p \\ \frac{1}{3} \int_{Q_d}^{Q_{\lambda_p}} q C(q) \frac{d^2 q}{(2\pi)^2} + \frac{7}{15\pi} \frac{\lambda_p}{d} \int_{Q_{\lambda_p}}^{Q_c} q C(q) \frac{d^2 q}{(2\pi)^2} & \text{if } d > \lambda_p, \end{cases} \quad (5)$$

where $Q_{\lambda_p} = 2\pi/\lambda_p$ and $Q_d = 2\pi/d$.

From combination of equations (1)–(5) we can compute the total free energy of the elastic film which is given by

$$U_{\text{total}} = U_{\text{ad}} + U_{\text{Cas}} + U_{\text{el}}. \quad (6)$$

Adhesion stability requires that $U_{\text{total}} < 0$ or alternatively $|U_{\text{ad}}| > |U_{\text{Cas}}| + |U_{\text{el}}|$.

3. RESULTS AND DISCUSSION

Computations of the roughness effects will require knowledge of the roughness spectra $\langle |h(q)|^2 \rangle$. A wide variety of surfaces and interfaces possess the so-called self-affine roughness [34]. In this case the roughness spectrum shows a power-law scaling [34–36, 40, 41] $\langle |h(q)|^2 \rangle \propto q^{-2-2H}$ if $q\xi \gg 1$ and $\langle |h(q)|^2 \rangle \propto \text{const}$ if $q\xi \ll 1$. This is satisfied by the analytic model [40, 41]

$$\langle |h(q)|^2 \rangle = 2\pi \frac{w^2 \xi^2}{(1 + a q^2 \xi^2)^{(1+H)}}, \quad (7)$$

with $a = (1/2H)[1 - (1 + a Q_c^2 \xi^2)^{-H}]$ if $0 < H < 1$, and $a = (1/2) \ln(1 + a Q_c^2 \xi^2)$ if $H = 0$ [40, 41]. Small values of H (approx. 0) characterize jagged or irregular surfaces, while large values of H (approx. 1) refer to surfaces with smooth hills and

valleys [34–36, 40, 41]. For other models see Refs [34–36, 42–44]. The parameter w is the root-mean-squared (rms) roughness amplitude.

We should point out that the roughness model in equation (6) describes bounded roughness not only because we assume finite correlation lengths ξ ($< +\infty$) but also because we consider roughness exponents $H < 1$. Indeed, the height difference correlation function $g(r) = \langle [h(r) - h(0)]^2 \rangle \propto \int \langle |h(q)|^2 \rangle [\exp(i\vec{k} \cdot \vec{r}) - 1] d^2q$ scales for $r \ll \xi$ as $g(r) \propto r^{2H}$ which ensures that the ratio $g(r)/r^2 \rightarrow 0$ for $r \rightarrow +\infty$ if and only if $H < 1$ ensuring bounded roughness besides the restriction of finite correlation length ξ .

If we substitute equation (7) into equations (1), (2) and (5) we can obtain analytic results for roughness exponents $H = 0, 0.5$ and 1 . Equation (7) yields for the average local slope ρ_{rms} the analytic form $\rho = (w/\sqrt{2}\xi a) \sqrt{(1-H)^{-1}[(1+aQ_c^2\xi^2)^{1-H} - 1] - 2a}$ [40, 41], and, therefore, the analytic expression for the adhesion energy for weak roughness ($\rho_{\text{rms}} < 1$) is given by

$$U_{\text{ad}} \cong -\Delta\gamma \left[1 + \frac{1}{2}\rho^2 + \sum_{n=2}^{+\infty} S(n)\rho^{2n} \right], \quad (8)$$

with $S(n) = \{1 \cdot 1 \cdot 3 \cdot 5 \cdots (2n-3)\}(-1)^{n-1}/2^n$. For the elastic energy we have analytic results for the characteristic exponents $H = 0, 0.5$ and 1 . If we set $w_0 = \sqrt{4(1-v^2)/E}$ we have

$$U_{\text{el}}|_{H=0} = A_{\text{flat}} \frac{w^2}{w_0^2} \left\{ \frac{1}{a}(Q_c) - \frac{1}{a^{3/2}\xi} [\tan^{-1}(X_c)] \right\}, \quad (9)$$

$$U_{\text{el}}|_{H=0.5} = A_{\text{flat}} \frac{w^2}{w_0^2} \left\{ \frac{1}{a^{3/2}\xi} [\sinh^{-1}(X_c)] - \frac{1}{a} [Q_c T_c^{-1/2}] \right\}, \quad (10)$$

$$U_{\text{el}}|_{H=1} = A_{\text{flat}} \frac{w^2}{w_0^2} \left\{ \frac{1}{a^{3/2}\xi} [\tan^{-1}(X_c)] - \frac{1}{2a} [Q_c T_c^{-1}] \right\}, \quad (11)$$

with $X_c = \sqrt{a}\xi Q_c$ and $T_c = (1 + aQ_c^2\xi^2)$. Finally, the roughness correction to the Casimir energy as expressed by the factor G is given for roughness exponents $H = 0, 0.5$, and 1 by the expressions

