

Fluctuation-Induced Forces in Nonequilibrium Diffusive Dynamics

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Fluctuations in nonequilibrium steady states generically lead to power law decay of correlations for conserved quantities. Embedded bodies which constrain fluctuations, in turn, experience fluctuation induced forces. We compute these forces for the simple case of parallel slabs in a driven diffusive system. Our model calculations show that the force falls off with slab separation d as $k_B T/d$ (at temperature T , and in all spatial dimensions) but can be attractive or repulsive. Unlike the equilibrium Casimir force, the force amplitude is nonuniversal and explicitly depends on dynamics. The techniques introduced can be used to study pressure and fluctuation induced forces in a broad class of nonequilibrium systems.

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External objects immersed in a medium may modify the underlying fluctuations and, in turn, experience fluctuation-induced forces (FIF) [1]. The textbook example is the Casimir force [2,3] arising from quantum fluctuations of the electromagnetic field. Its thermal analog in critical systems [4] has been observed in binary liquid mixtures [5,6], ^4He films [7,8], and liquid-vapor coexistences [9]. In both cases, quantum and classical, the underlying fluctuations are long-range correlated leading to forces that fall off as power laws. In the latter (for example, in an oil-water mixture), this is achieved by tuning to a critical point, while the former is a consequence of the massless nature of the photon field. Generically, in a fluid in equilibrium, correlations (and, hence, FIF) decay exponentially and are insignificant beyond a correlation length.

Nonequilibrium situations provide another route to long-range correlated fluctuations: Systems which, in equilibrium, have zero or short-ranged correlations [$C_{\text{eq}} \sim \delta^s(\mathbf{x})$ in s dimensions], quite generically exhibit power law correlations ($C_{\text{neq}} \sim 1/|\mathbf{x}|^\alpha$) with conserved dynamics when out of equilibrium [10–12]. Thus, it is natural to inquire about the nature (strength and range) of FIF in corresponding nonequilibrium settings (where there is no matching force in equilibrium). Indeed, such forces have been explored in a number of circumstances, including driven granular fluids [13–16], shear flow [17], active matter systems [18], and in ordinary fluids due to the Soret effect [19] or subject to a temperature gradient [20,21]. However, despite these studies, they are much less understood than other FIF.

Here, we explore FIF in diffusive systems which are far from thermal equilibrium. First, we consider, in detail, possibly the simplest (and, hence, analytically tractable) example of FIF in a system of diffusing particles which are subject only to hard core exclusion. The model is commonly referred to as the symmetric simple exclusion process (SSEP) [22]. Then, we present perturbative results

for general diffusive systems. The methods introduced can be used to investigate a large variety of models.

The setups examined are as follows: (a) The two dimensional system shown in Fig. 1(a); infinite in the y direction and connected to two reservoirs at $x = 0$ and $x = L$, with densities $\rho(0, y) = \rho_l$ and $\rho(L, y) = \rho_r$, respectively. Two slabs, a distance d from each other, span the system along the x direction. (b) The three dimensional extension of this setup is depicted in Fig. 1(b), with the two slabs replaced by a tube of square cross section. (c) A generalized setup in which the slabs (or tube in three dimensions) of length $R \leq L$, do not necessarily span the entire system. As is evident from the discussion below, this choice of configuration guarantees that only out-of-equilibrium FIF induce a force between the slabs.

Consider, first, the two dimensional setup of Fig. 1(a) for a SSEP. For equal reservoir densities, $\rho_l = \rho_r$, the system is in equilibrium and has short range correlations. The pressure is uniform throughout the box so that there is no average force on the slabs. When the reservoir densities are different, the (average) density profile varies linearly between the two reservoirs, and there is an average diffusive current of particles along the x direction. Its magnitude is $j = D\Delta\rho/L$, where D the diffusion constant of the particles and $\Delta\rho \equiv (\rho_l - \rho_r)$. Since the average density profile is the same on both sides of each slab, naively, one would again expect no force between the two plates. However, we find that the presence of nonequilibrium long-range correlations [10,22] for $\rho_l \neq \rho_r$ leads to a force between the two slabs, given by (for $d \ll L$)

$$\begin{aligned} F &= -\frac{k_B T}{d} (\Delta\rho)^2 g(\rho_l, \rho_r) \\ &= -\frac{k_B T}{d} \left(\frac{jL}{D}\right)^2 g(\rho_l, \rho_r). \end{aligned} \quad (1)$$

