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Reconciling nonlinear dissipation with the bilinear model of two Brownian particles

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The Brownian motion of a single particle is a paradigmatic model of the nonequilibrium dynamics of dissipative systems. In the system-plus-reservoir approach, one can derive the particle's equations of motion from the reversible dynamics of the system coupled to a bath of oscillators representing its thermal environment. However, extending the system-plus-reservoir approach to multiple particles in a collective environment is not straightforward, and conflicting models have been proposed to that end. Here, we set out to reconcile some aspects of the nonlinear and the bilinear models of two Brownian particles. We show how the nonlinear dissipation originally derived from exponential system-reservoir couplings can alternatively be obtained from the bilinear Lagrangian, with a modified spectral function that explicitly depends on the distance between the particles. We discuss applications to the contexts of anomalous diffusion and of hydrodynamic interactions. Our results thus broaden the applicability of the bilinear model.

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I. INTRODUCTION

The Brownian motion is a representative example of nonequilibrium dissipative dynamics. Theoretically, the irreversible dynamics of a subsystem can emerge from a reversible dynamics of the global system. To achieve that, one way is to phenomenologically model the particle's environment as a set of independent oscillators, each linearly coupled to the system of interest. This is the so-called system-plus-reservoir approach. By tuning the spectral function, which determines the weight of each frequency mode on the particle's dissipation rate, one can recover experimental observations. This framework has proven useful across classical and quantum domains [1–3].

Let us consider two particles immersed in the same bath. This can be relevant, for instance, when environment-induced effects on multiple degrees of freedom are being investigated, as in the cases of biologically inspired problems [4,5], of non-Markovianity [6], of synchronization [7] and of quantum entanglement [8–11], to name a few. One way to address this problem is to employ the so-called bilinear model, which assumes that each particle is linearly coupled to the same set of oscillators [4,6,7,9,10,12,13]. However, such a bilinear coupling may lead to unphysical results, namely, the free-particle motion of the relative coordinate, and the absence of mutual effects between proximal particles, as pointed out by Duarte and Caldeira [14]. To solve these issues, a nonlinear model for the system-environment couplings has been devised, which not only recovers the well-known single-particle case, but also predicts dissipation rates that are nonlinear functions of the distance between the pair of Brownian particles [14].

Despite the successes of the nonlinear model for two Brownian particles, the bilinear approach has its merits. Experimentally, it can yield correct results, as in the case of a recent demonstration of environment-induced entanglement in the optical domain [15]. Theoretically, linearity allows the model to be exactly solvable in the quantum and the classical regimes, making it a desirable tool. We highlight the method recently developed by Weiderpass and Caldeira to characterize entropy production by a quantum Brownian particle [16]. The ansatz proposed in Ref. [16] is strongly relying on the linearity of the model employed. Extending Weiderpass and Caldeira's ansatz to multiple quantum Brownian particles could contribute to the field of far-from-equilibrium thermodynamics of quantum many-body dissipative systems [17,18].

Here, we address the following question. Is there an alternative way to avoid the shortcomings of the standard bilinear model, without recurring to nonlinear system-environment couplings? To that end, we introduce a physically motivated spectral function that explicitly depends on the relative distance between a pair of Brownian particles, as derived from a nonlinear response theory of the bath of oscillators perturbed by the particles. By doing so, we obtain, from the bilinear model, the nonlinear dissipation that had been originally derived from exponential system-environment couplings [14]. Our method thereby broadens the applicability of the bilinear model, as it now embraces the description of nonlinear environment-induced forces. We discuss how our modified bilinear model can be adjusted so as to avoid anomalous diffusions, and to describe hydrodynamic forces between a pair of Brownian particles [19].

In Sec. II, we revisit the bilinear and the nonlinear models. In Sec. III, we show our main result, namely, how nonlinear dissipation can be obtained from the bilinear Lagrangian. For that, we define a spectral function that depends

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on the relative distance between the particles. In Sec. IV we explore the consequences of specific changes in our distance-dependent spectral function. We first show how our modified spectral function enables us to bridge the dissipation rates from the standard nonlinear and the standard bilinear models (Sec. IV A). We then discuss how to avoid an anomalous diffusion described in the original nonlinear model (Sec. IV B), and how our approach can be applied to model a pair of Brownian particles sharing a hydrodynamic environment (Sec. IV C). Finally, in Sec. V we present our conclusions and further considerations. Detailed derivations are presented in the Appendixes.

II. PRELIMINARY REMARKS

A. Standard bilinear model

In the bilinear model, the Lagrangian of two classical Brownian particles immersed in a collective environment reads

$$L = \frac{m}{2}\left(\dot{x}_1^2 + \dot{x}_2^2\right) + \sum_k \frac{m_k}{2}\left(\dot{R}_k^2 - \omega_k^2 R_k^2\right) - \sum_k R_k\left(C_k^{(1)}x_1 + C_k^{(2)}x_2\right) - \sum_k \frac{\left(C_k^{(1)}x_1 + C_k^{(2)}x_2\right)^2}{2m_k\omega_k^2}. \tag{1}$$

Here, $\dot{x}_{1,2}$ and $x_{1,2}$ are the velocity and the positions of the particles with mass m. R_k is the position of the kth bath oscillator, with frequency ω_k and mass m_k. The coupling between the system and each oscillator is assumed to be linear in their positions, with distinct coupling strengths $C_k^{(i)}$. The counterterm is added so as to offset the environment-induced modification of the external potential.

One can derive the Euler-Lagrange equations for both the system and the bath. From now on, we define the center of mass and relative coordinates as

$$q = \frac{x_1 + x_2}{2}, \tag{2}$$

and

$$u = x_1 - x_2, \tag{3}$$

which yield the following equations of motion,

$$\begin{aligned} m\ddot{q}(t) + (\eta + \eta_{12})\dot{q}(t) &= f_q(t),\\ m\ddot{u}(t) + (\eta - \eta_{12})\dot{u}(t) &= f_u(t). \end{aligned} \tag{4}$$

One can interpret $f_q(t)$ and $f_u(t)$ as the fluctuating forces for the center of mass and the relative coordinate,

$$f_q(t) = -\sum_k \frac{\left(C_k^{(1)} + C_k^{(2)}\right)}{2} \times \left(\dot{R}_k(0)\frac{\sin\omega_k t}{\omega_k} + \tilde{R}_k(0)\cos\omega_k t\right), \tag{5}$$

$$f_u(t) = \sum_k \left(C_k^{(1)} - C_k^{(2)}\right) \times \left(\dot{R}_k(0)\frac{\sin\omega_k t}{\omega_k} + \tilde{R}_k(0)\cos\omega_k t\right). \tag{6}$$

where $\tilde{R}_k(0) = R_k(0) + [C_k^{(i)}x_i(0) + C_k^{(j)}x_j(0)]/m_k\omega_k^2$. The statistical properties of these forces stem from the initial state of the total system.

