

Violation of the action-reaction principle and self-forces induced by nonequilibrium fluctuations

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We show that the extension of Casimir-like forces to fluctuating fluids driven out of equilibrium can exhibit two interrelated phenomena forbidden at equilibrium: self-forces can be induced on single asymmetric objects and the action-reaction principle between two objects can be violated. These effects originate in asymmetric restrictions imposed by the objects' boundaries on the fluid's fluctuations. They are not ruled out by the second law of thermodynamics since the fluid is in a nonequilibrium state. Considering a simple reaction-diffusion model for the fluid, we explicitly calculate the self-force induced on a deformed circle. We also show that the action-reaction principle does not apply for the internal Casimir forces between a circle and a plate. Their sum, instead of vanishing, provides the self-force on the circle-plate assembly.

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In his pioneering work, Casimir [1] showed that two metallic plates in the electromagnetic field of vacuum attract one another due to the restriction they impose on the quantum fluctuations of the field. Fluctuation-induced forces exerting between macroscopic objects have since been exhibited in a vast variety of other systems at equilibrium [2], such as critical fluids and crystal liquids in the nematic phase, in which thermal fluctuations of long range can develop [3,4]. These forces are thought to play an important role in the stability of equilibrium phases of mesoscopic particles embedded into complex fluids. While the theoretical achievements in this field are numerous, a direct measurement of the Casimir force in critical fluids has only been obtained recently [5].

Matter in nonequilibrium steady states also develops fluctuations that are generically of large amplitude and have long correlation lengths [6,7]. By analogy to the equilibrium situation, one expects these fluctuations to induce similar forces that may be responsible for the aggregation and/or segregation mechanisms observed in fluids driven out of equilibrium [8–10]. However, the calculation of these forces cannot rely on the derivation of an (equilibrium) thermodynamic potential. It is only recently that these forces have been obtained between two planar objects immersed into nonequilibrium driven systems [11], granular fluids [12], or reaction-diffusion systems that violate the detailed balance [13].

Whether at or out of equilibrium, accurate experimental measurements of fluctuation-induced forces need to go beyond the idealized geometry of infinitely long plates, predominant in theory for its simplicity. Although a long-studied topic, the proper account of the geometry dependence of Casimir forces is a notoriously difficult problem when dealing with nontrivial geometries [14]. To date, the most widely used technique by experimentalists [5] relies on the so-called Derjaguin construction [15] (proximity force approximation), which in essence integrates the two-plate expression of the force along the curved surfaces.

In this Rapid Communication, we show that when restricted by nonplanar objects, nonequilibrium fluctuations can lead to additional effects not possible in equilibrium systems. Namely, nonvanishing forces can be induced on single asymmetric obstacles and the action-reaction principle between two intruders can be violated. These phenomena can have significant consequences in experiments, as they lead to

directed motion and unevenness in the measures of the forces between two objects. Furthermore, an unbalance of action and reaction would impede the use of the Derjaguin approximation where it would normally be valid at equilibrium. We found no mention of these facts in the literature.

Since nonequilibrium systems are thermodynamically open, it is not entirely unexpected that self-forces can appear. In fact, provided that both the microscopic time-reversibility and space rotation-invariance symmetries are broken, such forces have been implicitly suggested by the occurrence of sustained motions in other nonequilibrium contexts, such as in ratchets [16], Brownian motors [16,17], molecular motors [18], or the adiabatic piston [19]. In these systems, the space asymmetry usually lies in an external temperature gradient or in an anisotropic field exerting on the object. More recently, however, the directed motion of an asymmetric object immersed into vibrated granular matter has been exhibited [20]. The direct calculation of self-forces that we present here allows for a better understanding of the different effects at play in such motions. It also makes possible the evaluation of additional stresses exerting on asymmetrical structures in microdevices and could be used in tailoring mechanisms for the self-assembly of ordered structures. The violation of the action-reaction principle between two intruders directly results from the presence of self-forces. It does not seem to be systematic, however, even in asymmetric setups [21]. Note that it prevents the two-body forces from being derived from an effective potential, in contrast to equilibrium cases. Let us add that a violation of Newton's third law has also been noted in depletion forces between identical spheres in a flowing fluid [22].