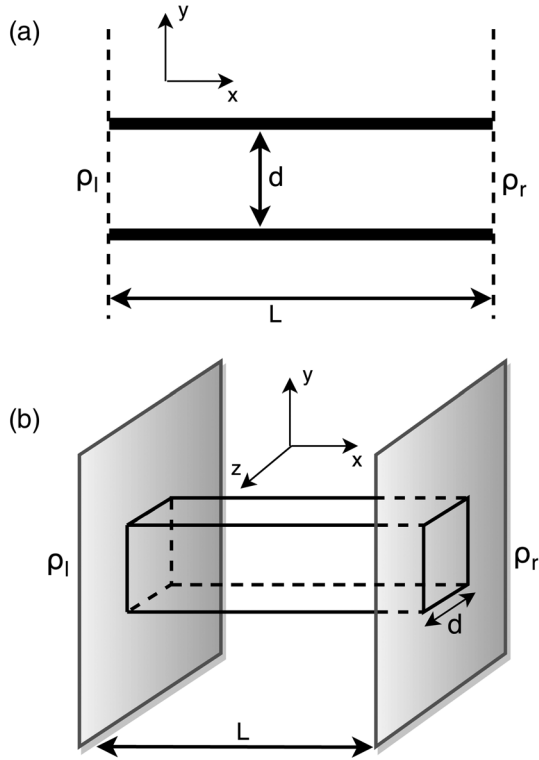


FIG. 1. The setups studied consist of (a) A two dimensional system, infinite in the y direction is connected to two reservoirs at $x = 0$ and $x = L$, with densities $\rho(0, y) = \rho_l$ and $\rho(L, y) = \rho_r$, respectively. Two slabs, a distance d from each other, span the system along the x direction. (b) The three dimensional generalization of the above, with the two slabs replaced by a tube of square cross section.

Here, k_B is the Boltzmann constant, T is the temperature of the surrounding bath, and $g(\rho_l, \rho_r)$ is a positive dimensionless function of order one. Note that the force is attractive and, when expressed in terms of current or the average density gradient $\overline{\nabla\rho} = \Delta\rho/L$, proportional to $L^2(\overline{\nabla\rho})^2$. Here, the overline denotes an average over the steady-state probability distribution. For $d \gg L$, the force still decays as $1/d$ but with a coefficient that is smaller by a factor of 2. When the three dimensional analog of the above setup is considered and a tube with a square cross section connects the two reservoirs [see Fig. 1(b)], the force between two parallel slabs has the same form, with the same function $g(\rho_l, \rho_r)$ for $d \ll L$.

For slabs or a tube of finite extension, R , and assuming that fluctuations at the edges of the slabs or tube are negligible, our results suggest that the force should behave as

$$\begin{aligned} F &= -\frac{k_B T R^2}{d L^2} (\Delta\rho)^2 g^* \left(\rho_l, \rho_r, \frac{R}{x_0} \right) \\ &= -\frac{k_B T}{d} R^2 \left(\frac{j}{D} \right)^2 g^* \left(\rho_l, \rho_r, \frac{R}{x_0} \right). \end{aligned} \quad (2)$$

Here g^* is a positive function of ρ_l, ρ_r and R , while x_0 is the distance of the left side of the slabs or tube from the left reservoir. For hard core particles, the force is attractive and proportional to R^2 .

Finally, we derive the form of FIF for general diffusive models to order $(\Delta\rho)^2$. The result shows that, in contrast to equilibrium systems, out-of-equilibrium FIF depend on the specific choice of dynamics. Moreover, the above scaling forms remain, and while the force is attractive for SSEP, it can be repulsive in other interacting diffusive systems. We argue that this is the case in boundary driven antiferromagnetic Ising models with spin conserving dynamics for a certain regime of parameters.