For the dissipation term, we assume an Ohmic bath [3], so the spectral functions read

$$J_i(\omega) = \frac{\pi}{2}\sum_k \frac{C_k^{(i)2}}{m_k\omega_k}\delta(\omega - \omega_k) \equiv \eta\omega\Theta(\Omega - \omega), \tag{7}$$

with the high-frequency cutoff Ω [20,21], and with Θ being the Heaviside step function. Similarly, a mixed spectral function appears,

$$J_{12}(\omega) = \frac{\pi}{2}\sum_k \frac{C_k^{(1)}C_k^{(2)}}{m_k\omega_k}\delta(\omega - \omega_k) = \eta_{12}\omega\Theta(\Omega - \omega), \tag{8}$$

as an indication of bath-mediated interactions between the particles. This introduces the dissipation rates η and η_{12}.

Two properties of this model call our attention. First, the case of identical couplings, $C_k^{(1)} = C_k^{(2)}$, (a reasonable hypothesis, as far as two proximal particles in the same environment are concerned) implies that $\eta = \eta_{12}$ and $f_u(t) = 0$, leading to vanishing dissipation and fluctuating forces [see Eqs. (4) and (6)]. It means that a free-particle motion is found, namely,

$$\ddot{u}(t) = 0. \tag{9}$$

Second, even for nonidentical couplings one finds that η_{12} is independent of the distance between the particles. Both considerations suggest instantaneous effects between spatially separate entities. As discussed by Duarte and Caldeira [14], these are undesirable features, arising from the lack of an appropriate length scale for environment-mediated phenomena.

B. Standard nonlinear model

In the nonlinear model as introduced by Ref. [14], the Lagrangian reads

$$L = \frac{m}{2}\left(\dot{x}_1^2 + \dot{x}_2^2\right) + \frac{1}{2}\sum_k m_k\left(\dot{R}_k\dot{R}_{-k} - \omega_k^2 R_k R_{-k}\right) - \frac{1}{2}\sum_k [(C_{-k}(x_1) + C_{-k}(x_2))R_k + (C_k(x_1) + C_k(x_2))R_{-k}], \tag{10}$$

where

$$C_k(x) = \kappa_k e^{ikx}. \tag{11}$$

The exponential couplings between the bath and the particles guarantee homogeneity and translational invariance. Note that the index k now has dimensions of $[L]^{-1}$, explaining why it introduces the required length scale.

The equations of motion are

$$m\ddot{x}_i(t) + \int_0^t K(x_i(t) - x_i(t'), t - t')\dot{x}_i(t')dt' + \int_0^t K(x_i(t) - x_j(t'), t - t')\dot{x}_j(t')dt' + \frac{\partial}{\partial x_i}V(x_i(t) - x_j(t)) = F_i(t) \tag{12}$$

with $i, j = 1, 2$, and once again $F_i(t)$ can be interpreted as the fluctuating force. The dissipation kernels are

$$K(r,\tau) = \sum_k \int_0^\infty d\omega\, 2k^2\kappa_k\kappa_{-k} \times \frac{\mathrm{Im}\chi_k^{(i)}(\omega)}{\pi\omega}\cos\omega_k\tau\cos kr, \tag{13}$$

written in terms of the imaginary part of the dynamical response of the environment oscillators, namely,

$$\mathrm{Im}\chi_k^{(i)}(\omega) \equiv \frac{\pi}{2m_k\omega_k}\delta(\omega-\omega_k). \tag{14}$$

In the present model, $\mathrm{Im}\chi_k^{(i)}(\omega)$ is equivalent to the spectral function in that it allows for the transformation of a discrete set of oscillators into a continuum. The δ function is thus replaced by a Lorentzian peaked around ω_k. The next step is to focus on the low-frequency limit of that Lorentzian so as to recover the Ohmic regime (linear in ω). This justifies the approximation

$$\mathrm{Im}\chi_k^{(i)}(\omega) \approx \tilde{f}(k)\,\omega\,\Theta(\Omega-\omega). \tag{15}$$

Nonlinear equations of motion are obtained, such that

$$m\ddot{q}(t) + (\eta + \eta_e[u(t)])\dot{q}(t) = f_q(t), \tag{16}$$

and

$$m\ddot{u}(t) + (\eta - \eta_e[u(t)])\dot{u}(t) + V_e'(u(t)) = f_u(t). \tag{17}$$

Here,

$$V_e(u) \equiv -\frac{2\Omega\eta}{\pi k_0^2\left(k_0^2u^2+1\right)} \tag{18}$$

represents an environment-induced potential, which depends on the relative distance. Similarly,

$$\eta_e[u] \equiv \eta\frac{\left(1-3k_0^2u^2\right)}{\left(k_0^2u^2+1\right)^3} \tag{19}$$

describes a distance-dependent bath-mediated dissipation rate. The constant k_0 is a characteristic inverse length introduced when the summation over k is transformed into an integral, $\sum_k \to (L/2\pi)\int dk$, and a density of spatial modes is postulated,

$$\eta g(k) \equiv \frac{L}{2\pi}\kappa_k\kappa_{-k}\tilde{f}(k), \tag{20}$$

such that $\int_0^\infty dk\, k^2 g(k) = 1$. Finally, the choice for

$$g(k) = \left(\frac{1}{2k_0^3}\right)e^{-k/k_0} \tag{21}$$

explains how k_0 is defined in Ref. [14].

Equations (16)–(19) describe rich environment-induced behaviors for proximal particles, while recovering the independent Brownian movement for arbitrarily large distances. The free-particle anomaly found in the bilinear model is no longer present, since both the dissipative and the fluctuating forces are finite for any finite $u(t)$. The statistical properties of the fluctuating forces are such that (see Appendix A)

$$\langle f_q(t)\rangle = \langle f_u(t)\rangle = 0,$$
$$\langle f_q(t)f_q(t')\rangle = k_BT(\eta+\eta_e[u(t)])\delta(t-t'),$$
$$\langle f_u(t)f_u(t')\rangle = 4k_BT(\eta-\eta_e[u(t)])\delta(t-t'),$$

in agreement with the fluctuation-dissipation theorem.

