Irreversible dynamics of a massive intruder in dense granular fluids

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Abstract – A Generalized Langevin Equation with exponential memory is proposed for the dynamics of a massive intruder in a dense granular fluid. The model reproduces numerical correlation and response functions, violating the Equilibrium Fluctuation-Dissipation Relations. The source of memory is identified in the coupling of the tracer velocity V with a spontaneous local velocity field U in the surrounding fluid: fluctuations of this field introduce a new time scale with its associated length scale. Such identification allows us to measure the intruder's fluctuating entropy production as a function of V and U, obtaining a neat verification of the fluctuation relation.

Models of granular fluids are a natural framework where the issues of non-equilibrium statistical mechanics can be addressed [1]. Due to dissipative interactions among the microscopic constituents, energy is not conserved and external sources are necessary in order to maintain a stationary state. Heat fluxes and currents continuously pass through the system, time reversal invariance is broken and consequently, properties such as the Equilibrium Fluctuation-Dissipation Relation (EFDR) do not hold. In recent years, a rather complete theory, at least in the dilute limit, has been developed and numerous aspects have been clarified, in good agreement with numerical simulations [2,3]. However, a general understanding of dense granular fluids is still lacking. A common approach is the so-called Enskog correction [2,4], which reduces the breakdown of molecular chaos to a renormalization of the collision frequency. In cooling regimes, the Enskog theory may describe strong non-equilibrium effects, due to the explicit cooling-time–dependence [5]. However it cannot describe dynamical effects in stationary regimes, such as large violations of the Einstein relation [6,7].

In this letter, we propose a model for the dynamics of a massive tracer moving in a gas of smaller granular particles, both coupled to an external bath. In particular, taking as reference point the dilute limit, where the system has a closed analytical description [8], we suggest a Generalized Langevin Equation (GLE) with an exponential memory kernel as first approximation capable of

describing the dense case. Here, the main features are: i) the decay of correlation and response functions is not simply exponential and shows backscattering [9,10] and ii) the EFDR [11,12] of the first and second kind do not hold. In the model we propose, detailed balance is not necessarily satisfied, non-equilibrium effects can be taken into account and the correct behavior of correlation and response functions is predicted. Furthermore, the model has a remarkable property: it can be mapped onto a two-variable Markovian system, *i.e.* two coupled Langevin equations with simple white noises. The auxiliary variable can be identified in the local velocity field spontaneously appearing in the surrounding fluid. This allows us to measure the fluctuating entropy production [13], and fairly verify the fluctuation relation [12,14,15]. This is a remarkable result, if considered the interest of the community [16] and compared with unsuccessful past attempts [17,18].

We consider an "intruder" disc of mass $m_0 = M$ and radius R, moving in a gas of N granular discs with mass $m_i = m$ $(i > 0)$ and radius r, in a two-dimensional box of area $A = L^2$. We denote by $n = N/A$ the number density of the gas and by ϕ the occupied volume fraction, *i.e.* $\phi = \pi(Nr^2 + R^2)/A$ and we denote by $\boldsymbol{V}$ (or $\boldsymbol{v}_0$) and $\boldsymbol{v}$ (or $\boldsymbol{v}_i$ with $i > 0$) the velocity vector of the tracer and of the gas particles, respectively. Interactions among the particles are hard-core binary instantaneous inelastic collisions, such that particle i, after a collision with particle j, comes out with a velocity

$$\boldsymbol{v}_i' = \boldsymbol{v}_i - (1+\alpha)\frac{m_j}{m_i + m_j}[(\boldsymbol{v}_i - \boldsymbol{v}_j)\cdot\hat{\boldsymbol{n}}]\hat{\boldsymbol{n}}, \qquad (1)$$

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where $\hat{\boldsymbol{n}}$ is the unit vector joining the particles' centers of mass and $\alpha \in [0,1]$ is the restitution coefficient ($\alpha = 1$ is the elastic case). The mean free path of the intruder is proportional to $l_0 = 1/(n(r+R))$ and we denote by τ_c its mean collision time. Two kinetic temperatures can be introduced for the two species: the gas granular temperature $T_g = m\langle \boldsymbol{v}^2 \rangle/2$ and the tracer temperature $T_{tr} = M\langle \boldsymbol{V}^2 \rangle/2$.