To derive the above results, we use fluctuating hydrodynamics [10,22,23]. In this approach, the dynamical equation of motion for the particle density can be shown, either through a microscopic derivation (for example, see [24]) or through a phenomenological approach, to be

$$\partial_t \rho(x, t) + \partial_x J(x, t) = 0, \quad (3)$$

with a stochastic current

$$J_\mu(x, t) = -D \partial_\mu \rho(\mathbf{x}, t) + \sqrt{\sigma(\rho)} \eta_\mu(\mathbf{x}, t). \quad (4)$$

Here, D is a diffusion coefficient and η_μ is an uncorrelated white noise vector with components $\mu = 1, \dots, s$, with s the system dimension. The noise has zero mean $\overline{\eta_\mu(\mathbf{x}, t)} = 0$, is uncorrelated $\overline{\eta_\mu(\mathbf{x}, t) \eta_\nu(\mathbf{x}', t')} = \delta_{\mu\nu} \delta(t - t') \delta(\mathbf{x} - \mathbf{x}')$; its variance $\sigma(\rho) = 2Dk_B T \rho^2 \kappa(\rho)$ satisfying a fluctuation-dissipation condition, where $\kappa(\rho)$ is the compressibility of the gas. For diffusing particles subject to hard-core exclusion, D is a constant independent of the density ρ , and $\sigma(\rho) = 2Da^s \rho(1 - \rho)$ [10,22]. Here, a is a UV cutoff given by the lattice size, and we use the standard convention where $0 \leq \rho \leq 1$ is dimensionless. For simplicity, in what follows, derivations are mostly restricted to two dimensions [Fig. 1(a)]; the extension to three dimensions [Fig. 1(b)] is straightforward and provided in the Supplemental Material [25].

The density is subject to the boundary conditions $\rho(0, y) = \rho_l$ and $\rho(L, y) = \rho_r$ at the reservoirs, while the normal component of the current must vanish on the two slabs. In steady-state, the average density profile is given by $\bar{\rho}(x, y) = \rho_l + \Delta\rho x/L$, with average current $\bar{\mathbf{j}} = (D\Delta\rho/L)\hat{\mathbf{x}}$.

It is important to note that the continuum equations are valid in the hydrodynamic limit of a corresponding lattice obtained as follows: Consider a (hyper-)cubic system of volume L^s divided into N^s boxes of size ξ^s , where ξ is a length scale such that $N\xi = L$. The hydrodynamic regime corresponds to first letting $\xi \rightarrow \infty$ and $L \rightarrow \infty$ with $L/\xi = N$, and then taking the limit $N \rightarrow \infty$. Eq. (3) is valid when the system is rescaled and length is measured in units where $\xi \rightarrow 0$ and $N\xi = L$ [22].

With this in mind and using ideas similar to Refs. [14,15,20,21], we write the average pressure to leading order in the fluctuations as

$$\overline{P(\rho(\mathbf{x}))} = \lim_{\xi \rightarrow \infty} \left(P(\bar{\rho}(\mathbf{x})) + \frac{1}{2} P''|_{\bar{\rho}(\mathbf{x})} \overline{\delta\rho(\mathbf{x})^2} \right). \quad (5)$$

Here, $\delta\rho(\mathbf{x}) = \rho(\mathbf{x}) - \bar{\rho}(\mathbf{x})$, and primes, henceforth, indicate derivatives with respect to the density ρ . To calculate the force between the plates, the pressure has to be evaluated on both sides of each slab. The hydrodynamic procedure described above implies that calculations have to be carried out using Eq. (3) with the cutoff ξ , and then with length scales rescaled at the end of the calculation so that L is finite. In practice, this implies that any divergent UV contributions to the pressure fluctuations need to be removed from the results of the calculation. In particular, in equilibrium and using the continuum result $\overline{\delta\rho(\mathbf{x})\delta\rho(\mathbf{x}')} = a^s \rho(1-\rho)\delta(\mathbf{x}-\mathbf{x}')$, one has $\overline{\delta\rho(\mathbf{x})^2} = a^s \rho(1-\rho)/\xi^s$, and the fluctuations do not contribute to the pressure as $\xi \rightarrow \infty$. Clearly, in the setup considered, at any location along the wall contributions from $P(\bar{\rho}(\mathbf{x}))$ cancel. However, as we now show, $\overline{\delta\rho(\mathbf{x})^2}$ varies on the opposing faces of each slab leading to a fluctuation induced force. [Note that, if the plates were tilted, the density along the two sides of a slab would be different due to the current. This would lead to a dominant contribution to the force between the two plates from $P(\bar{\rho}(\mathbf{x}))$. Since our focus is on FIF, we do not consider such cases.]