III. RESULTS

In this section we show how the nonlinear dissipation term $\eta_e[u(t)]$ in Eqs. (16) and (17) can be obtained from the bilinear Lagrangian. Let us consider once again the model from Eq. (1). Note that our results could be easily extended to include direct couplings between the particles and external potentials, as done in Ref. [9]. In order to avoid the anomalous free-particle motion for the relative coordinate ($\ddot{u} = 0$), we have to assume that each particle has a distinct coupling parameter ($C_k^{(1)} \neq C_k^{(2)}$), otherwise the bath decouples from $u = x_1 - x_2$. Using similar techniques as in the previous sections, one gets to the following equations of motion for each particle:

$$m\ddot{x}_i + \sum_k \frac{C_k^{(i)2}}{m_k\omega_k^2}\int_0^t \cos\omega_k(t-t')\dot{x}_i(t')dt' + \sum_k \frac{C_k^{(i)}C_k^{(j)}}{m_k\omega_k^2}\int_0^t \cos\omega_k(t-t')\dot{x}_j(t')dt' = f_i(t), \tag{22}$$

where $i \neq j = 1, 2$, and $f_i(t)$ term can be interpreted as the fluctuating force,

$$f_i(t) = -\sum_k C_k^{(i)}\left[\dot{\tilde{R}}_k(0)\frac{\sin\omega_k t}{\omega_k} + \tilde{R}_k(0)\cos\omega_k t\right], \tag{23}$$

where $\tilde{R}_k(0) = R_k(0) + (C_k^{(i)}x_i(0) + C_k^{(j)}x_j(0))(m_k\omega_k^2)^{-1}$.

To evidence the center of mass and the relative coordinates, we rewrite the equations of motion as

$$m\ddot{q}(t) + \frac{1}{2}\int_0^t [K_1(t-t')\dot{x}_1(t') + K_2(t-t')\dot{x}_2(t')]dt' + \int_0^t K_{ij}(t-t')\dot{q}(t')dt' = f_q(t), \tag{24}$$

and

$$m\ddot{u}(t) + \int_0^t [K_1(t-t')\dot{x}_1(t') - K_2(t-t')\dot{x}_2(t')]dt' - \int_0^t K_{ij}(t-t')\dot{u}(t')dt' = f_u(t). \tag{25}$$

The dissipation kernels in Eqs. (24) and (25) are different from the ones in Ref. [14] in the lack of a spatial dependence of the environment-induced effects, as we show below,

$$K_i(t-t') = \sum_k 2C_k^{(i)2}\int_0^\infty d\omega \frac{\mathrm{Im}\chi_k^{(i)}(\omega)}{\pi\omega}\cos\omega(t-t'), \tag{26}$$

$$K_{ij}(t-t') = \sum_k 2C_k^{(i)}C_k^{(j)}\int_0^\infty d\omega \frac{\mathrm{Im}\chi_k^{(ij)}(\omega)}{\pi\omega}\cos\omega(t-t'). \tag{27}$$

We have also used Eq. (14) to write the kernels in terms of the imaginary part of the bath susceptibility functions.

As far as the single-particle dissipation rates are concerned, we define

$$\eta \equiv \sum_k C_k^{(i)2} \tilde{f}(k), \tag{28}$$

where $\tilde{f}(k)$ comes from the Ohmic approximation in Eq. (26), i.e., $\mathrm{Im}\chi_k^{(i)}(\omega) \approx \tilde{f}(k)\,\omega\,\Theta(\Omega-\omega)$ [similarly to Eq. (15)]. To obtain the continuum limit, we replace $\sum_k C_k^{(i)2}\tilde{f}(k)$ by $\eta\int dk\; k^2 g(k)$, where $g(k) = 2^{-1}k_0^{-3}\exp(-k/k_0)$ (as in Sec. II B).

The key step in our derivation concerns the two-particle susceptibility $\chi_k^{(ij)}$. We consider that, when two Brownian particles are sufficiently close, the environment acting on each particle is composite: It is jointly formed by the free environment dynamics plus the perturbation of the other particle dynamics on that environment. Put differently, each particle is immersed in a structured environment due to the presence of the complementary particle, when these are close enough. As a consequence, our insight translates into a response function that should depend on the distance between the particles or, more generally, on the relative coordinate u. We thus postulate that, in our model,

$$\mathrm{Im}\chi_k^{(ij)}(\omega) \equiv \mathrm{Im}\chi_k^{(ij)}(\omega, u). \tag{29}$$

In Appendix B, we show that $\mathrm{Im}\chi_k^{(ij)}(\omega, u)$ results from a nonlinear response theory of the environment under the perturbation of the pair of particles. We obtain the Ohmic regime by choosing a linear function in ω, namely,

$$\mathrm{Im}\chi_k^{(ij)}(\omega, u) \approx h(k, u)\,\omega\,\Theta(\Omega-\omega), \tag{30}$$

where $h(k, u)$ is to be defined. The explicit choice for $h(k, u)$ will allow us to define the bath-related length scale. Because our main goal is to obtain nonlinear dissipation consistently with the standard nonlinear model, we write $h(k, u)$ in the form

$$h(k, u) = \tilde{F}(k)G(k, u). \tag{31}$$

Here, $\tilde{F}(k)$ is analogous to $\tilde{f}(k)$ in that it allows us to define

$$\eta_{\mathrm{eff}}[u] \equiv \eta\int dk\; g_{\mathrm{eff}}(k)G(k, u) \tag{32}$$

as the continuum limit of $\sum_k C_k^{(i)}C_k^{(j)}\tilde{F}(k)G(k, u)$. This summation is obtained from applying Eqs. (30) and (31) to (27). We have also defined

$$g_{\mathrm{eff}}(k) \equiv k^2 g(k), \tag{33}$$

motivated by the fact that we recover the single-particle dissipation rate by choosing $G(k, u) = 1$.

