Generalized Casimir forces in non-equilibrium systems

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Abstract. – In the present work we propose a method to determine fluctuation induced forces in non equilibrium systems. These forces are the analogue of the well known Casimir forces, which were originally introduced in Quantum Field theory and later extended to the area of Critical Phenomena. The procedure starts from the observation that many non equilibrium systems exhibit long-range correlations and the associated structure factors diverge in the long wavelength limit. The introduction of external bodies into such systems in general modifies the spectrum of these fluctuations and leads to the appearance of a net force between these bodies. The mechanism is illustrated by means of a simple example: a reaction diffusion equation with random noises.

Introduction. – In 1948 Casimir \textsuperscript{[1]} noticed that, since in the vacuum the quantum electromagnetic field fluctuates rather than vanishing as in the classical case, it would cost some energy to introduce a macroscopic body into it. Indeed, the presence of a macroscopic body limits the allowed wave-vectors and therefore the energy density. Casimir derived a simple expression for the resulting force per unit area acting between two large conducting neutral plates facing each other at distance \( x \). The result is: \( F/A = -\frac{\hbar c \pi^2}{240 x^4} \) and the presence of \( \hbar c \) comes from the quantal origin of the force. In the last twenty years, Casimir’s idea has been extended to some equilibrium systems where fluctuations are of thermal origin, rather than quantal and thus the force is proportional to \( k_B T \) \textsuperscript{[2]}. The unifying feature of systems displaying thermal Casimir forces is the presence of long ranged spatial correlations. The best known case are binary fluid mixtures at the critical point \textsuperscript{[3]}, where the Casimir force between two plates of area \( A \) and separation \( x \) is attractive and proportional to \(-k_B T A/x^3\). The inverse power law character of the force is originated by the divergence of the correlation length, \( \xi \), as the critical point is approached. The Ornstein-Zernike theory \textsuperscript{[4]} predicts long range density fluctuations characterized by a structure factor (defined as the Fourier transform}
of the density-density correlation function) like:

\[ S(k) \propto \frac{1}{\xi^2 + k^2}, \]  

(1)

where \( \xi \propto |T - T_c|^{-1/2} \). Then the structure factor behaves as \( S(k) \propto k^{-2} \) at the critical point and the free energy of the system scales as \( k_B T A/\ell^{d-1} \), from which one derives the above expression for the force.

Casimir forces also appear in equilibrium physical systems where long range correlations appear because a continuous symmetry is broken [5]. For instance in liquid crystals, with a vector order parameter, the structure factor for a smectic-A phase layered in the \( z \)-direction is anisotropic:

\[ S(k) \propto k_\perp^2 + \lambda^2 k_z^2. \]  

(2)

When the \( \mathbf{k} \) is parallel to the layers, i.e. \( k_z = 0 \), the structure factor diverges as \( k^{-2} \). Such smectic liquid crystals also exhibit a Casimir force: \( F \propto -k_B T A/\ell^2 \) [6]. Similar long ranged correlations appear also in nematic phases of liquid crystals, or magnetic liquid crystals immersed in a magnetic field \( H \) [7], in superfluid films [8], in solutions of ionic salts or micellar systems [9] among many others.

Although there seems to exist a relation between the presence of long range correlations and Casimir forces, this relation has not been elucidated when computing the force. In practice, Casimir forces are obtained by differentiating with respect to the typical distance the free energy change [2] induced by the confinement of the fluctuating field.

The goal of this Letter is to establish a general relation between correlation functions (or the associated structure factors) and Casimir forces. We also provide a novel method to compute the Casimir force, specially useful for non-equilibrium systems where long range correlations are ubiquitous [10], but arguments based on the free energy cannot be invoked.

In a previous work [11] we derived the Casimir force between large intruders in a granular system, starting from the structure factors. In that case the balance between the energy dissipation due to collisions and the energy injection, renders the system statistically stationary, but out of equilibrium. As a consequence it develops long range correlations, and the structure factors for density, velocity and temperature, as well as the cross structure functions, decay as \( k^{-2} \) [12].