For illustration purposes, we exhibit here these effects with the rather simplified nonequilibrium fluid that has been used in [13] in a planar geometry. The applicability of the model to nontrivial geometries can be greatly facilitated by devising an adequate Green-function formalism. We then show that the self-force exerting on a deformed circle is indeed nonzero at second order in the radius perturbation when dipolar deformations are considered. We also calculate the internal forces between a circle and a plate in an asymptotic regime where their separation is large and the circle's radius is small in comparison to the correlation length. In this situation, the action-reaction principle is not satisfied as the circle-plate assembly experiences a net self-force.

The fluctuating medium is described by a reaction-diffusion fluid, whose nonequilibrium steady state is achieved by violating the detailed balance [23]. The local density $\rho(\mathbf{r}, t)$ of the fluid fluctuates around a homogeneous reference density $\rho_0 \equiv \langle \rho(\mathbf{r}, t) \rangle$, where $\langle \cdot \rangle$ is a stochastic average. The density deviation $\Phi(\mathbf{r}, t) \equiv \rho(\mathbf{r}, t) - \rho_0$ is assumed to satisfy, at the mesoscopic scale, the stochastic reaction-diffusion equation

$$\frac{\partial \Phi}{\partial t} = (D\nabla^2 - \gamma)\Phi + \xi, \quad (1)$$

where D is a diffusion constant, γ is the reaction rate that drives the system to local equilibrium, and $\xi(\mathbf{r}, t)$ is a random white noise of correlation intensity Γ that takes into account the fluctuations on the reaction rates. Equation (1) primarily describes density fluctuations in a fluid with two reacting and diffusing chemical components (see [23]), but other nonequilibrium systems in their steady state are described by this model. The steady-state fluctuations are characterized by the bulk correlation length $\kappa^{-1} \equiv (\gamma/D)^{-1/2}$, which can be chosen as the mesoscopic scale.

Static objects immersed in the fluid prevent any flow of matter across their surface. Equation (1) is thus supplemented by the nonflux condition $\mathbf{n} \cdot \nabla \Phi = 0$ at the objects' surface, where $\mathbf{n}(\mathbf{r})$ is a unit normal vector pointing outward from the fluid's domain. In a steady state, one expects the pressure p of the fluid to be related to the density by a local equation of state $p = p(\rho(\mathbf{r}, t))$. This relation is experimentally measured in a number of cases of interest, like in driven granular media [24], for example. Here we only assume that it is expandable around the reference density ρ_0 and that density fluctuations stay small. The average pressure is thus modified by the fluctuations according to $\langle p \rangle = p_0 + \frac{p_0''}{2} \langle \Phi^2 \rangle$, where $p_0 = p(\rho_0)$ and $p_0'' = \frac{\partial^2 p}{\partial \rho^2}(\rho_0)$. The total force \mathbf{F}_S exerted by the fluid on an immersed object S results from summing this local pressure on every element $d\sigma$ of its surface. Since p_0 is a homogeneous pressure, it does not induce a force and one has

$$\mathbf{F}_S = \frac{p_0''}{2} \int_S d\sigma \mathbf{n} \langle \Phi^2 \rangle. \quad (2)$$

Starting from a dynamical model like (1), Casimir forces can be obtained by using Green functions [4]. After an initial transient of characteristic time of order $O(\gamma^{-1})$, the stationary solution of (1) is

$$\Phi_{st}(\mathbf{r}, t) = \int dt' \int_{\Omega} d\mathbf{r}' G(\mathbf{r}, \mathbf{r}', Dt - Dt') \xi(\mathbf{r}', t'), \quad (3)$$

where Ω is the domain occupied by the fluid.

