Eur. Phys. J. E **8**, 347–351 (2002)
DOI 10.1140/epje/i2002-10018-0

THE EUROPEAN PHYSICAL JOURNAL E
EDP Sciences

Acoustic instabilities in thin polymer films

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Received 5 March 2002 and Received in final form 21 May 2002

Abstract. The spatial confinement of a fluctuation spectrum leads to forces at the confining boundaries. While electromagnetic (EM) fluctuations lead to the well-known dispersion forces, the acoustic analogy has widely been neglected. We show that the strength of the forces resulting from confined acoustic modes may be of the same order of magnitude as van der Waals forces. Additionally, the predicted scaling behavior is identical to the non-retarded case of the EM fluctuations. Our results suggest that dewetting experiments using polymer films are strongly influenced by the acoustic dispersion forces.

PACS. 47.20.Ma Interfacial instability – 68.15.+e Liquid thin films – 05.40.-a Fluctuation phenomena, random processes, noise, and Brownian motion

1 Introduction

The attraction of two conducting plates due to the EM vacuum fluctuations was predicted by Casimir in 1948 [1]. This effect can be understood by considering the radiation pressure exerted by the plane waves of the random EM field [2]. Inside the cavity, modes with wavelengths larger than twice the plate spacing are suppressed, leading to a reduced density of states by the confinement. The difference in densities of states outside and inside the cavity leads to an imbalance in radiation pressures exerted onto the boundaries. This results in an attractive force between the two plates. In addition, the mode spectrum inside is discrete due to the large coherence length of the photons. The small resulting net force has been measured recently by Lamoreaux [3]. The concept introduced by Casimir is, however, not limited to electromagnetic waves. The confinement of any fluctuating field leads to a modification of the associated free energy and therefore to fluctuation induced forces [4]. Examples of Casimir forces caused by thermal fluctuations include finite-size corrections in the free energy of binary-liquid mixtures near the critical point, long-ranged correlations in superfluids and liquid crystals and the fluctuations of counterions in charged fluids [4]. In a macroscopic experiment, Larraza *et al.* [5] introduced the notion of an acoustic Casimir effect by measuring the force between two plates in an external sound field. In the absence of an external source, Bschorr [6] calculated a similar force based on the thermal pressure fluctuations in air.

A more general approach of EM phenomena in a confined medium leads to the disjoining pressure [7,8] which is known to destabilize thin liquid films [9]. Such dewetting instabilities have been the focus of recent research because of the fundamental interest in the interactions of liquids near surfaces, but also because of the practical relevance of film stability for a variety of applications. While several factors may contribute to the breakup of thin films, most effort has been devoted to investigate the effects of long-ranged van der Waals (vdW) forces [9–13]. Even though the mechanisms of film break-up by dispersive forces are well understood, quantitative agreement between models and experiments is often lacking [11,12,14]. Since the theory of vdW forces is well established [7,8], such disagreements are indicative of additional driving forces. Recent work demonstrates that electrostatic effects [15–17] and radiation pressures due to temperature gradients [18] may contribute to the destabilization of thin films. Here, we describe a new destabilizing mechanism, the acoustic analog to the Casimir pressure, or in more general terms, the acoustic disjoining pressure. The resulting dynamical instability is yet another variation of spinodal dewetting with driving forces comparable to dispersive interactions.

2 Theoretical model

Dzyaloshinskii *et al.* [8] have estimated the interfacial pressure of acoustic fluctuations (phonons). At low temperatures ($T = 0\,\mathrm{K}$), analogous to the EM case, an interfacial pressure $p_{\mathrm{ac}} \propto hu/d^4$ is predicted, where d is the