Using Eqs. (28) and (32), and taking the limit $\Omega \to \infty$ in Eqs. (26) and (27), the dissipation kernels become

$$K_i(t-t') = 2\eta\delta(t-t') \tag{34}$$

$$K_{ij}(t-t') = 2\eta_{\mathrm{eff}}[u]\delta(t-t'). \tag{35}$$

The length scale is now explicit, given by the dependence of the kernel on the relative coordinate u. By tuning $G(k, u)$, one controls the nonlinear dissipation force appearing in the equations of motion, be it in order to recover a certain theoretical model or to explain a specific experiment.

Finally, we find that

$$\begin{aligned} &m\ddot{q}(t) + (\eta + \eta_{\mathrm{eff}}[u])\dot{q}(t) = f_q(t), \text{ and} \\ &m\ddot{u}(t) + (\eta - \eta_{\mathrm{eff}}[u])\dot{u}(t) = f_u(t), \end{aligned} \tag{36}$$

where the center of mass and the relative fluctuating forces are given by $f_q(t) = (f_1 + f_2)/2$ and $f_u(t) = f_1 - f_2$. Note that, although nonlinear dissipation forces have been found, the effective bath-induced potential $V_e[u]$ from Eq. (17) could not be recovered in our modified bilinear model. This suggests that, if a given experiment reveals bath-mediated conservative forces, the theoretical model should probably start from nonlinear system-bath couplings.

IV. APPLICATIONS

The idea here is to illustrate the versatility of our main result, namely, Eq. (32). First, we recover the usual dissipation rates of both the standard nonlinear and the standard linear models. Finally, we discuss ways to avoid an anomalous diffusion that may appear, and revisit the Brownian dynamics of two particles with hydrodynamics interactions.

A. Recovering the dissipation rates

In order to recover the nonlinear dissipation $\eta_e[u]$ from Eq. (19), we choose

$$G(k, u) = \cos(ku). \tag{37}$$

We use this in Eq. (32), thus finding that

$$\eta_{\mathrm{eff}}[u] = \eta\int_0^\infty dk\; k^2\,\frac{e^{-k/k_0}}{2k_0^3}\,\cos(ku) = \eta\frac{\left(1 - 3k_0^2u^2\right)}{\left(k_0^2u^2 + 1\right)^3}. \tag{38}$$

As expected, $\eta_{\mathrm{eff}}[u] = \eta_e[u]$.

We also recover the constant dissipation rate η_{12} shown in Eq. (8), in the context of the standard bilinear model. By choosing $G(k, u) = G_0$, we find that

$$\eta_{\mathrm{eff}}[u] = \eta G_0 = \eta_{12}. \tag{39}$$

This means that our result bridges the standard bilinear and the standard nonlinear models, as far as dissipation is concerned.

B. Avoiding the anomalous diffusion

It is worth discussing the anomalous diffusion due to the specific form of $\eta_{\mathrm{eff}}[u]$ in Eq. (38). This can be seen from the diffusion coefficients appearing in the correlation functions of the Langevin forces, namely,

$$\langle f_1(t)f_2(t')\rangle = 2D_{12}(u)\delta(t-t'), \tag{40}$$

$\langle f_u(t)f_u(t')\rangle = 2D_u(u)\delta(t-t')$, and $\langle f_q(t)f_q(t')\rangle = 2D_q(u)\delta(t-t')$. The connection comes from the fact that

$$D_{12}(u) = \eta_{\mathrm{eff}}[u]k_BT. \tag{41}$$

Similarly, we also find

$$D_u(u) = 2(\eta - \eta_{\mathrm{eff}}[u])k_BT, \tag{42}$$

and

$$D_q(u) = \frac{(\eta + \eta_{\text{eff}}[u])}{2} k_B T. \tag{43}$$

The anomalous diffusion arises from

$$\eta_{\text{eff}}[|u| > k_0^{-1}/\sqrt{3}] < 0, \tag{44}$$

implying a reduction in the diffusion coefficient D_q at intermediate separations, as compared to arbitrarily far apart Brownian particles, as well as an anticorrelation ($D_{12} < 0$) between the Langevin forces acting on the particles.

If we are interested in describing a system where such anomalous diffusion is absent, we can tune $G(k, u)$ accordingly. To be concrete, let us take the example of the spectral functions used to model localized excitons interacting with a bath of acoustic phonons [22,23]. Their typical Gaussian features motivate us to set

$$G(k, u) = e^{-\frac{k}{k_0}(k_0 u)^2}. \tag{45}$$

This results in an effective dissipation rate given by

$$\eta_{\text{eff}}[u] = \frac{\eta}{\left(1 + k_0^2 u^2\right)^3}. \tag{46}$$

That is, Eq. (45) guarantees that $\eta_{\text{eff}}[u] \geqslant 0$, and also recovers the independent Brownian motions of two arbitrarily distant particles (in the limit of $|u| \to \infty$). In fact, any positive and convergent function [i.e., $G(k, u) \geqslant 0$ and $G(k, |u| \to \infty) = 0$] is sufficient to guarantee that $\eta_{\text{eff}}[u] \geqslant 0$, and that $\eta_{\text{eff}}[|u| \to \infty] = 0$.

Because our main point in this paper is to reconcile the bilinear and the nonlinear models as far as possible, it is worth highlighting that avoiding the anomalous diffusion is also possible in the standard nonlinear model. For that, one could tune $g(k)$ in Eq. (21) when applying it to Eq. (38). For instance, making $g(k) = k^{-2} k_0^{-1} \exp(-k/k_0)$ yields $\eta_{\text{eff}}[u] = \eta(1 + (k_0 u)^2)^{-1} > 0$. One advantage we see in our modified bilinear model is that it allows us to replace $\cos(ku)$ by some other function of u in Eq. (38). To our understanding, this provides a more practical means to recover the phenomenological dissipation rate under scrutiny.

C. Hydrodynamics-inspired model

In disagreement with the standard nonlinear model, our approach does not predict an effective potential between the particles, so that only bath-induced dissipation rates arise. Does it mean that our modified bilinear model is physically flawed? On the contrary, here we revisit a model of Brownian particles with hydrodynamic interactions [19], an instance that illustrates this feature of our model.