In order to maintain a granular medium in a fluidized state, an external energy source is coupled to each particle in the form of a thermal bath [19–21] (hereafter, exploiting isotropy, we consider only one component of the velocities):

$$m_i \dot{v}_i(t) = -\gamma_b v_i(t) + f_i(t) + \xi_b(t). \tag{2}$$

Here $f_i(t)$ is the force taking into account the collisions of particle i with other particles, and $\xi_b(t)$ is a white noise (different for all particles), with $\langle \xi_b(t) \rangle = 0$ and $\langle \xi_b(t)\xi_b(t') \rangle = 2T_b\gamma_b\delta(t-t')$. The effect of the external energy source balances the energy lost in the collisions and a stationary state is attained with $m_i\langle v_i^2 \rangle \leqslant T_b$.

For low packing fractions, $\phi \lesssim 0.1$, and in the large mass limit, $m/M \ll 1$, using the Enskog approximation it has been shown [8] that the dynamics of the intruder is described by a linear Langevin equation. In this limit the velocity autocorrelation function shows a simple exponential decay, with characteristic time M/Γ_E, where

$$\Gamma_E = \gamma_b + \gamma_g^E, \quad \text{with} \quad \gamma_g^E = \frac{g_2(r+R)}{l_0}\sqrt{2\pi m T_g}(1+\alpha), \tag{3}$$

and $g_2(r+R)$ is the pair correlation function for a gas particle and the intruder at contact. Time-reversal and the EFDR, which are very weakly modified for uniform dilute granular gases [6,22,23], become perfectly satisfied for a massive intruder. The temperature of the tracer is computed as $T_{tr}^E = (\gamma_b T_b + \gamma_g^E \frac{1+\alpha}{2} T_g)/\Gamma_E$. For a general study of a Langevin equation with "two temperatures" but a single time scale (which is always at equilibrium), see also [24].

As the packing fraction is increased, the Enskog approximation is less and less effective in predicting the memory effects and the dynamical properties of the system. In particular, velocity autocorrelation $C(t) = \langle V(t)V(0) \rangle/\langle V^2 \rangle$ and linear response function $R(t) = \delta V(t)/\delta V(0)$ (*i.e.* the mean response at time t to an impulsive perturbation applied at time 0) present an exponential decay modulated in amplitude by oscillating functions [10]. Moreover violations of the EFDR $C(t) = R(t)$ (Einstein relation) are observed for $\alpha < 1$ [7,25].

Molecular-dynamics simulations of the system have been performed by means of a standard event-driven algorithm to treat hard core interactions: the algorithm is supplemented with a "driving event" at times which are multiples of a small timestep (smaller than all time scales) which update the velocity of all particles by a
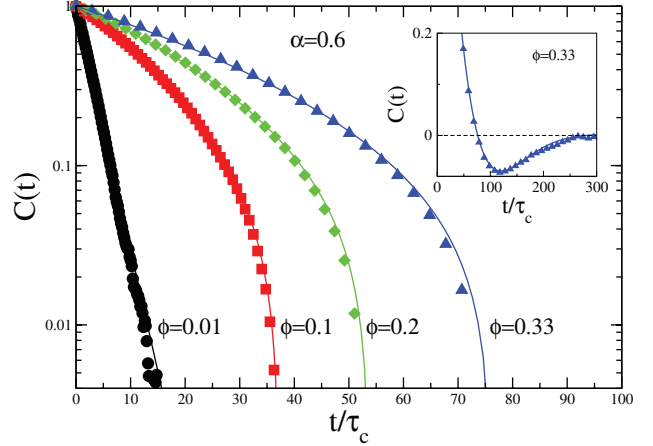

Fig. 1: (Color online) Semi-log plot of $C(t)$ (symbols) for different values of $\phi = 0.01, 0.1, 0.2, 0.33$ at $\alpha = 0.6$. Times are rescaled by the mean collision time τ_c. Continuous lines are the best fits obtained with eqs. (9). Inset: $C(t)$ and the best fit in linear scale for $\phi = 0.33$ and $\alpha = 0.6$.

discretized version of eq. (2). In the simulations we have measured $C(t)$ and $R(t)$, for several different values of the parameters α and ϕ. In fig. 1 symbols correspond to the velocity correlation functions measured in the inelastic case, $\alpha = 0.6$, for different values of the packing fraction ϕ. The other parameters are fixed: $N = 2500$, $m = 1$, $M = 25$, $r = 0.005$, $R = 0.025$, $T_b = 1$, $\gamma_b = 200$.