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thickness of the confined medium and h is Planck's constant. This has to be compared to the EM case, where $p_{\mathrm{r}} \propto hc/d^4$ for retarded or $p_{\mathrm{nr}} \propto hc/ad^3 \propto A/d^3$ for non-retarded vdW forces. u and c are the velocities of sound and light, a is a crossover length, and A is the Hamaker constant. Since typical sound velocities are approximately five orders of magnitude smaller than the speed of light, it is clear that p_{ac} is negligible for $T = 0\,\mathrm{K}$. However, for non-zero temperatures the influence of temperature is predominant. The criterion for classical behavior, $h\nu \ll kT$ (k Boltzmann's constant) with $\nu \propto u/d$ is for common liquids very well satisfied, whereas in the EM case the opposite is true ($\nu \propto c/d$). From dimensional considerations $p_{\mathrm{ac}} \propto kT/d^3$ in the classical regime. Since kT is of the same order of magnitude as typical Hamaker constants A [19], it is obvious that acoustic fluctuations may contribute to the destabilizing pressure at the film surfaces. Furthermore, since p_{ac} has the same scaling behavior as the non-retarded van der Waals force, it is experimentally very difficult to distinguish the two effects.

We consider a free-standing liquid film (medium 2; medium 1 refers to air or vacuum). To calculate the pressure contribution from the acoustic fluctuations in liquids, we follow the calculations of Debye [20] for the energy density of phonons in solids [21] in close analogy to the work of Bschorr [6]. We assume an elastic liquid medium with propagating longitudinal waves of sound velocity u_i up to the maximal Debye frequency $\nu_{\mathrm{D}}^{(i)} = u_i[9N/4\pi V_i]^{1/3}$, where N is Avogadro's number and V_i the molar volume. The radiation pressure exerted by plane waves incident vertically on a perfectly reflecting surface is $2j/u_i$ with the energy flux $j = u_i e$ and the phonon energy density e. Since the coherence length of phonons in liquids is fairly short, we can neglect interference effects and model the mode spectrum as a superposition of orthogonal waves. Therefore, the energy flux in one of the six orthogonal directions is $j/6$, resulting in a phonon pressure

$$p_{\mathrm{ac}} = \frac{1}{3} e\,. \tag{1}$$

The energy per phonon is kT (assuming $kT \gg h\nu$). To obtain the total energy density, the phonon energy has to be weighted by the density of states in medium i, $dn_i = 4\pi\nu^2/u_i^3 d\nu$, and integrated over the relevant phonon frequencies. Outside the film, phonons of all frequencies up to ν_{D} exist. Inside the film, the cutoff wavelength $\lambda_{\mathrm{c}} = 2d$ introduces a lower cutoff frequency

$$\nu_{\mathrm{c}} = \frac{u_i}{\lambda_{\mathrm{c}}} = \frac{u_i}{2d}\,. \tag{2}$$

The net radiation pressure (termed "acoustic Casimir effect" by Larraza *et al.* [5]) is readily calculated,

$$p_{\mathrm{ac}} = \frac{1}{3}\left[\int_0^{\nu_{\mathrm{D}}^{(1)}} kT\mathrm{d}n_1 - \int_{\nu_{\mathrm{c}}}^{\nu_{\mathrm{D}}^{(2)}} kT\mathrm{d}n_2\right] = \frac{\pi kT}{18d^3} + p_0'\,, \tag{3}$$

where p_0' is a pressure term that does not depend on film thickness.

In the absence of any further effects (*e.g.*, [15,18]), the total pressure acting inside a film with thickness d is given by

$$p\,(d) = p_0 + p_{\mathrm{L}} + p_{\mathrm{dis}} + p_{\mathrm{ac}} = p_0 - C_1\gamma\partial_{xx}d + \frac{A_{121}}{6\pi d^3} + \frac{\pi kT}{18d^3}\,, \tag{4}$$

where ∂_x denotes the partial derivative with respect to the lateral coordinate x. p_0 includes the d-independent contributions from the atmosphere. The second term describes the Laplace pressure with the surface tension γ between medium 1 and 2. The disjoining pressure p_{dis} given by the third term is characterized by the Hamaker constant A_{121} of medium 2 sandwiched by medium 1, for non-retarded dispersion forces.

The Navier-Stokes and continuity equations for an incompressible fluid yields the equation of motion for the lateral flux (in the x-direction) within the film

$$\partial_t d = \partial_x\left[\frac{d^3}{C_2\eta}\partial_x p\,(d)\right]\,, \tag{5}$$

where η is the zero-frequency viscosity. The constants C_1 and C_2 depend on the hydrodynamic boundary conditions at the two film surfaces. For two free boundaries, assuming a squeezing mode, we have $C_1 = 0.5$ and $C_2 = 12$ [9]. In the derivation of equation (3), we assumed the interface between medium 1 and 2 to be perfectly reflective. For the case that medium 1 is a gas and medium 2 a liquid, this condition is well satisfied.