$$G^{H=0} = w^2 \begin{cases} 0.4492 \left\{ \frac{1}{a} (Q_c - Q_d) - \frac{1}{a^{3/2}\xi} [\tan^{-1}(u)|_{X_d}^{X_c}] \right\} & \text{if } d < \lambda_p \\ \frac{1}{3} \left\{ \frac{1}{a} (Q_{\lambda_p} - Q_d) - \frac{1}{a^{3/2}\xi} [\tan^{-1}(u)|_{X_d}^{X_{\lambda_p}}] \right\} \\ \quad + \frac{7}{15\pi} \frac{\lambda_p}{d} \left\{ \frac{1}{a} (Q_c - Q_{\lambda_p}) - \frac{1}{a^{3/2}\xi} [\tan^{-1}(u)|_{X_{\lambda_p}}^{X_c}] \right\} & \text{if } d > \lambda_p, \end{cases} \quad (12)$$

$$G^{H=0.5} = w^2 \begin{cases} 0.4492 \left\{ \frac{1}{a^{3/2}\xi} [\sinh^{-1}(u)|_{X_d}^{X_c}] - \frac{1}{a} [Q_c T_c^{-1/2} - Q_d T_d^{-1/2}] \right\} & \text{if } d < \lambda_p \\ \frac{1}{3} \left\{ \frac{1}{a^{3/2}\xi} [\sinh^{-1}(u)|_{X_d}^{X_{\lambda_p}}] - \frac{1}{a} [Q_{\lambda_p} T_{\lambda_p}^{-1/2} - Q_d T_d^{-1/2}] \right\} \\ + \frac{7}{15\pi} \frac{\lambda_p}{d} \left\{ \frac{1}{a^{3/2}\xi} [\sinh^{-1}(u)|_{X_{\lambda_p}}^{X_c}] - \frac{1}{a} [Q_c T_c^{-1/2} - Q_{\lambda_p} T_{\lambda_p}^{-1/2}] \right\} & \text{if } d > \lambda_p, \end{cases} \quad (13)$$

$$G^{H=1} = w^2 \begin{cases} 0.4492 \left\{ \frac{1}{a^{3/2}\xi} [\tan^{-1}(u)|_{X_d}^{X_c}] - \frac{1}{2a} [Q_c T_c^{-1} - Q_d T_d^{-1}] \right\} & \text{if } d < \lambda_p \\ \frac{1}{3} \left\{ \frac{1}{a^{3/2}\xi} [\tan^{-1}(u)|_{X_d}^{X_{\lambda_p}}] - \frac{1}{2a} [Q_{\lambda_p} T_{\lambda_p}^{-1} - Q_d T_d^{-1}] \right\} \\ + \frac{7}{15\pi} \frac{\lambda_p}{d} \left\{ \frac{1}{a^{3/2}\xi} [\tan^{-1}(u)|_{X_{\lambda_p}}^{X_c}] - \frac{1}{2a} [Q_c T_c^{-1} - Q_{\lambda_p} T_{\lambda_p}^{-1}] \right\} & \text{if } d > \lambda_p, \end{cases} \quad (14)$$

with $L(x)|_A^B = L(B) - L(A)$, $X_d = \sqrt{a}\xi Q_d$, $X_{\lambda_p} = \sqrt{a}\xi Q_{\lambda_p}$, $T_{\lambda_p} = 1 + (X_{\lambda_p})^2$ and $T_d = 1 + (X_d)^2$. Note that we consider for the scattering function $P(q)$ the power-law regimes from which deviations occur for wave vectors $q < 10^{-3} \text{ nm}^{-1}$ where $P(q) \ll 1$ [25–28]. On the other hand, for $q \ll 1 \text{ nm}^{-1}$ (or more precisely $q\xi \ll 1$) the roughness spectrum approaches the asymptotic limit $\langle |h(q)|^2 \rangle \approx (2\pi)w^2\xi^2$. As a result the error by considering only the power-law approximation for $P(q)$ is not significant.

Calculations were performed (if not stated otherwise) for a choice of $\Delta\gamma = 9.6 \times 10^{-3} \text{ J/m}^2$, $\nu = 0.4$ and $E = 100 \text{ MPa}$. Figure 2 shows $|U_{\text{ad-el}}|/|U_{\text{Cas}}|$, with $U_{\text{ad-el}} = |U_{\text{ad}}| - |U_{\text{el}}|$, for different plate distances d . It shows that with increasing plate distance d the influence of the Casimir energy decreases. The minimum position around which we have $||U_{\text{ad}}| - |U_{\text{el}}|| \approx |U_{\text{Cas}}|$ is favoured for rougher surfaces either at short ($< \xi$) and/or long lateral roughness wavelengths ($> \xi$). Surface roughening leads to higher adhesion energy (increase in surface area and increased local surface slope). However, beyond certain parameter values, e.g., low exponent H , especially for $H < 0.5$, and/or high w/ξ ratio, the elastic energy becomes comparable with the adhesion energy and as a result it compensates the latter. In this regime as indicated by the dotted circle in Fig. 2 significant contributions can arise from the Casimir force that enhances the film deadhesion from the substrate. The left-hand side of the minimum corresponds to deadhesion regime, where the elastic energy and the Casimir force deadhere the elastic film

Figure 2. Energy ratio $|U_{ad-el}|/|U_{Cas}|$ as a function of the roughness exponent H for $w = 5$ nm, $\xi = 200$ nm, $\Delta\gamma = 9.6 \times 10^{-3}$ J/m², $\nu = 0.4$, $E = 100$ MPa and various plate separations d . The arrows indicate the adhesion and deadhesion regimes.