Notice that the Enskog approximation [2,8] cannot predict the observed functional forms, because it only modifies by a constant factor the collision frequency. In order to describe the full phenomenology, a model with more than one characteristic time is needed. As a first proposal, we consider a Langevin equation with a single exponential memory kernel [26,27]

$$M\dot{V}(t) = -\int_{-\infty}^{t} dt' \, \Gamma(t-t')V(t') + \mathcal{E}'(t), \tag{4}$$

where

$$\Gamma(t) = 2\gamma_0\delta(t) + \gamma_1/\tau_1 e^{-t/\tau_1} \tag{5}$$

and $\mathcal{E}'(t) = \mathcal{E}_0(t) + \mathcal{E}_1(t)$, with

$$\langle \mathcal{E}_0(t)\mathcal{E}_0(t') \rangle = 2T_0\gamma_0\delta(t-t'), \tag{6}$$

$$\langle \mathcal{E}_1(t)\mathcal{E}_1(t') \rangle = T_1\gamma_1/\tau_1 e^{-(t-t')/\tau_1} \tag{7}$$

and $\langle \mathcal{E}_1(t)\mathcal{E}_0(t') \rangle = 0$. In the limit $\alpha \to 1$, the parameter T_1 is meant to tend to T_0 in order to fulfill the EFDR of the 2nd kind $\langle \mathcal{E}'(t)\mathcal{E}'(t') \rangle = T_0\Gamma(t-t')$. Within this model the dilute case is recovered if $\gamma_1 \to 0$. In this limit, the parameters γ_0 and T_0 coincide with Γ_E and T_{tr}^E of the Enskog theory [8].

The exponential form of the memory kernel can be justified within the mode-coupling approximation scheme. In this framework [28], it can be written as a sum of two contributions: $\Gamma(t-t') = \beta_1\delta(t-t') + \beta_2\tilde{\Gamma}(t-t')$,

Table 1: Parameters of model (10), as obtained by fitting the numerical data (see text for details).

α	ϕ	T_{tr}	T_g	γ_0/M	T_0	T_1	γ_1/M	τ_1/τ_c	Γ_E/M	γ_g^E/M	T_{tr}^E	T_g^E
1.0	0.33	1.00	1.00	55	0.99	1.0	44	67	55	47	1.00	1.00
0.8	0.33	0.92	0.90	47	0.91	1.0	42	68	48	40	0.84	0.89
0.7	0.33	0.88	0.86	45	0.85	1.0	41	74	45	37	0.78	0.86
0.6	0.33	0.86	0.84	44	0.82	1.1	43	89	42	34	0.73	0.83
0.6	0.20	0.92	0.91	27	0.90	1.0	26	54	24	16	0.82	0.91
0.6	0.10	0.95	0.96	17	0.95	0.99	12	29	15	7	0.89	0.96
0.6	0.01	0.99	1.00	9.6	0.99	–	0	2.8	8.6	0.6	0.98	0.99
0.6	0.01*	0.88	0.94	21	0.88	–	0	21	20	12	0.85	0.93

where β_1 and β_2 are model-dependent coefficients, and $\tilde{\Gamma}(t-t')$ is a sum over modes q of $p(q)e^{-(\nu+D)q^2(t-t')}$, where $p(q)$ weights the modes relevant for the dynamics of the tracer. Here D and ν are the diffusion coefficient and the kinematic viscosity of the fluid, respectively. Following an old recipe [26], tested with success in equilibrium contexts, we assume that, for not too high packing fractions, memory arises due to re-collisions within a limited region at distance $\sim \lambda_1$ around the tracer and that this can be modeled by an effective $p(q)$ which is peaked around $q_1 = 2\pi/\lambda_1$, i.e. a single mode contributes to the sum, yielding $\tilde{\Gamma}(t-t') \sim e^{-(\nu+D)q_1^2(t-t')}$ and then

$$\tau_1 = \lambda_1^2 (2\pi)^{-2} (\nu + D)^{-1} \sim \tau_c^g (\lambda_1/l_0^g)^2, \qquad (8)$$

with τ_c^g and l_0^g the fluid mean free time and mean free path respectively. Equation (8) relates the time scale τ_1, characterizing the tail of the memory kernel, with a typical length scale λ_1 present in the system. This length scale will turn out to play a central role in the following.