There are a variety of other non-equilibrium systems for which the structure factors have been obtained. A major class is represented by systems under \emph{spatial gradients}, such as an isothermal uniformly sheared fluid in the \( z \)-direction (plane Couette flow) [13, 14]. It was found that the structure factor in the limit \( k \to 0 \) behaves as \( S(k) \propto -\gamma_0 \frac{k_z}{k_\perp^2 + \lambda^2 k_z^2} \), where \( \gamma_0 \) is the shear rate. In a similar fashion, a binary fluid in a Rayleigh-Bénard cell with a temperature gradient \( \nabla T \) below the onset of the instability, shows an enhancement of the structure factors proportional to \( (\nabla T)^2/k^4 \). Here the fluctuations are measured in the \( xy \)-plane, orthogonal to the gradient of the temperature [15]. In the same Rayleigh-Bénard cell, but heated from above, Li et al. showed that the concentration fluctuations in a polymer solution behave as \( (\nabla c)^2/k^4 \), where \( \nabla c \) is the concentration gradient created by the Soret effect. Pure concentration gradient were studied by Spohn [16] using lattice gases, who showed that the structure factors behave like \( (\nabla c)^2/(k^2 + k_0^2) \), with \( k_0 = \pi/(2L) \), being \( L \) the system size.

Another class of non-equilibrium systems exhibiting long range correlations are those lacking the \emph{detailed balance condition}. For instance, it has been shown [17, 18] that systems described by the Langevin equation with conservative dynamics and non-conservative noise
have long range spatial and temporal correlations. Analogously, non-equilibrium concentration fluctuations in reaction diffusion systems can be long ranged under certain conditions [19] that will be described in the next section.

The mesoscopic model. – As an example of how a Casimir force arises in a non-equilibrium system with long range correlations, we consider the simple case of a reaction-diffusion system in three dimensions, where the fluctuating density \( n \) around the homogeneous reference density \( n_0 \) obeys the equation

\[
\frac{\partial \phi}{\partial t} = \nabla \cdot (D \nabla \phi + \xi_1) - \lambda \phi + \xi_2, \tag{3}
\]

where \( \phi = n - n_0 \) is the fluctuating field, \( D \) is the diffusion coefficient, and \( \lambda > 0 \) is the relaxation rate. The terms \( \xi_1 \) and \( \xi_2 \) describe fluctuations in the diffusive flux and in the reaction rate. They are assumed to have white noise spectrum

\[
\langle \xi_1(r, t) \xi_1(r', t') \rangle = \Gamma_1 \delta_{i,k} \delta(r - r') \delta(t - t'),
\]

\[
\langle \xi_2(r, t) \xi_2(r', t') \rangle = \Gamma_2 \delta(r - r') \delta(t - t') \tag{4}
\]

where \( \Gamma_1 \) and \( \Gamma_2 \) are the noise intensities. This reaction-diffusion equation models, for instance, the set of chemical reactions:

\[
A \xrightarrow{k_1} B, \quad A + B \xrightarrow{k_2} 2A,
\]

that do not satisfy the detailed balance condition [20]. In this case, \( \phi \) is the density fluctuation of either the species \( A \) or \( B \) relative to its average stationary density and \( \lambda = n_0 k_2 - k_1 \). When \( \lambda = 0 \) the reaction shows a critical point at density \( n_0 = k_1/k_2 \). We assume that the pressure in the system is a function of the local density \( p(n) \).

Equation (3) not only models the chemical reaction described above, but governs many other physical systems, like liquid crystals [21] or superfluid films [2]. All these systems possess structure factors similar to those mentioned in the Introduction and the results obtained in this Letter can be generalized to those systems.

