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Non-equilibrium fluctuation-induced interactions

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Abstract

We discuss non-equilibrium aspects of fluctuation-induced interactions. While the equilibrium behavior of such interactions has been extensively studied and is relatively well understood, the study of these interactions out of equilibrium is relatively new. We discuss recent results on the non-equilibrium behavior of systems whose dynamics is of the dissipative stochastic type and identify a number of outstanding problems concerning non-equilibrium fluctuation-induced interactions.

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1. Introduction: fluctuation-induced interactions

The most famous example of a fluctuation-induced interaction is the Casimir effect [1]. Here, the presence of two conducting plates of area A modifies the zero point energy of the electromagnetic field and leads to an attractive force via the celebrated formula

$$f = -\frac{A\pi^2 \hbar c}{240 L^4}, \quad (1)$$

where L is the separation between the plates. At non-zero temperature thermal fluctuations come into play, giving rise to additional temperature-dependent interactions. The full theory of the electromagnetic Casimir effect is most compactly expressed via the Lifshitz theory, which is formulated in terms of the general dielectric and conductive properties of the interacting bodies [2]. At the level of pairwise, or two body, interaction, the Lifshitz theory recovers the general theory of van der Waals or dispersion forces [2].

In 1978 Fisher and de Gennes predicted that a critical Casimir effect would exist in critical (or near-critical systems) [3], a prediction that has been recently experimentally confirmed [4]. This critical Casimir force arises because objects or surfaces in critical systems modify the thermal fluctuations of the order parameter. In such systems the interaction can be tuned by, for instance, chemical treatment of the surfaces so they either favor the same or differing phases, for example, in a binary critical fluid. If L is the distance between two plates immersed in a critical fluid, then

the free energy is a function of L and the average force between the two plates is given by

$$f = -\frac{\partial F}{\partial L}, \quad (2)$$

where $F = -T \ln(Z)$ and Z is the partition function for the system (T being the temperature in units where the Boltzmann constant is 1). If we consider a simple Gaussian scalar field theory with Hamiltonian

$$H = \frac{1}{2} \int d\mathbf{x} [(\nabla\phi)^2 + m^2\phi^2], \quad (3)$$

then we can compute the partition function $Z = \int d[\phi] \exp(-\beta H)$ by integrating over fields ϕ with the appropriate boundary conditions. For instance, the equilibrium force for parallel plates of area A imposing the Dirichlet–Dirichlet (DD) boundary conditions or Neumann–Neumann boundary conditions is attractive and given as [5]

$$f = -\frac{2AT}{(4\pi)^{\frac{d-1}{2}} \Gamma(\frac{d-1}{2})} \times \int k^{d-2} dk \frac{\sqrt{k^2 + m^2} \exp(-2L\sqrt{k^2 + m^2})}{1 - \exp(-2L\sqrt{k^2 + m^2})}, \quad (4)$$

where d denotes the dimension of space and $\Gamma(z)$ is the Euler gamma function. In agreement with the more general considerations of Fisher and de Gennes, we note that the force becomes long range when the Gaussian field theory is critical;

that is, when $m = 0$ we have

$$f = -\frac{AT\Gamma(d)\zeta(d)}{(16\pi)^{\frac{d-1}{2}}\Gamma(\frac{d-1}{2})L^d}, \quad (5)$$

where ζ is the Riemann zeta function. A wide range of Casimir-type interactions also occur in lipid membranes, where local membrane inclusions can couple to various physical parameters of the membrane, for instance, the curvature, lipid composition and lipid order parameters such as the lipid phase (solid–liquid–gel), the thickness of lipid bilayers or even lipid orientation. Similarly, regions of lipids of varying bending and elastic energies can experience effective interactions due to membrane height fluctuations; thus, membrane fluctuations can potentially modify the phase diagram of lipid bilayers composed of lipid mixtures [6]. Also, colloids and other objects in liquid crystals can couple to the local nematic order parameter and this leads to effective interactions between such objects [7].