Let us consider two Brownian particles immersed in a viscous fluid. The fluid can mediate interactions between the particles, so that the dissipative forces may depend on the interparticle distance, as shown in Ref. [19]. Here, we restrict our discussion to the limit where the radius of each Brownian particle is vanishingly small as compared to their relative distance (i.e., the regime of validity of the so called Oseen tensor). We also assume the one-dimensional limit of the Brownian motion. In that case, an effective dissipation rate mediated by the hydrodynamic environment, as derived with the help of the fluctuation-dissipation relation, is given by Ref. [19] (see Appendix C)

$$\eta_{\text{eff}}^{\text{hydro}}[u] \approx \gamma_h |u|, \tag{47}$$

where γ_h is a constant proportional to the solvent viscosity.

To model Eq. (47), we can simply choose

$$G(k, u) = k|u|, \tag{48}$$

since

$$\eta_{\text{eff}}[u] = \eta \int_0^\infty dk\, k^2 \frac{e^{-\frac{k}{k_0}}}{2k_0^3} k|u| = \gamma_m |u|, \tag{49}$$

with $\gamma_m = 3\eta k_0$.

We have thus shown that an environment-mediated dissipation of hydrodynamic nature can be mapped into a fictitious bath of harmonic oscillators with no need for nonlinear system-environment couplings. In other words, we can in principle always find the appropriate $G(k, u)$ able to reproduce the phenomenological behavior of interest, including more sophisticated hydrodynamic models than the one studied here.

V. CONCLUSIONS

In summary, we have widened the degree of applicability of the bilinear model for the dynamics of two Brownian particles in a collective environment. In particular, we have reconciled a controversy between the bilinear and the nonlinear approaches, namely, whether distance-dependent dissipation forces mediated by the environment, and affecting both the center of mass and the relative coordinate, should exist or not, and in which circumstances.

Our main result was the derivation of the effective dissipation rate $\eta_{\text{eff}}[u]$ departing from a bilinear Lagrangian. Our method was based on the introduction of a distance-dependent nonlinear spectral function (response function) $\chi_k^{(ij)}(\omega, u)$, which settles a length scale to the dynamics of the Brownian particles (as also achieved with the standard nonlinear model, but not with the standard bilinear model). This allowed us to recover the dissipation forces as derived both from the standard bilinear and the standard nonlinear models. We have also discussed the physical meaning of $\chi_k^{(ij)}(\omega, u)$ in terms of a nonlinear response theory of the bath perturbed by the Brownian particles.

As applications, we have discussed some consequences of tuning our distance-dependent spectral function. For instance, a change from a trigonometric function to a Gaussian in $G(k, u)$ made it possible to avoid the anomalous diffusion presented in the original nonlinear model. Also, we described hydrodynamic correlations between a pair of Brownian particles in a viscous fluid by means of our phenomenological distance-dependent spectral function. These examples illustrate how our results represent a simple and versatile way to express diverse nonlinear dissipative forces in the dynamics of pairs of Brownian particles.

As a perspective, we would like to generalize our distance-dependent spectral function to a larger number of Brownian degrees of freedom in a common environment (N particles in a three-dimensional space). We believe it to be a feasible goal, given the pairwise character that typically underlies effective interactions. This could allow us to characterize relaxation processes and entropy production in nonequilibrium dissipative many-body systems, across classical and quantum regimes [17]. For instance, we could think of generalizing a recent study concerning entropy production of a single quantum Brownian particle [16].

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APPENDIX A: TWO-TIME CORRELATION FUNCTIONS AND THE FLUCTUATION DISSIPATION THEOREM

From the condition of thermal equilibrium, we have the following identities:

$$\langle \tilde{R}_k(0)\rangle = 0, \quad \langle \dot{R}_k(0)\rangle = 0, \tag{A1}$$

$$\langle \tilde{R}_k(0)\dot{R}_{k'}(0)\rangle = 0, \quad \langle \dot{R}_k(0)\tilde{R}_{k'}(0)\rangle = 0, \tag{A2}$$

$$\langle \dot{R}_k(0)\dot{R}_{k'}(0)\rangle = \frac{k_BT}{m_k}\delta_{kk'}, \quad \langle \tilde{R}_k(0)\tilde{R}_{k'}(0)\rangle = \frac{k_BT}{m_k\omega_k^2}\delta_{kk'}, \tag{A3}$$

where k_B is the Boltzmann constant. As defined in the main text, the formal expressions for the fluctuating forces are given by

$$f_i(t) = -\sum_k C_k^{(i)}\left[\dot{R}_k(0)\frac{\sin\omega_k t}{\omega_k} + \tilde{R}_k(0)\cos\omega_k t\right], \tag{A4}$$

where the displaced equilibrium positions of the oscillators (due to their couplings with the particles) are $\tilde{R}_k(0) = R_k(0) + (C_k^{(i)}x_i(0) + C_k^{(j)}x_j(0))(m_k\omega_k^2)^{-1}$. We have also defined $f_q = (f_1 + f_2)/2$, and $f_u = f_1 - f_2$. With the above expressions at hands, we obtain the general form for the two-time correlation functions,

$$\langle f_\alpha(t)f_\beta(t')\rangle = 2D_{\alpha\beta}(u)\delta(t-t'), \tag{A5}$$

where $D_{\alpha\beta}(u)$ is a type of diffusion coefficient having a different form according to the choice of forces we are dealing with. To explicitly compute them, we apply the continuum limit in the same way we did in Sec. III. We thus find Eqs. (41), (42), and (43) as results.

For the sake of completeness, and also to highlight that Eq. (A5) is indeed a general form of the fluctuation-dissipation theorem, we recall the original version of the theorem below. The Langevin equation for a free Brownian particle reads

$$m\dot{v}(t) = -\eta v(t) + F(t), \tag{A6}$$

where η is a friction coefficient, and $F(t)$ is a fluctuating force caused by collisions of the particle with the atoms of the surrounding fluid. The fluctuating force fulfills

$$\langle F(t)\rangle = 0, \qquad \text{and } \langle F(t)F(t')\rangle = 2D\delta(t-t'), \tag{A7}$$

where D can be seen as a measure of the strength of the fluctuating force. The δ function in time indicates that there is no correlation between impacts at any distinct time intervals. The solution for the linear, first-order, and inhomogeneous differential equation reads [1]

$$v(t) = e^{-\eta t/m}v(0) + \int_0^t dt' e^{-\eta(t-t')/m}F(t')/m. \tag{A8}$$

We can get the mean-squared velocity and evaluate for long times, thus

$$\langle v^2(\infty)\rangle = \frac{D}{\eta m}. \tag{A9}$$

At thermal equilibrium, $\langle v^2\rangle_{\text{eq}} = k_BT/m$ (equipartition theorem), hence

$$D = \eta k_B T. \tag{A10}$$

This relates the strength D of the random noise, or fluctuating force, to the magnitude η of the friction, or dissipation rate, explaining why it is known as the fluctuation-dissipation theorem. It expresses the balance between friction and noise that is required to have thermal equilibrium state at long times.