A linear stability analysis determines whether the interfacial fluctuations of the thermal capillary wave spectrum with wave number q and amplitude δd_0, $d\,(x,t) = d_0 + \delta d_0 \exp(iqx + t/\tau)$ are exponentially amplified (growth rate $\tau^{-1} > 0$) or damped ($\tau^{-1} < 0$). In the long-wavelength approximation ($\delta d_0 \ll d \ll 1/q$), the dispersion relation is

$$\frac{1}{\tau} = -\frac{d_0^3}{C_2\eta}\left[\gamma C_1 q^4 + \partial_d\,(p_{\mathrm{dis}} + p_{\mathrm{ac}})\,q^2\right]\,. \tag{6}$$

The film is unstable when $\partial_d\,(p_{\mathrm{dis}} + p_{\mathrm{ac}}) < 0$ and all modes with $q < q_{\mathrm{c}}$ are amplified. The fastest growing mode q_m is then given by the maximum of equation (6),

$$q_m^2 = \frac{1}{2}q_{\mathrm{c}}^2 = -\frac{1}{2\gamma C_1}\partial_d\,(p_{\mathrm{dis}} + p_{\mathrm{ac}}) \tag{7}$$

with the maximal growth rate

$$\frac{1}{\tau_m} = \frac{C_1}{C_2}\frac{\gamma d_0^3}{\eta}q_m^4\,. \tag{8}$$

Since both p_{dis} and p_{ac} have the same dependence on d, the scaling of q_m with d is also identical. Thus, the force balance between the dispersive and acoustic terms depends exclusively on their absolute strength. It is therefore important to explore several corrections and higher-order effects that may contribute to p_{ac}.

While for most experimental systems the temperature is larger than the Debye temperature $\Theta_\mathrm{D} = h\nu_\mathrm{D}/k$ (for polystyrene $\Theta_\mathrm{D} \approx 100\,\mathrm{K}$), the classical limit $T \gg \Theta_\mathrm{D}$ needs to be verified. For $T \leq \Theta_\mathrm{D}$, the quantum-statistical character of the phonon energy has to be taken into account. Using $x = h\nu/kT$, the energy per phonon is given by

$$\frac{h\nu}{e^{\frac{h\nu}{kT}}-1} = kT\left[\frac{x}{e^x-1}\right] = kT\left[1-\frac{1}{2}x+\sum_{n=1}^{\infty}(-1)^{n+1}\frac{B_n x^{2n}}{(2n)!}\right]. \qquad (9)$$

The series converges for $|x|<2\pi$, where $B_n = 2(2n)!\zeta(2n)/(2\pi)^{2n}$ are the Bernoulli numbers with the Riemann zeta-function $\zeta(z)$. Using equation (9) to calculate the acoustic pressure in equation (3), we obtain

$$p_\mathrm{ac} = p_0'\left(\Theta_\mathrm{D}/T\right) + \frac{\pi kT}{18d^3}\left[1-\frac{3}{16}\frac{hu}{kTd} + \sum_{n=1}^{\infty}\frac{6(-1)^{n+1}\zeta(2n)}{(2n+3)(4\pi)^{2n}}\left(\frac{hu}{kTd}\right)^{2n}\right]. \qquad (10)$$

The sum converges rapidly for $\frac{hu}{kTd} \leq 1$. For $u = O(1000)$ m/s, $T = O(300)\,\mathrm{K}$, and $d \geq 1\,\mathrm{nm}$, $\frac{hu}{kTd} \leq O(0.1)$ and only the first term in the expansion has to be considered. Down to very thin films ($d \approx 1\,\mathrm{nm}$) this correction is less than 5%. Therefore, even for temperatures below Θ_D where the quantum statistics is important, the effect is still dominated by kT.