Many of the examples of thermal fluctuation-induced interactions we have cited occur in soft matter systems, where time scales can be very long and dynamics slow. It is therefore natural to ask how fluctuation-induced interactions evolve with time toward their equilibrium value [8–10]. In addition, one can analyze a number of situations where one has non-equilibrium steady states: for instance, when the system is driven by external noise that does not obey the detailed balance necessary for thermal equilibrium [9–12]. Non-equilibrium Casimir effects have also been observed and predicted in a variety of driven granular systems [13, 14], as well as in systems undergoing chemical reactions [15]. The imposition of temperature [16] or other thermodynamic gradients in systems will also lead to modifications of fluctuation-induced interactions with respect to the equilibrium situation and may provide a means of tuning fluctuation-induced interactions.

2. Non-equilibrium situations: the problem of computing the force

Out of equilibrium we cannot derive forces as the derivatives of a free energy, and in general, the force will depend on the dynamics, non-equilibrium driving and/or gradients in the system. In order to determine the non-equilibrium force, we thus need an expression for the force in terms of the field configuration. There are basically three approaches to this problem:

- *The phenomenological approach.* In non-thermal systems such as granular materials and systems of reaction diffusion equations, one can define a local pressure based on local kinetic arguments. For instance, the local order parameter ϕ can be associated with a local density $\rho(\phi)$ and the local pressure defined via a local equation of state, for instance $P = T\rho$, if one uses the ideal gas approximation [15]. In granular materials, the equation of state for hard spheres has also been used [13].
- *The stress tensor.* The instantaneous force on a surface S in the direction i is given by

$$f_i = \int_S T_{ij} dS_j, \quad (6)$$

where T_{ij} is the stress tensor; this approach has been widely used in both equilibrium and non-equilibrium situations [8, 11, 12, 17].

- *The principle of virtual work.* Boundary conditions are effectively imposed by assuming that the surface has a coordinate L and a field ϕ effectively exists on the surface and interacts with the field via an interaction term [9, 10, 18], for instance,

$$H_{\text{int}} = \int d\mathbf{x} \delta(z - L)V(\phi), \quad (7)$$

for a surface on the plane $z = L$. The total energy is thus a function $H[\phi, L]$ of both the field ϕ and the coordinate L . The force conjugate to the parameter L is thus given by the principle of virtual work as $f = -\frac{\partial H}{\partial L}$. As an example, taking $V(\phi) = \lambda\phi^2/2$ and then taking the limit $\lambda \rightarrow \infty$ imposes Dirichlet boundary conditions on the surface at $z = L$.

It is straightforward to see that the principle of virtual work and stress tensor gives the same expressions for the average force in equilibrium. However, the computation of the force seems to require clarification of the field theory at a microscopic level; for instance, one needs to know how the field ϕ changes when the surface is displaced [10, 19]. In electrostatics, there is no ambiguity as the force acting on a volume V is given by $\mathbf{f} = \int_V d\mathbf{x} \rho \mathbf{E}$, where \mathbf{E} is the local electric field and ρ is the charge density [20]. From this expression, we can, in fact, derive the stress tensor expression for the force. In a binary mixture, the forces on the plates will be generated by the interaction between the molecules composing the plate and the molecules of the surrounding fluid; in a molecular dynamics simulation the force on the plates can thus be unambiguously computed. However, if we coarse grain the binary fluid and replace it with an Ising model, we immediately see that the numerical calculation of the instantaneous force for a given spin configuration is much less obvious.

3. The results for fluctuation-induced interactions governed by stochastic dissipative dynamics

Here we consider a free field theory with a general quadratic Hamiltonian

$$H = \frac{1}{2} \int d\mathbf{x} d\mathbf{x}' \phi(\mathbf{x}) \Delta(\mathbf{x}, \mathbf{x}', L) \phi(\mathbf{x}'), \quad (8)$$

and we will compute the force using the principle of virtual work discussed in the previous section. Here Δ is a self-adjoint operator, i.e. $\Delta(\mathbf{x}, \mathbf{x}') = \Delta(\mathbf{x}', \mathbf{x})$, and L represents any suitable free parameter in the problem, but for concreteness it could be the position of a plate that interacts with the field.