To evaluate the nonequilibrium fluctuation induced contribution to pressure, note that, for the SSEP, the fluctuation-dissipation relation $\sigma(\rho) = 2Dk_B T \rho^2 \kappa(\rho)$, with $\kappa(\rho) = \frac{1}{\rho} \frac{d\rho}{dP}$ and $\sigma = 2Da^s \rho(1-\rho)$, gives

$$\frac{1}{2} P''|_{\bar{\rho}(\mathbf{x})} = \frac{1}{2a^s} \frac{k_B T}{[1 - \bar{\rho}(\mathbf{x})]^2}. \quad (6)$$

To compute $\overline{\delta\rho(\mathbf{x})^2}$ on the faces of the slabs, we evaluate the fluctuations along the walls in chambers of size $L \times d$ and $L \times \infty$, respectively. The first corresponds to the chamber between the walls, and the second to the semi-infinite surrounding spaces.

To evaluate $\overline{\delta\rho(x, y)^2}$ we use standard methods [27,28], expanding the equation of motion to linear order in $\delta\rho$ about the steady-state profile. The current to linear order is

$$J_\mu(x, t) = -D\partial_\mu \delta\rho(\mathbf{x}, t) + \sqrt{\sigma(\bar{\rho}(\mathbf{x}))} \eta_\mu(\mathbf{x}, t). \quad (7)$$

The dynamical equation is then linear in $\delta\rho$ so that the correlation function $C(\mathbf{x}, \mathbf{x}') = \overline{\delta\rho(\mathbf{x})\delta\rho(\mathbf{x}')}$ satisfies a Lyapunov equation [27,28]. After several straightforward manipulations, this can be brought to the form

$$\begin{aligned} & (\nabla_{\mathbf{x}} D \nabla_{\mathbf{x}} + \nabla_{\mathbf{x}'} D \nabla_{\mathbf{x}'}) C_{\text{neq}}(\mathbf{x}, \mathbf{x}') \\ & = -\frac{1}{2} \delta(\mathbf{x} - \mathbf{x}') \nabla_{\mathbf{x}'}^2 \sigma(\bar{\rho}(\mathbf{x}')), \end{aligned} \quad (8)$$

where $C_{\text{neq}}(\mathbf{x}, \mathbf{x}') = C(\mathbf{x}, \mathbf{x}') - \frac{1}{2D} \sigma(\bar{\rho}(\mathbf{x}')) \delta(\mathbf{x} - \mathbf{x}')$ is the nonequilibrium part of the correlation function. Using the average density profile, $\bar{\rho}(x, y) = \rho_l + \Delta\rho x/L$, the above equation reduces to calculating the Green's function of a Poisson equation

$$(\nabla_{\mathbf{x}}^2 + \nabla_{\mathbf{x}'}^2) C_{\text{neq}}(\mathbf{x}, \mathbf{x}') = 2\delta(\mathbf{x} - \mathbf{x}') (\Delta\rho)^2 a^2 / L^2. \quad (9)$$

The boundary conditions are such that $C_{\text{neq}} = 0$ when either \mathbf{x} and \mathbf{x}' are on the reservoirs (since the density on the reservoirs is fixed, $\delta\rho = 0$ identically), while, on the slabs, its normal derivative is zero (no current). To calculate the force, density fluctuations have to be calculated on the slabs, e.g., $c_{\text{neq}}(x) \equiv C_{\text{neq}}(\{x, y = 0\}, \{x, y = 0\})$, evaluated at the same point $\mathbf{x} = \mathbf{x}'$ on one of the slabs. Using standard Fourier methods, one finds

$$c_{\text{neq}}(x) = \sum_n A_n \sin^2\left(\frac{n\pi}{L} x\right), \quad (10)$$

with

$$A_n = -\frac{a^2 (\Delta\rho)^2}{Ld} \left[\left(\frac{1}{n\pi}\right)^2 + \frac{d}{n\pi L} \coth\left(\frac{n\pi d}{L}\right) \right]. \quad (11)$$

In the limit $d \gg L$, one finds to order L/d

$$A_n = -a^2 (\Delta\rho)^2 \left[\frac{1}{(n\pi)^2 L^2} + \frac{1}{(n\pi)^2 Ld} \right]. \quad (12)$$