The additivity of p_dis and p_ac in equation (4) implies the assumption that thermal and EM fluctuations are not correlated. For dielectric materials there is no direct coupling between the EM and vibrational molecular modes. While higher-order cross-correlations have to be worked out in a more detailed model, we restrict our treatment to the approximation, in which the EM and Debye density of states are decoupled.

So far, we have restricted our model to free-standing films. Since experiments on free-standing films are difficult to perform [14], most experiments consider supported films (medium 2 (liquid) sandwiched between medium 1 (air) and medium 3 (substrate)) [11–13]. The theoretical framework for supported films is similar to the case of free-standing films: in equation (4), A_{121} has to be replaced by A_{123} and the changed hydrodynamic boundary condition at the solid substrate changes the prefactors in equations (4-8) to $C_1 = 1$ and $C_2 = 3$ [10].

In addition, two further effects arise due to the elastic coupling of the film to an external medium. First, the coupling modifies the mode spectrum in the film. This changes primarily the lower cutoff ν_c in equation (3), to allow lower-frequency modes. Effective elastic coupling requires similar elastic moduli for medium 2 and 3. In typical experimental systems such as polystyrene on silicon, however, the elastic moduli differ by two orders of magnitude ($\approx 3\,\mathrm{GPa}$ and $\approx 100\,\mathrm{GPa}$ for polystyrene and silicon, respectively). Second, due to the finite difference between the acoustic impedances of the two media, $Z = \rho_i u_i$ (with the density ρ_i of medium i), a finite reflectivity $R_{23} = \left[(Z_1-Z_2)/(Z_1+Z_2)\right]^2 < 1$ between medium 2 and 3 has to be introduced. For polystyrene on silicon, $R_{23} \approx 0.7$. Since $R_{23} < 1$, p_ac should be somewhat reduced in this case, but remain comparable to p_dis.

For dewetting experiments employing polymer films, the frequency-dependent rheology of macromolecular melts has to be taken into account. Above (but close to the) glass transition temperature, polymer melts are viscous liquids in the zero-frequency limit, but are glassy at the the relevant phonon frequencies [22]. The density of states for glasses deviates from the Debye theory, showing in the lower THz region a positive anomaly ("boson" peak) [23]. However, the cutoff frequency ν_c lies below this range and the boson peak only changes the constant p_0' in equation (3) [24].

The dissipation of phonons takes place in the form of phonon absorption and re-emission, possibly with a different direction and frequency. However, the density of states is on the average always and everywhere fulfilled, based on the principle of detailed balance in thermal equilibrium. In general, the finite reflectivities are frequency dependent [25]. Below several hundred GHz, phonons behave acoustically and exert a large radiation pressure, while above this frequency range diffusive behavior is observed associated with low reflectivities. Thus, thermal equilibration of the film is ensured by the high-frequency phonons. The effect described here is, however, caused by the exclusion of a part of the low-frequency spectrum where the acoustic behavior dominates.

3 Discussion

It is instructive to predict the effect of the acoustic disjoining pressure in systems that can be studied experimentally. As a first example, we consider polystyrene (PS) films on a glass substrate, at $T = 150\,^\circ\mathrm{C}$. In Figure 1, the predicted wavelength of a dewetting instability ($\lambda = 2\pi/q_m$) is plotted *versus* the film thickness. The dashed line is based on vdW forces (p_dis) only, while the solid line contains contributions of both vdW and acoustic forces ($p_\mathrm{dis} + p_\mathrm{ac}$). Since the range in film thicknesses is larger than the crossover length d_c, we use an empirical expression for the derivative of the disjoining pressure $\partial_d p_\mathrm{dis}$:

$$\partial_d p_\mathrm{dis} = \frac{1}{2}\left[\left(\partial_d p_\mathrm{nr} + \partial_d p_\mathrm{r}\right) - \left(\partial_d p_\mathrm{nr} - \partial_d p_\mathrm{r}\right)\tanh\left(\frac{d-d_\mathrm{c}}{w}\right)\right], \qquad (11)$$

where $p_\mathrm{nr} = A_{123}/6\pi d^3$ and $p_\mathrm{r} = B_{123}/d^4$ for non-retarded and retarded disjoining pressures, respectively. With $d_\mathrm{c} = 43\,\mathrm{nm}$ and $w = 20\,\mathrm{nm}$, the crossover from non-retarded to retarded vdW forces causes a rapid increase in λ for $d > 40\,\mathrm{nm}$. The added contribution of p_ac has two