The solution of Eq. (3) predicts that, after an initial transient, the density \( \phi \) is statistically homogeneous and stationary so that in Fourier space we have:

\[
\langle \phi_k \rangle = 0, \quad \langle \phi_k \phi_q \rangle = V S(k) \delta_{k, -q} \tag{5}
\]

where the symbol \( \langle \cdot \rangle \) represents the average over the two noises \( \xi_1 \) and \( \xi_2 \). The structure factor \( S(k) \) is given by (see e.g. Chap. (8.3) of [19]):

\[
S(k) = \frac{\Gamma_1 k^2 + \Gamma_2}{2(Dk^2 + \lambda)} = \frac{\Gamma_1}{2D} + \frac{\Gamma/2D}{k^2 + k_0^2} \tag{6}
\]

with \( \Gamma = \Gamma_2 - \Gamma_1 \lambda/D \) and \( k_0 = \sqrt{\lambda/D} \). The corresponding real space correlation reads

\[
G(r) = \frac{\Gamma_1}{2D} \delta(r) + \frac{\Gamma}{2D} e^{-k_0 r} \tag{7}
\]

The second contribution, stemming from the \( k \)-dependent term in (6), represents fluctuations with a correlation length that depends on the reaction parameter, \( \lambda \), and, therefore, are of macroscopic size. In particular, near the critical point \( \lambda \to 0 \), the correlation length diverges. If the reaction satisfies the fluctuation-dissipation theorem [19,22] then \( \Gamma_1 = 2k_B T D \) and \( \Gamma_2 = 2k_B T \lambda \), where \( T \) is the temperature, implying that \( \Gamma \) vanishes along with the macroscopic correlations. On the contrary, in non-equilibrium systems which violate the
fluctuation-dissipation theorem, $\Gamma$ does not vanish and macroscopic correlations are present. The $\delta$-term in Eq. (7), coming from $\Gamma_1/2D$, describes the microscopic self-correlation of the particles that a mesoscopic model cannot resolve. Therefore, they can be eliminated. This corresponds to subtracting the asymptotic value of $S$ for large values of $k$. From now on, we will consider the macroscopic part (or, equivalently, the non-equilibrium part) of the structure factor $S^\ast(k) = S(k) - \lim_{q \to \infty} S(q) = \Gamma/[2D(k^2 + k_0^2)]$. This is equivalent to suppress the vectorial noise $\xi \ast$ and keep only a scalar noise $\xi$ with an intensity $\Gamma$.

We study now the effect of confining the system between two plates, parallel and infinite in the $y$ and $z$-directions, located at $x = 0$ and $x = L$, and calculate the force between the plates surrounded by a fluid described by Eq. (3). The force derives from the pressure, $p(n)$, exerted by the particles over the plates. To proceed, we consider the system in a volume $L_x \times L_y \times L_z$, periodic in all directions. In this volume we place two plates at distance $L$ with non flux boundary conditions at them, as natural for a reacting system. The total volume $V$ results divided into two regions: Region I between the plates of volume $L \times L_y \times L_z$, and Region II outside the plates of volume $(L_x - L) \times L_y \times L_z$. The limit $L_x, L_y, L_z \to \infty$ will eventually be taken.

In order to perform the analysis in the two regions let us consider a case of a general volume $V = X \times L_y \times L_z$, where $X = L$ for Region I and $X = L_x - L$ for Region II. The density field is expanded, taking into account the non flux boundary conditions on the $x$-direction, as

$$\phi(r, t) = V^{-1} \sum_{k_x} \sum_{k_y} \sum_{k_z} \phi_k(t) \cos(k_x x) e^{ik_y y} e^{ik_z z}$$

where $k_x = \pi n_x / X$, $k_y = 2\pi n_y / L_y$, $k_z = 2\pi n_z / L_z$, $n_x = 0, 1, 2, \ldots$ and $n_y, n_z = \ldots, -1, 0, 1, \ldots$. The noise $\xi$ is expanded in a similar way with

$$\langle \xi_k(t) \xi_q(t') \rangle = \gamma_{k_x} VT \delta_{k, q} \delta(t - t')$$