Conversely, for $d \ll L$ (indicated by the superscript 1) and to leading order in d/L

$$A_n^1 = -a^2 \frac{2(\Delta\rho)^2}{Ld} \left(\frac{1}{n\pi}\right)^2. \quad (13)$$

The Fourier series with $A_n \propto (n\pi)^{-2}$ corresponds to a parabola. For $d \ll L$, this gives

$$c_{\text{neq}}^1(x) = -a^2 \frac{(\Delta\rho)^2}{Ld} \frac{x}{L} \left(1 - \frac{x}{L}\right), \quad (14)$$

which is, in fact, the expected behavior of a one-dimensional SSEP [10,22]. For $d \gg L$, Eq. (12) leads to a constant contribution, corresponding to the $d \rightarrow \infty$ limit, and a contribution similar to $c_{\text{neq}}^1(x)$ with a coefficient that is smaller by two. Using the hydrodynamic procedure described earlier, we observe that $\overline{\delta\rho(\{x, y = 0\})^2} = c_{\text{neq}}(x)$. Namely, only the long-range part of the correlation function contributes to the pressure.

The fluctuation-induced correction to the pressure in Eq. (5) is the product of two factors: the first [given in Eq. (6)] is positive, while the second [from Eq. (14)] is negative. This leads to a negative contribution to pressure, corresponding to attraction between the slabs. In the limit $d \ll L$, the contribution from the semi-infinite surrounding spaces is negligible. Integrating the local pressure over the slab leads to a fluctuation-induced force

$$F = \int dx \frac{1}{2} P''|_{\bar{\rho}(x)} c_{\text{neq}}^1(x) = -\frac{k_B T (\Delta\rho)^2}{d} \int_0^1 dz \frac{z(1-z)}{2[1-\bar{\rho}(z)]^2}, \quad (15)$$

consistent with Eq. (1). Here, $\bar{\rho}(z) = \rho_l + \Delta\rho z$. Evaluating the integral shows that the total force is a concave function, vanishing at $\rho_l = \rho_r$. It is straightforward to use the above results to verify that, in the limit $d \gg L$, the force decays in the same form with a coefficient that is smaller by two. The calculation can be repeated in three dimensions for the configuration depicted in Fig. 1(b) (See [25] for details). The force is now calculated between two opposite slabs, say in the y direction, and yields the exact same result as above.

The negative result in Eq. (14) may appear counterintuitive, since it originates from a computation of $\delta\rho(x)^2$. To validate this conclusion, and the underlying hydrodynamic procedure, we performed Monte-Carlo simulations on the SSEP model in two dimensions and measured the pressure along the slab (see [25] for details). The results in the limit $d/L \ll 1$ and for different lattice sizes are shown in Fig. 2.

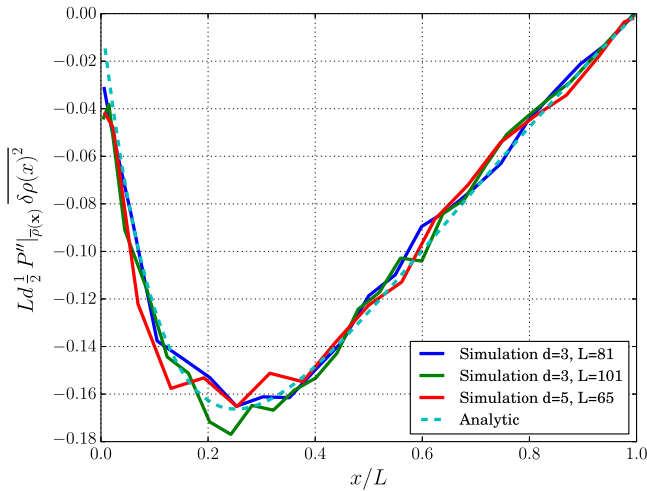


FIG. 2 (color online). Numerical results for the fluctuation induced pressure in two-dimensions, given by the integrand of Eq. (15), multiplied by Ld . Here, $\rho_l = 0.1$, $\Delta\rho = 0.6$, and three values of L, d such that $d \ll L$, are shown. Units are chosen such that the lattice spacing is $a = 1$ and $k_B T = 1$. Solid lines depict numerical results; the dashed line is the analytic calculation. The method for measuring the pressure is described in the Supplemental Material [25].