Figure 3. Energy ratio $|U_{ad-el}|/|U_{Cas}|$ as a function of the roughness exponent H for $w = 5$ nm, various correlation lengths ξ , $\Delta\gamma = 9.6 \times 10^{-3}$ J/m², $\nu = 0.4$, $E = 100$ MPa and $d = 30$ nm.

from the rough substrate (or $|U_{ad}| < |U_{Cas}| + |U_{el}|$), while the right-hand side corresponds to the adhesion regime ($|U_{ad}| > |U_{Cas}| + |U_{el}|$).

Figure 3 shows that with increasing lateral correlation length ξ or smoother surfaces at long wavelengths, the position of the minimum and therefore of

Figure 4. Energy ratio $|U_{ad-el}|/|U_{Cas}|$ as a function of the correlation length ξ (normalized with respect to the plate distance d) for various roughness exponents H , $w = 5$ nm, $\Delta\gamma = 9.6 \times 10^{-3}$ J/m², $\nu = 0.4$, $E = 100$ MPa, and $d = 30$ nm (the arrows indicate the displacement of the apparent minimum, as well as the adhesion and deadhesion regimes).

the regime where the contribution of vacuum fluctuations is important shifts considerably to lower roughness exponents H in order to compensate for the effect of smoothing at long wavelengths. Alternatively, as shown in Fig. 4, the minimum occurs for smaller correlation lengths with increasing roughness exponent H . This is in qualitative agreement with the fact that the Casimir force decreases the critical wavelength of the surface perturbation as found in earlier studies assuming periodic rough profiles [6]. However, as the arrows indicate with increasing roughness exponent H the minimum position where quantum effects are operative changes less drastically, especially close to the asymptotic value H approx. 1.

Figure 5 shows that the minimum position is strongly affected by the roughness amplitude. Lowering the roughness amplitude w leads also to a minimum position at lower roughness exponents H that are required for compensation of the long wavelength smoothing. This leads to significantly lower elastic energies and, therefore, to a negligible influence of vacuum fluctuations. The effect of the rms roughness amplitude w in comparison to that of the correlation length ξ and the roughness exponent H (compared with Figs 2–4) appears to be very significant.

In any case, as a generic finding we observe that the influence of the Casimir force on the film adhesion properties is significant for small separations d or more precisely lower than the plasmon wavelength λ_P which is the regime where surface coatings are poor reflectors and surface roughness plays a significant role in vacuum fluctuations [25–27]. Notably, for plate separations less than 10 nm, we should consider directly the effect of van der Waals forces on adhesion stability studies

Figure 5. Energy ratio $|U_{\text{ad-el}}|/|U_{\text{Cas}}|$ as a function of the roughness exponent H for two roughness amplitudes w , correlation length $\xi = 200$ nm, $\Delta\gamma = 9.6 \times 10^{-3}$ J/m², $\nu = 0.4$, $E = 100$ MPa and $d = 30$ nm.

since any retardation effect becomes negligible [29–31]. The latter case will be considered in more detail in future studies.

4. CONCLUSIONS

In summary, we studied the influence of the Casimir effect on the adhesion of a thin elastic film on a self-affine rough surface. It is shown that although an initial surface roughening leads to higher adhesion energy, beyond certain parameter values (low exponents H and/or high w/ξ ratios) the elastic energy becomes comparable with the adhesion energy and it leads to significant contribution by the Casimir force. Indeed, it is shown that with increasing lateral correlation length ξ and/or decreasing roughness amplitude w leading to long wavelength smoothing, the regime of roughness exponents H where the contribution of vacuum fluctuations is significant shifts considerably to lower values. The latter occurs in order that the short wavelength roughening (expressed by the roughness exponent H) compensates the effect of long wavelength smoothing that strongly decreases the elastic and Casimir energies and, therefore, preserves the energy balance $|U_{\text{ad}}| - |U_{\text{Cas}}| \approx |U_{\text{el}}|$.

Our results demonstrate that in micro/nanoelectromechanical systems where thin elastic films may be involved, quantum fluctuations at separations below 100 nm can play a significant role in the delamination from rough substrates or collapse of nanostructures [33]. The latter imposes restrictions onto the system functionality, and clearly demands a precise knowledge of the corresponding surface roughness parameters. This can be achieved, for example, using scanning probe microscopy,

X-ray reflectivity, electron diffraction, etc. [34–36, 40–44] in order to estimate correctly their effect on film adhesion/delamination properties.

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