It can be established that the temporal Fourier transform of the Green function $G(\mathbf{r}, \mathbf{r}', \omega) = \int d\tau e^{i\omega\tau} G(\mathbf{r}, \mathbf{r}', \tau)$ with $\tau = Dt$, is, up to a factor, the static structure factor of the fluid that enters into the force (2) when evaluated at $\omega=0$:

$$\begin{aligned} \langle \Phi_{st}(\mathbf{r}, t) \Phi_{st}(\mathbf{r}', t) \rangle &= \frac{\Gamma}{D} \int \frac{d\omega}{2\pi} \int_{\Omega} d\mathbf{r}'' G(\mathbf{r}, \mathbf{r}'', \omega) G(\mathbf{r}'', \mathbf{r}', -\omega) \\ &= \frac{\Gamma}{2D} G(\mathbf{r}, \mathbf{r}', \omega=0). \end{aligned} \quad (4)$$

The first equality in (4) follows directly from (3) and the convolution theorem. The second results from the differential equation satisfied by $G(\mathbf{r}, \mathbf{r}', \omega)$ (see details in [25]):

$$(-\nabla^2 + \kappa^2 - i\omega)G(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}'), \quad (5)$$

$$\mathbf{n}(\mathbf{r}) \cdot \nabla G(\mathbf{r}, \mathbf{r}', \omega)|_{\mathbf{r} \in S} = 0 \quad \forall \mathbf{r}' \in \Omega, \forall \omega. \quad (6)$$

In view of (4) one can omit any further reference to ω and calculate $G(\mathbf{r}, \mathbf{r}')$ as the solution of (5) and (6) in which ω is set to 0 right away. This is an appreciable simplification; \mathbf{F}_S only depends linearly on the static Green function.

To deal with the difficulties brought about by nonplanar objects in Casimir forces, a natural way is to use multiple-scattering techniques [14]. If G_0 is the free-space (unconstrained fluid) Green function, (5) and (6) are also equivalent to [25]

$$G(\mathbf{r}, \mathbf{r}') = G_0(\mathbf{r} - \mathbf{r}') - \int_S d\sigma_1 G(\mathbf{r}, \mathbf{r}_1) \mathbf{n}_1 \cdot \nabla_1 G_0(\mathbf{r}_1 - \mathbf{r}'), \quad (7)$$

which we abbreviate as $G = G_0 + G \star \nabla G_0$. The recursive iteration of this integral equation expands G as a series of multiple scatterings of G_0 on the surface S . In three dimensions, $G_0(\mathbf{r}) = \exp\{-\kappa|\mathbf{r}|\}/4\pi|\mathbf{r}|$, and in two dimensions, $G_0(\mathbf{r}) = K_0(\kappa|\mathbf{r}|)/2\pi$, where K_0 is the modified Bessel function of order 0. As is obvious from (7) and the above expressions for G_0 , the problem of calculating the force (2) as it stands is ill defined: short-range divergences appear. They are due to the inaccuracy of the continuous model on the microscopic scale [13]. A ‘‘bulk’’ divergence occurs when evaluating G at a same point \mathbf{r} , as well as a ‘‘wall’’ divergence once this point is approached to a surface. The first divergence is trivial to remove: it is independent of the immersed objects and thus consists in a homogeneous (although infinite) pressure unable to produce a force. The wall divergence plays an important role: it originates in the boundary condition imposed on $G(\mathbf{r}, \mathbf{r}')$ by the immersed objects and it is integrated all along their surface in calculating the force. The issue is then to understand how this integrated divergence compensates itself between different sides of the objects to yield a finite result. This compensation does not occur for any shape. To illustrate this, we consider the Green function G_p of a fluid restricted to a half space by a plate. The condition (6) on the plate can be replaced by the addition of an ‘‘image’’ source $\delta(\mathbf{r} - \mathbf{r}'^*)$ on the right-hand side (rhs) of (5), where \mathbf{r}'^* is the point symmetric to \mathbf{r}' with respect to the plate. The solution hence reads $G_p(\mathbf{r} - \mathbf{r}') = G_0(\mathbf{r} - \mathbf{r}') + G_0(\mathbf{r} - \mathbf{r}'^*)$. It is clear that evaluating $(G_p - G_0)(\mathbf{r}, \mathbf{r})$ [having subtracted the bulk divergence $G_0(\mathbf{r} - \mathbf{r}')$ as $\mathbf{r}' \rightarrow \mathbf{r}$] at the plate's surface produces a divergent collapse of \mathbf{r} and its image \mathbf{r}^* . For a smooth surface, this divergence is only slightly modified by the curvature and