The model (4) predicts $C = f_C(t)$ and $R = f_R(t)$ with

$$f_{C(R)} = e^{-gt}[\cos(\omega t) + a_{C(R)} \sin(\omega t)]. \qquad (9)$$

g, ω, a_C and a_R are known algebraic functions of $\gamma_0, T_0, \gamma_1, \tau_1$ and T_1. In particular, the ratio $a_C/a_R = [T_0 - \Omega(T_1 - T_0)]/[T_0 + \Omega(T_1 - T_0)]$, with $\Omega = \gamma_1/[(\gamma_0 + \gamma_1)(\gamma_0/M\tau_1 - 1)]$. Hence, in the elastic ($T_1 \to T_0$) as well as in the dilute ($\gamma_1 \to 0$) limit, one gets $a_C = a_R$ and recovers the EFDR $C(t) = R(t)$. In fig. 1 the continuous lines show the result of the best fits obtained using eqs. (9) for the correlation function, at restitution coefficient $\alpha = 0.6$ and for different values of the packing fraction ϕ. The functional form fits the numerical data very well.

Looking for an insight of the relevant physical mechanisms underlying such a phenomenology and in order to make the meaning of the parameters clear, it is useful to map eq. (4) onto a Markovian equivalent model by introducing an auxiliary field [29]:

$$M\dot{V} = -\gamma_0(V - U) + \sqrt{2T_0\gamma_0}\mathcal{E}_V,$$
$$\dot{U} = -\frac{U}{\tau_1} - \frac{\gamma_1}{\gamma_0\tau_1}V + \sqrt{2\frac{T_1\gamma_1}{\gamma_0^2\tau_1^2}}\mathcal{E}_U, \qquad (10)$$

where $\mathcal{E}_V$ and $\mathcal{E}_U$ are white noises of unitary variance. The variable

$$U(t) \propto \gamma_1/(\tau_1\gamma_0)\int_{-\infty}^t e^{-\frac{t-t'}{\tau_1}}[V(t') + \mathcal{E}_1(t')]\mathrm{d}t' \qquad (11)$$

is determined up to a multiplicative factor, as can be checked by direct substitution. In the chosen form (10), the dynamics of the tracer is remarkably simple: indeed V follows a memoryless Langevin equation in a *Lagrangian frame* with respect to a local field U. In the dilute limit this is exact (see Appendix of [8]) if U is the *local average velocity field* of the gas particles colliding with the tracer. Extrapolating such an identification to higher densities, we are able to both assign a meaning and predict a value for most of the parameters of the model: 1) the self-drag coefficient of the intruder in principle is not affected by the change of reference to the Lagrangian frame, so that $\gamma_0 \sim \Gamma_E$; 2) for the same reason $T_0 \sim T_{tr}$ is roughly the temperature of the tracer; 3) τ_1 is the main relaxation time of the average velocity field U around the Brownian particle; 4) γ_1 is the intensity of coupling felt by the surrounding particles after collisions with the intruder; 5) finally T_1 is the "temperature" of the local field U, easily identified with the bath temperature $T_1 \sim T_b$: indeed, thanks to momentum conservation, inelasticity does not affect the average velocity of a group of particles almost only colliding among themselves.

To find a confirmation of the above hypothesis, we have explored the region of the space of parameters $\alpha \in [0.6, 1]$ and $\phi \in [0.01, 0.33]$. From the simultaneous fit of the numerical data for correlation and response functions against eqs. (9) we can determine the set of parameters $\{g, \omega, a_C, a_R, \langle V^2 \rangle\}$. Then, by inverting the relations between them and the set $\{\gamma_0, T_0, \gamma_1, \tau_1, T_1\}$, we are eventually able to determine all the parameters entering (4). In table 1 such values are reported, together with the predictions given by the Enskog approximation (last four columns). The statistical error on these values is about 1%. We used the external parameters mentioned before, changing α or the box area A (to change ϕ): this makes the limit $\phi \to 0$ equivalent to $\gamma_g \sim 1/l_0 \to 0$ ("super-dilute" limit). The last row reports about the true dilute limit: i.e. R is reduced, at fixed l_0 (equal to

the value of the previous case $\phi = 0.2$), in order to get $\phi = 0.01$ and $\gamma_g > 0$. Notice that in the two dilute cases the simple Langevin equation is recovered ($\gamma_1 = 0$) and the dependence on the parameter T_1 disappears. Remarkably our predictions $\gamma_0 \sim \Gamma_E$, $T_0 \sim T_{tr}$ and $T_1 \sim T_b$ are fairly verified. The coupling time τ_1 increases with the packing fraction and, weakly, with the inelasticity. In the most dense cases it appears that $\gamma_1 \sim \gamma_g^E \propto \phi$: this is confirmed in the "super-dilute" limit, but cannot hold in the dilute one, where $\gamma_1 \to 0 \ll \gamma_g^E$. It is also interesting to notice that at high density $T_{tr} \sim T_g \sim T_g^E$, which is probably due to the stronger correlations among particles. Finally we notice that, at large ϕ, $T_{tr} > T_{tr}^E$, which is coherent with the idea that correlated collisions dissipate *less* energy.