The numerics compare well with the theoretical predictions.

Equation (5) suggests that the pressure and, therefore, the force can be either positive or negative, depending on the relative signs of P'' and c_{neq} . To explore this further, we carry out a perturbation theory in $\Delta\rho$ for a general model with a density dependent diffusion constant $D(\rho)$. The equation for the average density is then $\nabla[D(\bar{\rho}(\mathbf{x})) \cdot \nabla\bar{\rho}(\mathbf{x})] = 0$, and the Lyapunov Eq. (8) now has D as a function of ρ . Setting $\bar{\rho}(x) = \rho_l + \rho_1(x)\Delta\rho + \rho_2(x)(\Delta\rho)^2 + \dots$, it is straightforward to show that, to order $(\Delta\rho)^2$, the result in Eq. (14) is replaced by

$$c_{\text{neq}}^1(\mathbf{x}) \simeq \frac{k_B T (\Delta\rho)^2}{2Ld} \left[\left(\frac{\rho}{P'} \right)'' + \left(\frac{\rho}{P'} \frac{D'}{D} \right)' \right] \frac{x}{L} \left(1 - \frac{x}{L} \right), \quad (16)$$

resulting in a force

$$F \simeq \frac{k_B T (\Delta\rho)^2}{24d} P'' \left[\left(\frac{\rho}{P'} \right)'' + \left(\frac{\rho}{P'} \frac{D'}{D} \right)' \right], \quad (17)$$

where the derivatives with respect to the density are evaluated at ρ_l . The second term on the right-hand-side shows the explicit dependence of the results on the dynamics through the appearance of the diffusion coefficient. Moreover, there are no apparent restrictions on the sign of the force. For example, consider a model with $D = k^2[1 - q^2(\rho - \rho_0)]$, $\sigma(\rho) = r^2[1 + t^2(\rho - \rho_0)^2]$ and boundary conditions with $\rho_l = \rho_0$. While we are not aware of a direct microscopic realization of this formula, it can be considered as an approximation for an Ising model with repulsive interactions evolving under Kawasaki dynamics, with ρ denoting, say, the density of down spins. There, it is known that in one dimension $\sigma(\rho)$ has a minimum around some ρ_0 which depends on the parameters of the model, with $D(\rho)$ peaked around ρ_0 [29,30]. (On general grounds this behavior is expected to persist in higher dimensions.) Using the above expressions, it is straightforward to check that, to order $(\Delta\rho)^2$, the fluctuation induced force, $F \simeq \frac{k_B T (\Delta\rho)^2 t^2}{12d}$, is repulsive.

The nonextensivity of the force in Eq. (15) is somewhat surprising, and different from, say, the critical Casimir force which behaves as $F \propto k_B T L^{s-1}/d^s$ for generalized slabs of side L in s dimensions [1]. This is because c_{neq} scales inversely with the volume of the confining box, resulting in a local pressure that vanishes for a large slab. As such, we expect this force to be more relevant to small inclusions as opposed to macroscopic slabs. While the exact solution of the force between two inclusions is beyond the scope of this Letter, we can provide an estimate based on dimensional grounds for the SSEP. To this end, we consider parallel slabs of dimension R , and neglect the fluctuations of

density at the open sides of the corresponding enclosure. One is then left with evaluating the pressure fluctuations in a chamber of size $R \times d^{s-1}$ with boundary densities specified by the mean density at the edges of the slab. It is then straightforward to see that, in the limit $d \ll L$, the force is now given by (for SSEP)

$$F = -\frac{2k_B T (\Delta\rho)^2 R^2}{d L^2} \int_0^1 dz \frac{z(1-z)}{[1-\bar{\rho}(z)]^2}, \quad (18)$$

irrespective of dimension s , where $\bar{\rho}(z) = \rho_l + (\Delta\rho)(x_0 + Rz)/L$ as advertised in Eq. (2).

To conclude, we analyzed the force between two parallel slabs immersed in a diffusive system driven out of equilibrium. The origin of the FIF in these systems is very different from the one leading to thermodynamic Casimir effects [4], and its behavior is distinct from, for example, equilibrium slab geometries [31,32]. It will be interesting to study the behavior of the force in other geometries.

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