one expects the total force to be finite. By contrast, objects with sharp corners produce several images of \mathbf{r} [for instance, three in a right corner in two dimensions (2D)]. They generate additional divergences in the edges that are not likely to be compensated (unless a symmetric corner exists on the other side of the object). We restrict ourselves to objects with radii of curvature large enough to avoid such a complication. The proper mathematical way of removing the divergences is to introduce short-range (e.g., “hard-core”) cutoffs that are removed at the end of the calculation, similar to classical Coulombic systems between opposite charges and at metallic walls. Using (4) and (2), the regularized force \mathbf{F}_S on the object S is thus given by

$$\mathbf{F}_S = \lim_{\epsilon \rightarrow 0} F_0 \kappa \int_S d\sigma \mathbf{n}(\mathbf{r}) [G - G_0](\mathbf{r} - \boldsymbol{\epsilon} \mathbf{n}(\mathbf{r}), \mathbf{r} - \boldsymbol{\epsilon} \mathbf{n}(\mathbf{r})), \quad (8)$$

where $F_0 \equiv p_0'' \Gamma / (4D\kappa)$ has the dimension of a force. We now apply this general framework to calculate the fluctuation-induced force (8) on two distinct systems embedded into the fluid. For simplicity, we limit here the fluid to two dimensions, but the conclusions also apply to three-dimensional cases.

Deformed circle. Since the force (8) on a single circle must vanish by symmetry, we deform its radius R according to $R(\theta) = R + \eta s(\theta)$ (in polar coordinates) and assume $\eta \ll \kappa^{-1}, R$. The general solution of (5) for a finite object is

$$G(\mathbf{r}, \mathbf{r}') = G_0(\mathbf{r} - \mathbf{r}') + \sum_{m,n \in \mathbb{Z}} \frac{e^{im\theta + in\theta'}}{2\pi} a_{mn} K_m(\kappa\rho) K_n(\kappa\rho'), \quad (9)$$

where (ρ, θ) are polar coordinates for \mathbf{r} , (ρ', θ') for \mathbf{r}' , and K_m is the modified Bessel function of order m . The coefficients a_{mn} satisfy $a_{mn} = a_{nm} = a_{-m, -n}^*$ to ensure that the Green function is real and symmetric under the interchange of \mathbf{r} and \mathbf{r}' . They still need to be determined from the boundary condition at $\rho = R(\theta)$. Substituting (9) into (6), one can obtain them perturbatively in η in terms of the Fourier coefficients of $s(\theta)$, $s_n \equiv (2\pi)^{-1} \int_0^{2\pi} d\theta e^{-in\theta} s(\theta)$.

In the force (8), it must be noted that the dependence on $R(\theta)$ is double: explicit in $d\sigma$, \mathbf{n} , and G (via a_{mn}) and implicit since the Green function is evaluated on the boundary of the deformed circle. The whole expression is expanded in η , and it is verified that the zeroth-order contribution, which corresponds to the force on the undeformed circle, vanishes. The contribution linear in the perturbation also vanishes. Indeed, by linearity, each Fourier mode of s can be analyzed separately; the mode $n=0$ merely corresponds to a change in the radius of the circle; the dipolar modes $n = \pm 1$ are equivalent to a small displacement of the unperturbed circle; all remaining modes $|n| \geq 2$ correspond to symmetric perturbations, not having any preferred direction. The first contribution to the force thus comes from the second order in η and has the form

$$\mathbf{F} = F_0 (\kappa\eta)^2 \sum_m f_m s_m s_{-1-m}, \quad (10)$$

where the two components of \mathbf{F} are expressed on the rhs as a complex number. This particular combination of the (complex) Fourier coefficients s_n is necessary to ensure that the

force correctly transforms as a vector, or, equivalently in Fourier modes, as a dipole. The real coefficients f_m are non-trivial series of Bessel functions evaluated at R or $R + \epsilon$, and the limit $\epsilon \rightarrow 0$ must be taken after summing them.