For concreteness we consider the problem where the system is prepared in a state $\phi = 0$ at the time $t = 0$ and then let it relax at some non-zero temperature T close to a critical point where it is massless. We also assume that the system is of infinite extent; that is to say the field exists outside the two plates. This means that the resulting force per unit area is a

disjoining pressure; it has no bulk term and thus tends to zero when the plate separation becomes large.

We assume that the field obeys general over-damped stochastic dynamics [21]

$$\begin{aligned}\frac{\partial \phi(\mathbf{x})}{\partial t} &= - \int d\mathbf{x}' R(\mathbf{x}, \mathbf{x}') \frac{\delta H}{\delta \phi(\mathbf{x}')} + \eta(\mathbf{x}, t) \\ &= - \int d\mathbf{x}' R \Delta(\mathbf{x}, \mathbf{x}') \phi(\mathbf{x}') + \eta(\mathbf{x}, t),\end{aligned}\quad (9)$$

where $R\Delta$ indicates the composed operator $R\Delta(\mathbf{x}, \mathbf{x}') = \int d\mathbf{y} R(\mathbf{x}, \mathbf{y}) \Delta(\mathbf{y}, \mathbf{x}')$. To satisfy detailed balance with noise that is uncorrelated in time, we chose the spatial noise correlator to be

$$\langle \eta(\mathbf{x}, t) \eta(\mathbf{x}', t') \rangle = 2T \delta(t - t') R(\mathbf{x}, \mathbf{x}'). \quad (10)$$

When $R(\mathbf{x}, \mathbf{x}') = \delta(\mathbf{x} - \mathbf{x}')$ we have non-conserved model A dynamics, and when $R(\mathbf{x}, \mathbf{x}') = -\nabla^2 \delta(\mathbf{x} - \mathbf{x}')$ we have conserved model B dynamics. It can be shown [9, 10] that the Laplace transform of the time-average-dependent force is given by

$$\mathcal{L}f(s) = \frac{T}{s} \frac{\partial}{\partial L} \ln(Z(\Delta_s)), \quad (11)$$

where the operator Δ_s is given by

$$\Delta_s = \Delta + \frac{s}{2} R^{-1} \quad (12)$$

and $Z(\Delta_s)$ is the partition function for a Gaussian field theory defined via the operator Δ_s . This result means that the Laplace transform of the time-dependent Casimir force can be expressed in terms of the static Casimir force for another free field theory, and if this static partition function is known the time-dependent force can be extracted by inverting the Laplace transform. The late time equilibrium result is recovered from the pole at $s = 0$ in equation (11). Note that in the case of model B dynamics, the operator Δ_s is non-local due to the term $R^{-1} = -\nabla^{-2}$. Thus, intriguingly, the study of the temporal evolution of fluctuation-induced forces can lead us to consider static Casimir forces arising from non-local field theories. In the case of model B dynamics, additional boundary conditions have to be specified for the flux of the field or its chemical potential at the interfaces [22]. This choice of boundary conditions still needs to be specified within the formalism described here, and the cases when it can be applied are yet to be worked out—the non-equilibrium evolution of the Casimir force under model B dynamics thus remains an open problem even for Gaussian field theories.

Let us consider the case of model A dynamics, i.e. where the dynamical operator $R(\mathbf{x} - \mathbf{x}') = \delta(\mathbf{x} - \mathbf{x}')$. This is the easiest case to analyze and it is the case that has been most studied in the literature via the other approaches mentioned above. We take H to be of the form of equation (3) in the critical case $m = 0$ and impose DD boundary conditions.

The Laplace transform can be almost inverted analytically, the temporal derivative of the out of equilibrium force is given by

$$\frac{df}{dt} = - \frac{2AT}{(8\pi)^{\frac{d}{2}} t^{\frac{d-1}{2}}} \frac{\partial}{\partial t} \sum_{n=1}^{\infty} \frac{1}{\sqrt{t}} \exp\left(-\frac{L^2 n^2}{2t}\right), \quad (13)$$

where A is the area of the plates. Using the Poisson summation formula, we find that

$$\begin{aligned}\frac{df}{dt} &= - \frac{AT}{2(8\pi)^{\frac{d}{2}} t^{\frac{d+2}{2}}} + \frac{AT}{2(8\pi)^{\frac{d-1}{2}} t^{\frac{d+1}{2}}} \\ &\quad \times \frac{\partial}{\partial L} \left[\sum_{n=1}^{\infty} \exp\left(-\frac{2\pi^2 n^2 t}{L^2}\right) \right].\end{aligned}\quad (14)$$