where $\delta_{k_x, q_x} \delta_{k_y, -q_y} \delta_{k_z, -q_z}$ is a modified 3D Kronecker delta. Moreover the factor $\gamma_{k_x}$ ($\gamma_{k_x} = 1/2$ if $k_x = 0$ and $\gamma_{k_x} = 1$ otherwise) appears because of the non-flux boundary condition in the $x$-direction. Replacing these expansions in (3) it is found that

$$\langle \phi_k \rangle = 0, \quad \langle \phi_k \phi_q \rangle = \gamma_{k_x} \delta_{k, q} V S^\ast(k)$$

with the same structure factor $S^\ast(k)$ as in the homogeneous case. Finally, the density field fluctuations in real space are given by:

$$\langle \phi(r) \rangle = 0, \quad \langle \phi(r)^2 \rangle = \frac{\Gamma}{2Dk_0^2} V^{-1} \sum_{q} \frac{1}{q^2 + 1} \cos(q_x k_0 x)^2.$$

where $q = k / k_0$ and the prime in the sum means that the term $q_x = 0$ has a factor 1/2.

The sum in Eq. (11) contains an ultraviolet divergence ($q \to \infty$). Therefore, in order to perform the summation a regularization prescription is needed. We introduce a regularizing kernel in Eq. (11) of the form $1/(1 + \epsilon^2 q^2)$, that equals 1 for $\epsilon \to 0$, limit that will be taken at the end of the calculations. The election of a rational function instead of an exponential one is made to keep the calculations as simple as possible. The technique of a regularizing kernel is equivalent to imposing a cutoff in the $q$-vectors of the order of $q_c \sim \epsilon^{-1}$ or to the zeta-function regularization method [21].

Next, we take the limit $L_y, L_z \to \infty$ allowing us to replace the sums on $q_y$ and $q_z$ by integrals that can be carried out, with the result:

$$\langle \phi(r)^2 \rangle = \frac{\Gamma}{8\pi DX} \frac{1}{1 - \epsilon^2} \sum_{q_x} \log \left( \frac{1 + \epsilon^2 q_x^2}{\epsilon^2(1 + q_x^2)} \right) \cos(q_x k_0 x)^2.$$
Figure 1 shows the density fluctuations (12) in the \( x \)-direction when the plates are located at \( x = 0 \) and \( k_0L = 1/2, \epsilon = 0.05 \) and \( L_x \to \infty \). The set of \( q_x \)-vectors entering in the sum of Eq. (12) for Regions I and II are different. In Region I, the allowed \( q_x \) vectors are quantized as: \( q_x = \pi n_x/(k_0L) \), while in Region II they form a continuum in the limit \( L_x \to \infty \). This difference produces a jump of \( \langle \phi^2 \rangle \) at the plate, that is shown in the inset.

**Casimir forces.** As shown in Fig. 1, the density fluctuations in a confined system depend on the position but also on the system size, and therefore are different in the region in between the plates and the region outside them. If the pressure is a function of the local density field \( p(n) \), these differences in the density fluctuations create an unbalance in pressure between both sides of each plate, and consequently a net force. To calculate the force, we expand the local pressure around the reference density \( n_0 \), and take the statistical average, finding that

\[
\langle p(x) \rangle = p(n_0) + \frac{1}{2} \left. \frac{\partial^2 p}{\partial n^2} \right|_{n_0} \langle \phi(x)^2 \rangle.
\]

The net force acting on the plate located at \( x = L \) is the difference between the pressure inside \( p(x \to L^-) \) and the pressure outside \( p(x \to L^+) \), that come from density fluctuations in Regions I and II respectively. Then the net effective force between the plates by unit area is twice the pressure difference in one of the plates:

\[
\frac{F}{A} = \frac{2}{16\pi D(1-\epsilon^2)} \left[ \frac{1}{L} \sum_{q_x=-\infty}^{\infty} \log \left( \frac{1+\epsilon^2 q_x^2}{\epsilon^2(1+q_x^2)} \right) - \frac{k_0}{\pi} \int_{-\infty}^{\infty} dq_x \log \left( \frac{1+\epsilon^2 q_x^2}{\epsilon^2(1+q_x^2)} \right) \right]
\]