For simplicity, we choose a specific deformation that leads to a nonvanishing force. It can be checked that $f_0 + f_{-1} = 0$, as expected because the joint deformation given by s_0 and s_1 produces again a circle that is merely expanded and translated: its self-force vanishes. The first nontrivial cases are given by the coupling of the $n=1$ and $n=-2$ modes of $s(\theta)$. Hence considering $s(\theta) = 2s_1 \cos(\theta) + 2s_2 \cos(2\theta)$, we find

$$\mathbf{F} = -F_0 s_1 s_2 (\kappa\eta)^2 H(\kappa R) \hat{\mathbf{x}}, \quad (11)$$

where H is a dimensionless function whose numeric computation is accurately compatible with $2/(\kappa R)$ in the range $0.1 \leq \kappa R \leq 100$.

A nonvanishing self-force is therefore produced on a deformed circle at order η^2 for the simple deformation considered here, made of a dipolar and quadrupolar combination.

Circle-plate system. In systems at equilibrium, fluctuation-induced forces can only appear between two or more objects. Because the global force on the total system must vanish, such two-body forces always satisfy the action-reaction principle. The picture is different in systems driven out of equilibrium.

Indeed, consider two objects S and S' immersed in the fluid. The total force \mathbf{F}_S on S can be separated into a self-contribution \mathbf{F}_S^0 , which may already be present in the absence of S' , and a contribution $\mathbf{F}_{S \leftarrow S'} = \mathbf{F}_S - \mathbf{F}_S^0$ due to the additional asymmetry provoked by the presence of S' . Denoting by $G_{SS'}$ and G_S^0 the Green functions associated with the two-object and single-object S systems, respectively, one has, from Eq. (8), $\mathbf{F}_{S \leftarrow S'} = F_0 \kappa \int_S d\sigma \mathbf{n} (G_{SS'} - G_S^0)$ and

$$\mathbf{F}_{S \leftarrow S'} + \mathbf{F}_{S' \leftarrow S} = \mathbf{F}_{SS'} - \mathbf{F}_S^0 - \mathbf{F}_{S'}^0, \quad (12)$$

where $\mathbf{F}_{SS'}$ is the global force exerting on the assembly $S \cup S'$ considered as a whole. Note that the quantity $G_{SS'} - G_S^0$ entering in $\mathbf{F}_{S \leftarrow S'}$ is well defined: the bulk divergence and the wall divergence on S , present in both $G_{SS'}$ and G_S^0 , compensate in the subtraction. We take as definition of the action-reaction principle for the internal (two-body) forces of such system the vanishing of the rhs of (12). Since fluctuation-induced forces are not additive, this vanishing will not happen in general in the presence of self-forces, except from obvious symmetry reasons.

As an example, we take an assembly made of a circle C of radius R and a thin and long plate P in a 2D fluid. Their separation at the closest point is d (see Fig. 1). The plate is taken much longer than κ^{-1} to avoid boundary effects. Since either object is symmetric, $\mathbf{F}_C^0 = \mathbf{F}_P^0 = 0$. We calculate both terms on the lhs of (12) in the regime $R \ll \kappa^{-1} \ll d$ to show that the total force on the assembly, \mathbf{F}_{CP} , is nonzero; equivalently, the action-reaction principle is not satisfied in this situation.