A fundamental feature of this model is its ability to reproduce violations of EFDR. In fig. 2, we plot the correlation and response functions in a dense case (elastic and inelastic): symbols correspond to numerical data and continuous lines to the best-fit curves. In the inelastic case, deviations from EFDR $R(t) = C(t)$ are clearly observed. In the inset of fig. 2 the ratio $R(t)/C(t)$ is also reported. It is interesting to note that a relation between the response and correlations measured in the unperturbed system still exists, but —in the non-equilibrium case— must take into account the contribution of the cross correlation $\langle V(t)U(0)\rangle$, *i.e.*:

$$R(t) = aC(t) + b\langle V(t)U(0)\rangle \tag{12}$$

with $a = [1 - \gamma_1/M(T_0 - T_1)\Omega_a]$ and $b = (T_0 - T_1)\Omega_b$, where Ω_a and Ω_b are known functions of the parameters (see for instance [29]). At equilibrium, where $T_0 = T_1$, the EFDR is recorvered.

The mathematical definition of the auxiliary variable U, eq. (11), which requires the knowledge of a part of the noise $\mathcal{E}_1$, makes it very difficult to be measured in simulations or in experiments. But the above discussion has shown that U represents a spontaneous local velocity field interacting with the tracer: therefore it can be measured in the following manner. We fix a distance l and average the velocity of the gas particles within a circle $\mathcal{C}_l$ of radius $l + R$ centered on the tracer. In this way we define $U_l = 1/N_l \sum_{i \in \mathcal{C}_l} v_i$, where N_l is the number of particles in $\mathcal{C}_l$. Two methods are available to estimate the correct length l^*, which is difficult to be predicted on a general ground. A first guess is provided by identifying it with λ_1, which can be obtained by inverting eq. (8) after having measured τ_1, using the known values of D and ν in a granular fluid. The second method is to measure the correlations $\langle VU_l \rangle$ and $\langle U_l^2 \rangle$ and find the best value l_{cor} such that $\langle VU_{l_{cor}} \rangle \sim \langle VU \rangle$ and $\langle U_{l_{cor}}^2 \rangle \sim \langle U^2 \rangle$ (where $\langle VU \rangle$ and $\langle U^2 \rangle$ are easily computed from the model, once all the parameters have been determined fitting $C(t)$ and $R(t)$). Remarkably, the two estimates give compatible results and identify a narrow range of values for $l^* \sim \lambda_1 \sim l_{cor}$. Hence, one can identify $U \sim U_{l^*}$ and the auxiliary variable can be directly measured in numerical simulations and experiments.
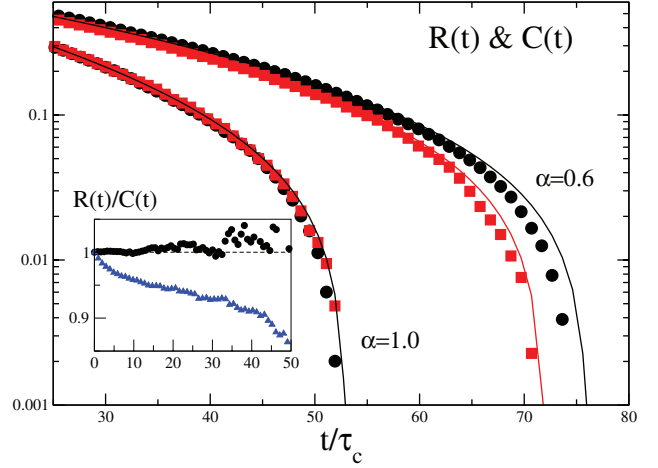

Fig. 2: (Color online) Correlation function $C(t)$ (black circles) and response function $R(t)$ (red squares) for $\alpha = 1$ and $\alpha = 0.6$, at $\phi = 0.33$. Continuous lines show the best-fits curves obtained with eqs. (9). Inset: the ratio $R(t)/C(t)$ is reported in the same cases.