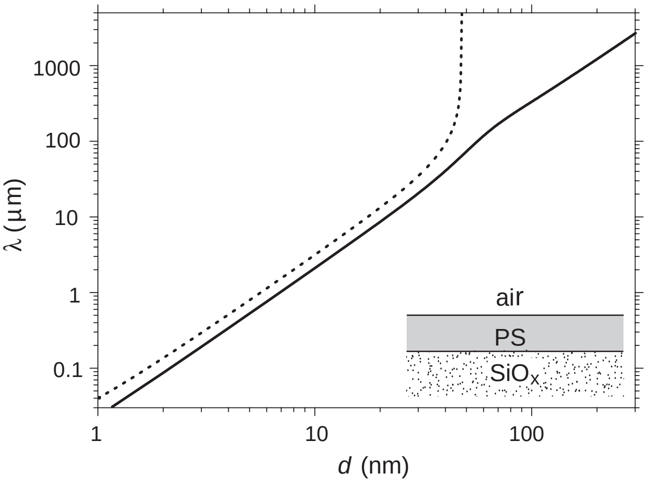

Fig. 1. Calculated instability wavelength λ *vs.* film thickness d for PS ($\gamma = 30\,\mathrm{mN/m}$) on glass, for $T = 150\,^\circ\mathrm{C}$. The dashed line is the contribution of vdW forces, exhibiting a crossover from the destabilizing non-retarded ($A = 1.8 \cdot 10^{-20}$ J [19]) to the stabilizing retarded ($B = -2.2 \cdot 10^{-29}$ Jm [8,26]) regime, for $d_\mathrm{c} = 43\,\mathrm{nm}$ and $w = 20\,\mathrm{nm}$ in equation (11). The solid line contains both p_dis and p_ac terms (Eq. (7)).

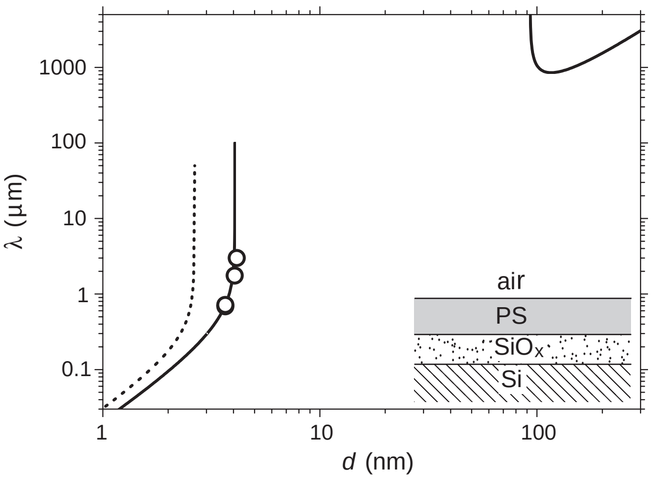

Fig. 2. Instability wavelength λ *vs.* film thickness d for PS ($\gamma = 30.8$ mN/m) on a SiO$_x$ covered Si substrate (see inset). The circles are data from [13]. The dashed line is a prediction (Eq. (7)) based exclusively on non-retarded vdW forces (Eq. (12), with $A_{\mathrm{SiO}_x} = 1.8 \cdot 10^{-20}$ J, $A_\mathrm{Si} = -2.2 \cdot 10^{-19}$ J [13]). The solid line takes both p_dis (Eq. (11)) and p_ac into account. For values of $d > 100\,\mathrm{nm}$, p_ac dominates over the much weaker retarded vdW forces ($B_\mathrm{Si} = -7.1 \cdot 10^{-29}$ Jm, $d_\mathrm{c} = 8\,\mathrm{nm}$, $w = 1\,\mathrm{nm}$), leading to a re-entrant instability (upper right corner).

consequences: 1) The instability wavelength is lower. Data analysis considering only p_dis would result in a "Hamaker constant" that is too high. 2) If both p_dis and p_ac are taken into account, the crossover behavior seen for vdW forces is suppressed, since the effect of p_dis is negligible for large values of d. While using a pure vdW model may lead to a misinterpretation of the data (*i.e.* the non-retarded vdW forces seemingly extend to very large values of d), the crossover to retarded vdW forces should provide a criterion, by which the effect of the acoustic disjoining pressure can be detected in an experiment.