In the multiple-scattering expansion of $G_{CP}(\mathbf{r}, \mathbf{r})|_{\mathbf{r} \in C}$, Eq. (7), the free-space correlation G_0 is scattered on both surfaces C and P . It is clear that when the separation d is much

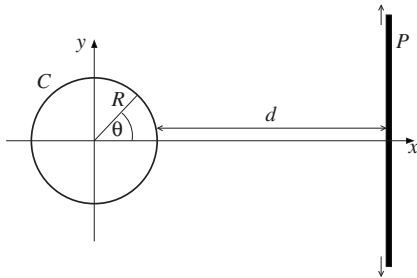


FIG. 1. A circle C of radius R and a thin, long, plate P are immersed in the fluctuating fluid. Their separation at closest point is d .

larger than the correlation length κ^{-1} , the dominant terms in this expansion contain the least number of propagations between C and P . Those containing none rebuild the series of scatterings of G_0 on C that gives the Green function of the single circle, G_C^0 . All other terms contain scatterings on P . Since the initial and final points are both $\mathbf{r} \in C$, making a scattering on P necessarily involves at least two interspace propagations, one from C to P and one to come back to C . The next dominant terms therefore contain exactly two of the propagators G_0 evaluated at points separated by at least d . However, before and after these interspace propagations, any number of scatterings from C to C or from P to P can be done. Summing them up into the quantities G_C^0 and G_P^0 , one eventually has

$$(G_{CP} - G_C^0)(\mathbf{r}, \mathbf{r})|_{\mathbf{r} \in C} \sim G_P^0 * \nabla G_C^0 + G_C^0 * \nabla G_P^0 * \nabla G_C^0.$$

Explicit expressions for G_C^0 and G_P^0 are easy to obtain. The function G_C^0 is straightforward to calculate from (9) with the result $a_{mn} = -[I'_m(\kappa R)/K'_m(\kappa R)]\delta_{n,-m}$, where I'_m and K'_m are the derivatives of the modified Bessel functions of order m .

We shall here only state the result of an asymptotic analysis of $\mathbf{F}_{C \rightarrow P}$ and $\mathbf{F}_{P \rightarrow C}$, based on small- κR expansions and steepest-descent values of integrals as $\kappa d \rightarrow \infty$ (explicit calculations, including in other regimes, will be developed in [25]). On the x axis, the forces exerting on the circle and the

plate in the regime $R \ll \kappa^{-1} \ll d$ are found to be

$$F_{C \rightarrow P} \sim -F_0 \frac{\sqrt{\pi}(\kappa R)^2 e^{-2\kappa d}}{\sqrt{\kappa d}}, \quad F_{P \rightarrow C} \sim -\frac{3}{2} F_{C \rightarrow P}. \quad (13)$$

Clearly, the action-reaction principle is not satisfied. Furthermore, the circle-plate assembly experiences a nonzero global self-force along x of the same order: $F_{CP} \sim \frac{1}{2} F_0 \frac{\sqrt{\pi}(\kappa R)^2 e^{-2\kappa d}}{\sqrt{\kappa d}}$.

In conclusion, we have shown that in nontrivial geometries the extension of Casimir-like forces to fluctuating fluids driven out of equilibrium must take into account two interrelated phenomena forbidden at equilibrium: the possibility that a self-force may be induced on a single asymmetric rigid body and that the action-reaction principle for the forces between two objects may be strongly violated. The latter fact impedes that an effective interaction potential holds in nonequilibrium. Its occurrence would prevent the use of the Derjaguin approximation. As the magnitude of this violation can be of the same order as the internal forces, special care should be taken when obtaining these forces in experiments or simulations.

The complexity of dealing with nonplanar objects has been overcome here by considering a very simple model for the nonequilibrium fluid and by devising a Green-function and multiple-scattering technique. Clearly, to allow quantitative comparison with real fluids (such as colloidal solutions, dusty plasmas, etc.), one would need to refine the initial model.

The presence of self-forces leads to directed motion if the objects are let free to move as in the case of ratchets. Self-forces can lead to arrangements of composites of intruders that could be tailored to produce microdevices by self-assembling. Their dynamical properties, however, need a more thorough analysis, for their motion will affect the fluid's fluctuations and a self-dynamical interaction could appear.

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