APPENDIX B: RESPONSE THEORY

Here we set out to discuss the physical meaning of our phenomenological response function as defined in Eq. (29).

1. Linear response theory of an environment perturbed by a single particle

First, we revisit Caldeira [3], so as to provide a more detailed, self-contained discussion on the standard single-particle scenario (cf. p. 107 of Ref. [3]). The bath's equations of motion for a single Brownian particle with linear couplings are given by

$$m_k\ddot{R}_k + m_k\omega_k^2R_k - C_kx = 0, \tag{B1}$$

which gives that

$$\tilde{R}_k = -\frac{C_k}{m_k\left(\omega^2 - \omega_k^2\right)}\tilde{x}(\omega) \tag{B2}$$

for each mode with frequency ω. An effective (and collective) coordinate for the environment has been defined in Ref. [3] as the linear combination

$$R_{\text{eff}} \equiv \sum_k C_k\tilde{R}_k. \tag{B3}$$

It can also be decomposed in the Fourier space as

$$R_{\text{eff}} = \chi_{\text{env}}(\omega)\tilde{x}(\omega), \tag{B4}$$

where the susceptibility is defined as

$$\chi_{\mathrm{env}}(\omega) \equiv -\sum_k \frac{C_k^2}{m_k\left(\omega^2-\omega_k^2\right)}. \tag{B5}$$

The imaginary part of $\chi_{\mathrm{env}}(\omega)$ comes from replacing

$$\omega \pm \omega_k \rightarrow \omega \pm \omega_k + i\epsilon, \;\; (\text{with } \epsilon \rightarrow 0), \tag{B6}$$

and using identity

$$\frac{1}{\omega \pm \omega_k + i\epsilon} = \mathcal{P}\left(\frac{1}{\omega \pm \omega_k}\right) - i\pi\delta(\omega \pm \omega_k). \tag{B7}$$

It thus follows that

$$\mathrm{Im}\,\chi_{\mathrm{env}}(\omega) \equiv \chi''_{\mathrm{env}}(\omega) \tag{B8}$$

$$= \frac{\pi}{2}\sum_k \frac{C_k^2}{m_k\omega_k}[\delta(\omega-\omega_k)+\delta(\omega+\omega_k)]. \tag{B9}$$

Since ω and $\omega_k > 0$, we get the spectral function,

$$\chi''_{\mathrm{env}}(\omega) = J(\omega) = \frac{\pi}{2}\sum_k \frac{C_k^2}{m_k\omega_k}\delta(\omega-\omega_k), \tag{B10}$$

in agreement with Eq. (7). This concludes our revision of Ref. [3]. In what follows, we address an issue that has not been discussed so far, to the best of our knowledge.

2. Nonlinear response theory of an environment perturbed by a couple of particles within the nonlinear couplings model

What should be the effective (collective) coordinate of the environment R_{eff}, as analogous to that defined in Eq. (B3), in the case of nonlinear system-bath couplings? As a matter of fact, the equations of motion for the bath modes, namely,

$$m_k\ddot{R}_{-k} + m_k\omega_k^2 R_{-k} + C_{-k}(x_1) + C_{-k}(x_2) = 0, \;\; \text{and} \tag{B11}$$

$$m_k\ddot{R}_k + m_k\omega_k^2 R_k + C_k(x_1) + C_k(x_2) = 0, \tag{B12}$$

imply that Eqs. (B2) and (B3) no longer apply. The function $C_k(x_i)$ mixes the Fourier components of the particle's coordinates, $\tilde{x}_i(\omega)$, generally in a nonlinear manner. Physically speaking, if the environment causes on the pair of particles a dissipative force that nonlinearly depends on the interparticle distance, we expect as a reaction that the particles disturb the environment in a similar manner, thus making R_{eff} to depend nonlinearly on the interparticle distance.

To address this question, we assume that $R_k(t) = \tilde{R}_k\exp(-i\omega t)$, and $C_k(x_1) + C_k(x_2) \approx [C_k(x_1^0) + C_k(x_2^0)]\exp(-i\omega t)$. This can be seen as a quasi-static (Born-Oppenheimer) approximation, in the sense that, in the limit of very slowly varying Brownian movement of the particles as compared with the fast bath dynamics ($\omega \ll \omega_k$), we have that $x_i^0 \approx x_i = x_i(t)$, and $\exp(-i\omega t) \approx 1$. Such approximations are equivalent to the low-frequency limit employed when assuming the Ohmic regime. We thus get that

$$\tilde{R}_{-k} \approx -\frac{C_{-k}(x_1) + C_{-k}(x_2)}{m_k\left(\omega^2-\omega_k^2\right)}, \;\; \text{and} \tag{B13}$$

$$\tilde{R}_k \approx -\frac{C_k(x_1) + C_k(x_2)}{m_k\left(\omega^2-\omega_k^2\right)}. \tag{B14}$$

By extrapolating Eq. (B3), we define an effective (collective) coordinate of the environment as a linear combination of $\tilde{R}_k'$s with coefficients now given by $C_{\pm k}(x_i)$, that is,

$$R_{\mathrm{eff}} \equiv \sum_k (\tilde{R}_k(C_{-k}(x_1) + C_{-k}(x_2)) + \tilde{R}_{-k}(C_k(x_1) + C_k(x_2))). \tag{B15}$$

This definition allows us to find the emergence of both "$\cos(ku)$" and "$\delta(\omega-\omega_k)$" terms in the dynamics of the effective environment, as they appear in Eqs. (13) and (14). Indeed,

$$R_{\mathrm{eff}} = -2\sum_k \frac{\kappa_k\kappa_{-k}}{m_k\left(\omega^2-\omega_k^2\right)}(1 + 2\cos ku), \tag{B16}$$

after having made $C_k(x_i) = \kappa_k e^{ikx_i}$ in Eq. (B15). We find "$\delta(\omega-\omega_k)$" when we apply Eq. (B7) to (B16). By expanding R_{eff} in a power series of u, an effective nonlinear susceptibility $\chi_{\mathrm{env},u}(\omega, u)$ shows up,

$$R_{\mathrm{eff}} = \chi_{\mathrm{env},u}(\omega, u)\, u. \tag{B17}$$

In the following, we build on this line of thought in order to explain the physical meaning of our nonlinear susceptibility, as we have introduced it in Eq. (29).