As model A dynamics is diffusive, we see that L^2 sets a time scale. The short-term behavior of the force as defined by the regime $t/L^2 \ll 1$ can be obtained from equation (13) and is given by

$$f(t) \sim - \frac{2AT}{(8\pi t)^{\frac{d}{2}}} \exp\left(-\frac{L^2}{2t}\right). \quad (15)$$

The late time, defined via $t/L^2 \gg 1$, decay to the equilibrium force f_{eq} can be extracted from equation (14) and is given by

$$f(t) \sim f_{\text{eq}} + \frac{AT}{d(8\pi)^{\frac{d}{2}} t^{\frac{d}{2}}}. \quad (16)$$

In equation (14), one sees the appearance of the eigenvalues for DD boundary conditions in the sum on the right-hand side. The first term can be seen to be due to the bulk on the exterior of the system. The results here also agree with those predicted using the stress tensor [8]; in the case of DD boundary conditions, the two approaches can be seen to be equivalent even out of equilibrium.

We now turn to a model of interacting dipoles which has the advantage of predicting a temporarily evolving Casimir, or van der Waals, force that can be, in principle, confronted with experiment and where, due to its electrostatic origin, the force can be precisely defined.

We consider a model dielectric medium [23] made up of local polarization fields $\mathbf{p}_v(\mathbf{x})$ at the point \mathbf{x} of the medium. Here v corresponds to a type or species of polarization field that notably has its own polarizability per unit volume denoted by $\chi_v(\mathbf{x})$. Next, we take two semi-infinite regions (slabs) V^+ and V^- defined via the sign of the coordinate z , such that $z > 0$ in V^+ and $z < 0$ in V^- . The two regions are separated in the z -direction by a distance L . The total energy for a given configuration of the dipole fields is

$$H = \frac{1}{2} \int d\mathbf{x} d\mathbf{y} \sum_{vv'} \mathbf{p}_v(\mathbf{x}) \cdot A_{vv'}(\mathbf{x}, \mathbf{y}, L) \mathbf{p}_{v'}(\mathbf{y}), \quad (17)$$

where

$$A_{vv'}(\mathbf{x}, \mathbf{y}, L) = \frac{\delta(\mathbf{x} - \mathbf{y}) I \delta_{vv'}}{\chi(\mathbf{x})} + D(\mathbf{x}, \mathbf{y}, L). \quad (18)$$

Here I is the identity matrix in three-dimensional space and the polarization energy equation (17) corresponds to the classical harmonic energy needed to generate a local polarization field. The second term D is the usual dipole-dipole interaction.

We assume that the dipole field dynamics is of the model A type

$$\frac{\partial p_{iv}(\mathbf{x})}{\partial t} = -\kappa_v(\mathbf{x}) \frac{\delta H}{\delta p_{iv}(\mathbf{x})} + \zeta_{iv}(\mathbf{x}, t), \quad (19)$$

where $\kappa_\nu(\mathbf{x})$ determines the local relaxation in the region \mathbf{x} of space for dipoles of type ν and $\zeta_{i\nu}(\mathbf{x}, t)$ is Gaussian white noise in time with spatial correlation function satisfying detailed balance. The choice of model A type dynamics is based on the observation that polarization is not a conserved quantity (i.e. it can be generated locally at a given point without the need for charge transport to that point). The parameters in the above model can then be related to the frequency-dependent dielectric function of the slabs via

$$\epsilon(\omega) = \epsilon_0 + \sum_\nu \frac{\chi_\nu}{1 + \frac{i\omega\chi_\nu}{\kappa_\nu}}; \quad (20)$$

this form of the dielectric function further justifies the choice of model A dynamics as it has the form of a superposition of the classical Debye expression for the dielectric function. We now consider the situation where two such dielectric slabs are brought suddenly into proximity, parallel and at distance L along their axes. The development of the van der Waals force between the two slabs is physically due to the onset of the correlation between the dipoles in the two slabs. It can be shown [23] that the time-dependent Casimir force is given by $f(t) = -\frac{\partial F(t, L)}{\partial L}$, where F is a time-dependent free energy whose Laplace transform is given by