An important independent assessment of the effectiveness of model 4 comes from the study of the fluctuating entropy production [13] which quantifies the deviation from detailed balance in a trajectory. Given the trajectory in the time interval $[0, t]$, $\{V(s)\}_0^t$, and its time-reversed $\{\mathcal{I}V(s)\}_0^t \equiv \{-V(t-s)\}_0^t$, in ref. [30] it has been shown that the entropy production for the model (4) takes the form

$$\Sigma_t = \log \frac{P(\{V(s)\}_0^t)}{P(\{\mathcal{I}V(s)\}_0^t)} \approx \gamma_0 \left(\frac{1}{T_0} - \frac{1}{T_1} \right) \int_0^t \mathrm{d}s \, V(s)U(s). \tag{13}$$

Boundary terms —in the stationary state— are subleading for large t and have been neglected. This functional vanishes exactly in the elastic case, $\alpha = 1$, where equipartition holds, $T_1 = T_0$, and is zero on average in the dilute limit, where $\langle VU \rangle = 0$. Formula (13) reveals that the leading source of entropy production is the energy transferred by the "force" $\gamma_0 U$ on the tracer, weighed by the difference between the inverse temperatures of the two "thermostats". Following the procedure described above, in the case $\phi = 0.33$ and $\alpha = 0.6$, we estimate for the correlation length $l^* \sim 9r \sim 6l_0$. Then, measuring the entropy production of eq. (13) (by replacing $U(t)$ with U_{l^*}) along many trajectories of length t, we can compute the probability $P(\Sigma_t = x)$ and compare it to $P(\Sigma_t = -x)$, in order to verify the fluctuation relation

$$\log \frac{P(\Sigma_t = x)}{P(\Sigma_t = -x)} = x. \tag{14}$$

In fig. 3 we report our numerical results. The main frame confirms that at large times the fluctuation relation (14) is well verified within the statistical errors. The inset shows the collapse of $\log P(\Sigma_t)/t$ onto the large deviation rate function for large times. Notice also that formula (13) does not contain further parameters but the ones already
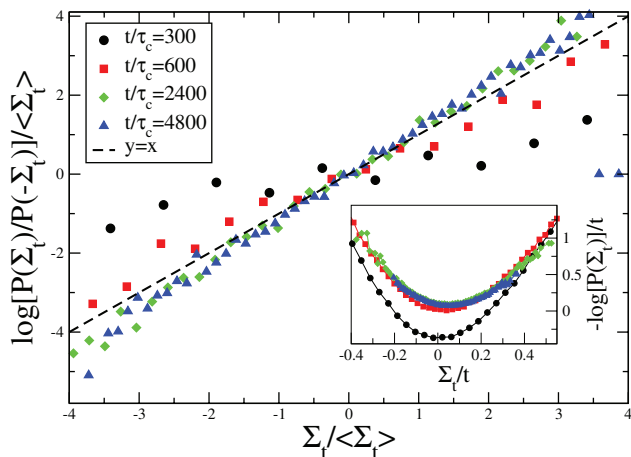
Fig. 3: (Color online) Check of the fluctuation relation (14) in the system with $\alpha = 0.6$ and $\phi = 0.33$. Inset: collapse of the rescaled probability distributions of Σ_t at large times onto the large deviation function.

determined by correlation and response measure, *i.e.* the slope of the graph is not adjusted by further fits. Indeed a wrong evaluation of the weighing factor $(1/T_0 - 1/T_1) \approx (1/T_{tr} - 1/T_b)$ or of the "energy injection rate" $\gamma_0 U(t) V(t)$ in eq. (13) could produce a completely different slope in fig. 3.

In conclusion, we designed a first granular dynamical theory describing non-equilibrium correlators and responses for a massive tracer. The value of this proposal is to offer a significant insight into the mechanisms of re-collision and dynamical memory and their unexplored relation with the breakdown of equilibrium properties. It is remarkable that velocity correlations $\langle V(t)U(t') \rangle$ between the intruder and the surrounding velocity field are responsible for both the violations of the EFDR and the appearance of a non-zero entropy production, provided that the two fields are *at different temperatures*. Small non-Gaussian corrections [23], always present in granular fluids, are neglected here in favor of the largest contribution given by memory terms to violations of EFDR and entropy production. For some of the parameters in the theory ($\gamma_0 \sim \Gamma_E$, $T_0 \sim T_{tr}$ and $T_1 \sim T_b$) we have reasonable predictions, while τ_1 and γ_1, related to the coupling between U and V, deserve further investigations. Close analytical predictions of all the parameters could be obtained through a full kinetic theory (beyond Enskog), also to deduce eventual extensions to the case $M \sim m$, larger densities, and hard spheres.

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