3. Nonlinear response theory of an environment perturbed by a couple of particles within the bilinear couplings model

An analogy with the effective (collective) bath coordinate from Eq. (B15) allows us to assign a physical meaning to our nonlinear susceptibility, as given in Eq. (29). The equations of motion for the bath within the bilinear couplings model are given by

$$m_k\ddot{R}_k + m_k\omega_k^2 R_k + C_k^{(1)}x_1 + C_k^{(2)}x_2 = 0. \tag{B18}$$

Inspired by Eq. (B14), we set

$$\tilde{R}_k = -\frac{\left(C_k^{(1)}x_1 + C_k^{(2)}x_2\right)}{m_k\left(\omega^2-\omega_k^2\right)},$$

where the coordinates $x_i = x_i(t)$ now explicitly depend on time. In terms of the center of mass and the relative coordinates, we get that

$$\tilde{R}_k(q, u) = -\frac{\left(\bar{c}q + \Delta c\frac{u}{2}\right)}{m_k\left(\omega^2-\omega_k^2\right)}, \tag{B19}$$

where

$$\bar{c} = C_k^{(1)} + C_k^{(2)}, \tag{B20}$$

$$\Delta c = C_k^{(1)} - C_k^{(2)}. \tag{B21}$$

Instead of assuming that the nonlinearity arises from the nonlinear couplings $C_k(x_i)$, as in Eq. (B15), we now consider that the collective coordinate is itself a nonlinear combination of bath modes,

$$R_{\text{eff}} \equiv \sum_k \left(C_k^{(1)} F_k^{(1)}[\tilde{R}_k(q,u)] + C_k^{(2)} F_k^{(2)}[\tilde{R}_k(q,u)]\right), \tag{B22}$$

where the functions $F_k^{(i)}[\bullet]$ must be appropriately chosen so as to reproduce the phenomenological behavior of the system of interest. Without loss of generality, $F_k^{(i)}[\bullet]$ can be expanded in a power series as

$$F_k^{(1)} = \sum_{n=0}^{\infty} F_{k,n}^{(1)} \frac{[\tilde{R}_k(q,u)]^n}{n!}, \text{ and} \tag{B23}$$

$$F_k^{(2)} = \sum_{m=0}^{\infty} F_{k,m}^{(2)} \frac{[\tilde{R}_k(q,u)]^m}{m!}. \tag{B24}$$

We also assume that $F_{k,n}^{(1)}$ and $F_{k,m}^{(2)}$ can be renormalized as

$$F_{k,n}^{(1)} = f_{k,n}^{(1)}\left[m_k\left(\omega^2 - \omega_k^2\right)\right]^{n-1}, \text{ and} \tag{B25}$$

$$F_{k,m}^{(2)} = f_{k,m}^{(2)}\left[m_k\left(\omega^2 - \omega_k^2\right)\right]^{m-1}, \tag{B26}$$

so that

$$\begin{aligned} R_{\text{eff}} &= \sum_k C_k^{(1)}\left(\sum_{n=0}^{\infty} F_{k,n}^{(1)} \frac{[\tilde{R}_k(q,u)]^n}{n!}\right) + \sum_k C_k^{(2)}\left(\sum_{m=0}^{\infty} F_{k,m}^{(2)} \frac{[\tilde{R}_k(q,u)]^m}{m!}\right) \\ &= \sum_k C_k^{(1)}\left(\sum_{n=0}^{\infty} f_{k,n}^{(1)}\left(m_k\left(\omega^2-\omega_k^2\right)\right)^{n-1} \frac{[\tilde{R}_k]^n}{n!}\right) + \sum_k C_k^{(2)}\left(\sum_{m=0}^{\infty} f_{k,m}^{(2)}\left(m_k\left(\omega^2-\omega_k^2\right)\right)^{m-1} \frac{[\tilde{R}_k]^m}{m!}\right) \\ &= -\sum_k \frac{1}{m_k(\omega^2-\omega_k^2)}\left(C_k^{(1)} \sum_{n=0}^{\infty} \frac{(-1)^{n-1}}{n!} f_{k,n}^{(1)}\left[\bar{c}q + \Delta c\frac{u}{2}\right]^n + C_k^{(2)} \sum_{m=0}^{\infty} \frac{(-1)^{m-1}}{m!} f_{k,m}^{(2)}\left[\bar{c}q + \Delta c\frac{u}{2}\right]^m\right). \end{aligned} \tag{B27}$$

Using the binomial expansion $(a+b)^n = \sum_{p=0}^{n} \binom{n}{p} a^{n-p} b^p$, we obtain

$$\begin{aligned} R_{\text{eff}} &= -\sum_k \frac{1}{m_k\left(\omega^2-\omega_k^2\right)}\left[C_k^{(1)} \sum_{n=0}^{\infty} \frac{(-1)^{n-1}}{n!} f_{k,n}^{(1)} \sum_{p=0}^{n} (\bar{c}q)^{n-p}\left(\Delta c\frac{u}{2}\right)^p \binom{n}{p} + C_k^{(2)} \sum_{m=0}^{\infty} \frac{(-1)^{m-1}}{m!} f_{k,m}^{(2)} \sum_{p'=0}^{m} (\bar{c}q)^{m-p'}\left(\Delta c\frac{u}{2}\right)^{p'} \binom{m}{p'}\right] \\ &= -\sum_k \frac{1}{m_k\left(\omega^2-\omega_k^2\right)} \sum_{n'=0}^{\infty} \frac{(-1)^{n'-1}}{n'!}\left(C_k^{(1)} f_{k,n'}^{(1)} + C_k^{(2)} f_{k,n'}^{(2)}\right) \sum_{m'=0}^{n'} \binom{n'}{m'} (\bar{c}q)^{n'-m'}\left(\Delta c\frac{u}{2}\right)^{m'} \\ &= -\sum_k \frac{1}{m_k\left(\omega^2-\omega_k^2\right)} \sum_{n'=0}^{\infty} \frac{(-1)^{n'-1}}{n'!}\left(C_k^{(1)} f_{k,n'}^{(1)} + C_k^{(2)} f_{k,n'}^{(2)}\right) \sum_{m'=0}^{n'} \binom{n'}{m'} \bar{c}^{n'-m'}\left(\frac{\Delta c}{2}\right)^{m'} q^{n'-m'} u^{m'} \\ R_{\text{eff}} &= \sum_{n'=0}^{\infty} \sum_{m'=0}^{n'} \alpha_{n'm'} q^{n'-m'} u^{m'}, \end{aligned} \tag{B28}$$