$$\mathcal{L}F = -\frac{T}{s} \ln \left(Z \left[\epsilon \left(-i\frac{s}{2} \right) \right] \right), \quad (21)$$

where $Z[\epsilon(-i\frac{s}{2})]$ is the partition function for the static problem with

$$Z \left[\epsilon \left(-i\frac{s}{2} \right) \right] = \int d[\phi] \exp \left(-\frac{\beta}{2} \int d\mathbf{x} \epsilon \left(\mathbf{x}, -i\frac{s}{2} \right) [\nabla\phi(\mathbf{x})]^2 \right). \quad (22)$$

The static result, which is identical to that predicted by Lifshitz theory, is recovered from the pole at $s = 0$. The Laplace transform is, in general, difficult to invert; however, in the case of the parallel slab geometry, it can be shown that the force is given by

$$f(L, t, T) = -\frac{TA}{L^3} H(t), \quad (23)$$

where $H(t)$ is a time-dependent *Hamaker* constant that starts at zero and evolves to its static value as $t \rightarrow \infty$. We thus see that the scaling with distance L and time is completely different from that of model A dynamics for a massless scalar field, even though the static values have the same $1/L^3$ scaling. The behavior of $H(t)$ depends on the choice of dielectric function. For simple Debye-type dielectric functions, the decay to the equilibrium force is exponential; however, for certain dielectric functions [24], typical of glassy polymer-based materials, the decay can be of a power-law type [23].

4. Conclusions and open questions

We have seen that results on the out of equilibrium dynamics of fluctuation-induced forces are restricted to the simplest models of stochastic dynamics and that the field is replete with open questions and new directions for future study. To conclude this paper, I list a number of questions and challenges I believe would be interesting to address.

- *The definition of the out of equilibrium force.* There seem to be a number of unresolved issues as to how one can define the instantaneous force in out of equilibrium situations for coarse-grained models. As well as permitting the determination of out of equilibrium forces, such methods, especially numerical ones, could lead to better schemes for measuring Casimir forces in simulations, even in equilibrium. Notably, such new methods will be useful to determine the fluctuations of fluctuation-induced interactions [17].
- *Force dynamics for interacting field theories.* Even within the framework of simple dissipative stochastic dynamics, the only existing results on dynamics are for free or Gaussian field theories. The treatment of interacting field theories remains an open challenge. A problem that seems attackable is the problem of an N components ϕ^4 field theory in the limit $N \rightarrow \infty$ whose dynamics can be solved [26].
- *Beyond overdamped stochastic dynamics.* Clearly, the dynamics of binary liquid mixtures will be sensitive to hydrodynamic effects in most geometries and the coupling between hydrodynamics and the dynamics of the order parameter [21, 26] will change the temporal evolution of fluctuation-induced forces. Understanding the interplay between phase ordering in the presence of plates or inclusions and hydrodynamics is thus essential. Also it would be interesting, from a purely academic point of view, to see what effect the inclusion of inertial effects has on the evolution of fluctuation-induced forces; this should give the force propagation a wave-like component. Possibly, models for fluctuation-induced interactions in granular materials [13, 14] could incorporate inertial effects.
- *Non-equilibrium steady states.* One of the most active fields in the quantum electrodynamic Casimir effect is the study of forces between objects held at different temperatures [25]. In the realm of classical fluctuation-induced interactions, less work has been done. It can be shown that in the parallel plate geometry, if the three regions (outside left, inside and outside right) are held at different temperatures then the effective interaction is dominated by a bulk-like term [16] that, is mathematically similar in origin to the bulk difference in radiation pressure between regions of space at different temperatures. In soft matter systems, the absence of vacuum makes it natural to study the effect of temperature gradients, imposed for instance by holding the plates at different temperatures. The interacting dipole model presented in the previous section presents an interesting test bed for exploring the effects of temperature gradients and may indeed be realistic enough to make experimentally verifiable predictions.

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