The integral in this equation is simply \( 2\pi(\epsilon - 1)/\epsilon \), and Eq.(1.431,2) of [23], allows us to evaluate the sum. The diverging terms \( O(\epsilon^{-1}) \) cancel and the result for the force, in the limit of a vanishing cutoff \( \epsilon \), is surprisingly simple:

\[
\frac{F}{A} = F_0 \left( 1 - \frac{\log(2\sinh l)}{l} \right)
\]

where \( l = k_0L_x \) and \( F_0 = \Gamma k_0(\partial^2 p/\partial n^2)/(8\pi D) \). Let us note that the final expression of the Casimir force, is a universal function of the reduced distance, \( l = k_0L_x \). Moreover, there is
no dependence on the cutoff length, as the two divergences in the cutoff, one stemming from the discrete sum and another from the integral, exactly cancel each other. The regularizing kernel, a technique well known in the field of Casimir forces [24], has allowed us to obtain a finite result as a difference of two diverging quantities.

The analysis of the Casimir force, Eq. (15) can be performed in the limits of far plates \((l \gg 1)\) or near ones \((l \ll 1)\). In the first case, \((l \gg 1)\), implies that the distance \(L_x \gg k_0^{-1}\) and therefore the plates are outside the correlation length, \(k_0^{-1}\) (see Eq. (7)). Then, one expects a very fast decay of the Casimir forces. In the opposite limit \((l \ll 1)\), when the plates are well inside the correlation length, the force is much stronger. These forces are

\[
F^{\text{far}} / A = F_0 e^{-2l}, \quad F^{\text{near}} / A = -F_0 \frac{\log l}{l}.
\]

These results are presented in Fig. 2 where we plot the exact force Eq. (15) as a function of the dimensionless distance \(l\), together with the far and near plate approximations.

![Figure 2](image_url)

Fig. 2 – Dimensionless force as a function of the dimensionless distance \(l\), together with the asymptotic form for \(l \gg 1\) (dashed line) and \(l \ll 1\) (dotted line).

**Summary and Conclusions.** – We have calculated the fluctuation induced Casimir force between two parallel plates produced by a scalar field whose evolution is given by a reaction-diffusion equation. This system is out of equilibrium because the noise terms do not satisfy the fluctuation-dissipation relation, developing long range correlations. From these correlations one can calculate the pressure that the fluctuating field exerts on the two parallel plates.

The Casimir force obtained in this way depends on a macroscopic parameter \(k_0\) that defines a characteristic length as \(k_0^{-1}\), which is a combination of the diffusion constant and reaction rates, similar to the cases described in other physical systems [16, 20]. The calculated force exhibits a crossover at plate separation of the order of \(k_0^{-1}\), distance at which the correlation have decayed (see Eq. (7)). For short separations, the correlations play an important role and the force shows a strong distance dependence, while for \(L \gg k_0^{-1}\) the density correlations have decayed and the force vanishes exponentially.

Finally the force is repulsive if the pressure is such that \(\partial^2 p / \partial n^2 > 0\), as it is usually the case for molecular fluids far from the liquid-gas transition. However, the sign might change for different geometries of the obstacles (for instance plate-sphere, or two concentric spheres [25]).
To conclude, we have developed a method to calculate Casimir forces starting from the structure factors. This method is valid for both equilibrium and non-equilibrium systems, where the usual derivation of Casimir forces based on free energy calculations is not applicable. The necessary ingredients to obtain a Casimir force are a fluctuating field with correlations that extend over a macroscopic range, and a confinement of the fluctuation spectrum induced by the presence of the plates or other obstacles.

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