where $\alpha_{n'm'}$ is defined as

$$\begin{aligned} \alpha_{n'm'} \equiv & -\sum_k \frac{(-1)^{n'-1}}{m_k\left(\omega^2-\omega_k^2\right)n!}\left(C_k^{(1)} f_{k,n'}^{(1)} + C_k^{(2)} f_{k,n'}^{(2)}\right) \\ & \times \binom{n'}{m'} \bar{c}^{n'-m'}\left(\frac{\Delta c}{2}\right)^{m'}. \end{aligned} \tag{B29}$$

By writing the effective bath coordinate as

$$R_{\text{eff}} = \left(\sum_{n'=0}^{\infty} \sum_{m'=0}^{n'} \alpha_{n'm'} q^{n'-m'} u^{m'-1}\right) u, \tag{B30}$$

we find a nonlinear susceptibility, such that

$$R_{\text{eff}} = \chi_{\text{env}}(\omega, q, u)\, u. \tag{B31}$$

Explicitly, we have that

$$\begin{aligned} \chi_{\text{env}}(\omega, q, u) = & -\sum_k \sum_{n'} \frac{(-1)^{n'-1}}{m_k\left(\omega^2-\omega_k^2\right)n!} \\ & \times \left(C_k^{(1)} f_{k,n'}^{(1)} + C_k^{(2)} f_{k,n'}^{(2)}\right) \\ & \times \sum_{m'=0}^{n'} \binom{n'}{m'} \bar{c}^{n'-m'}\left(\frac{\Delta c}{2}\right)^{m'} q^{n'-m'} u^{m'-1}. \end{aligned} \tag{B32}$$

The imaginary part of the susceptibility comes from employing Eq. (B6) and Eq. (B7), which gives that

$$\begin{aligned}\mathrm{Im}\,\chi_{\mathrm{env}}(\omega,q,u) &\equiv \chi''_{\mathrm{env},q}(\omega,q,u)\\ &= \frac{\pi}{2}\sum_{k}\frac{h'(k,q,u)}{m_k\omega_k}[\delta(\omega-\omega_k)+\delta(\omega+\omega_k)],\end{aligned} \tag{B33}$$

where we have defined

$$\begin{aligned}h'(k,q,u) \equiv \sum_{n'}\frac{(-1)^{n'-1}}{n!}\left(C_k^{(1)}f_{k,n'}^{(1)}+C_k^{(2)}f_{k,n'}^{(2)}\right)\\ \sum_{m'=0}^{n'}\binom{n'}{m'}\bar{c}^{n'-m'}\left(\frac{\Delta c}{2}\right)^{m'}q^{n'-m'}u^{m'-1}.\end{aligned} \tag{B34}$$

Since ω and $\omega_k > 0$, only $\delta(\omega-\omega_k)$ remains in Eqs. (B34), as usual. Finally, to keep consistency with our choice in Eq. (30), we set

$$h'(k,q,u) = h(k,u), \tag{B35}$$

showing how our distance-dependent susceptibility,

$$\mathrm{Im}\,\chi_k^{(ij)}(\omega) = \chi''_{\mathrm{env}}(\omega,u), \tag{B36}$$

results from a nonlinear response theory of a bath of oscillators perturbed by a pair of Brownian particles. In summary, both in the standard nonlinear couplings model and in our bilinear model with a modified spectral function, the bath of oscillators reacts nonlinearly when subjected to the disturbance provided by the pair of Brownian particles.

APPENDIX C: DISSIPATION RATE OF TWO BROWNIAN PARTICLES SHARING A HYDRODYNAMIC ENVIRONMENT

Here we discuss the dissipation rate we used in Eq. (47). In Ref. [19], we find that the so-called Oseen tensor is given by

$$\mathcal{D}_{ij} = \frac{k_B T}{8\pi\kappa r_{ij}}\left(\mathbb{I}+\frac{\vec{r}_{ij}\vec{r}_{ij}}{r_{ij}^2}\right), \tag{C1}$$

for particles i and j, with $i \neq j$. Here, κ is the fluid viscosity, $\mathbb{I}$ the identity matrix, $\vec{r}_{ij}$ is the vector linking the centers of each particle, and $r_{ij} = |\vec{r}_{ij}|$. Because $\mathcal{D}_{ij}$ describes diffusion in configuration space, its dimensions are $[\mathcal{D}_{ij}] = [L]^2[T]^{-1}$. The fluid viscosity has dimensions of $[\kappa] = [M][L]^{-1}[T]^{-1}$. The dimension of the dissipation rate in our model is $[\eta_{\mathrm{eff}}] = [M][T]^{-1}$ (see Sec. III). Hence, $[\kappa][r_{ij}] = [M][T]^{-1} = [\eta_{\mathrm{eff}}^{\mathrm{hydro}}]$. Guided by this dimensional analysis, we can infer how the Oseen tensor provides us with an effective (tensorial) dissipation rate depending on the relative coordinate for a pair of Brownian particles in a fluid, namely,

$$\mathcal{D}_{ij} = \frac{k_B T}{\eta_{\mathrm{eff}}^{\mathrm{hydro}}[\vec{r}_{ij}]}, \tag{C2}$$

where

$$\eta_{\mathrm{eff}}^{\mathrm{hydro}}[\vec{u}] \equiv 8\pi\kappa|\vec{u}|\left(\mathbb{I}+\frac{\vec{u}\vec{u}}{u^2}\right)^{-1}. \tag{C3}$$

In the one-dimensional model of the Brownian motion, we make $\vec{u} \to u$, and $\mathbb{I} \to 1$. Our effective dissipation rate mediated by the hydrodynamic environment is thus found to be

$$\eta_{\mathrm{eff}}^{\mathrm{hydro}}[u] = 4\pi\kappa|u| = \gamma_h|u|, \tag{C4}$$

where $\gamma_h = 4\pi\kappa$.

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