As a second example, we re-analyze the data from Seemann *et al.* [13] (Fig. 2). They studied the stability of PS films on a Si wafer covered by a $\ell = 2.4\,\mathrm{nm}$ thick oxide layer. The two-layer sandwich (inset in Fig. 2) requires a correction of the non-retarded disjoining pressure:

$$p_\mathrm{nr} = \frac{A_{\mathrm{SiO}_x}}{6\pi d^3} - \frac{A_{\mathrm{SiO}_x} - A_\mathrm{Si}}{6\pi (d+\ell)^3}\,, \qquad (12)$$

where A_{SiO_x} and A_Si are the A_{123} Hamaker constants for SiO$_x$ and Si substrates, respectively. The dashed and solid lines in Figure 2 are the predictions from equation (11), with p_nr from equation (12) and a value of $B_{123} = B_\mathrm{Si} = -7.1 \cdot 10^{-29}$ Jm [8,26].

Since the Hamaker constants of Si and SiO$_x$ have opposite signs, a spinodal instability is observed only for thin films. While the vdW theory predicts a stabilization of the film (*i.e.* a divergence of λ, dashed line in Fig. 2), the experimental divergence lies at a value of d, which is too large. The prediction of equation (7) on the other hand (excluding the higher-order effects described above), is a better description of the experimental data. For this system, an interesting prediction can be made. If the film thickness is larger than 100 nm a re-entrant instability should occur. There, the stabilizing effect of the vdW forces becomes negligible, leading to a dominance of p_ac in equation (7). While re-entrant film destabilization may not be measurable in this specific experimental system (the film will break up due to heterogeneous nucleation first [13]), it should in principle be experimentally accessible, providing a further possibility to verify our theory.

While attractive forces stemming from a geometric confinement of a fluctuating medium are generic and apply to any fluctuating field, the range of these forces is related to the correlation length of the fluctuations [4]. In the case of simple liquids, where the mean free path of phonons is short ($\sim 1\,$Å), attractive forces stemming from the exclusion of part of the phonon spectrum should be limited to extremely thin films and are therefore negligible. The glassy behavior of polymer melts at high frequencies [22], however, leads to much larger phonon mean-free-path lengths ($\sim 1\,\mu$m) [27] and therefore to a measurable acoustic disjoining pressure in dewetting experiments that employ $\sim 100\,\mathrm{nm}$ thick polymer films.

The only liquid thin film system other than polymer melts, in which the acoustic disjoining pressure could also play a role, is liquid helium. Only in cryogenic liquids is the phonon correlation length larger than a molecular dimension. The main difference to polymer melts is, however, the role of the temperature. At $T \approx 0\,\mathrm{K}$, phonons (and therefore p_ac) obey quantum statistics, while for $T > \Theta_\mathrm{D}$ $p_\mathrm{ac} \propto kT$. At very low temperatures (as considered by Dzyaloshinskii and co-workers [8]) $p_\mathrm{ac} \ll p_\mathrm{dis}$ because of the quantum statistic where $p_\mathrm{ac} \propto hu$ (see above).

In conclusion, we have shown that forces in thin polymer films stemming from non-electromagnetic fluctuations are predicted to contribute equally to film instabilities as the EM dispersion forces and have the same scaling behavior (compared to the non-retarded EM case). Under certain conditions, the acoustic disjoining pressure may even dominate the EM analog.

Useful discussions with R. Blossey and M. Morariu are gratefully acknowledged. This work was partially funded by the Deutsche Forschungs Gemeinschaft (DFG) through the Sonderforschungsbereich 513 and the Dutch "Stichting voor Fundamenteel Onderzoek der Materie" (FOM).

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