Intermediate regimes in granular Brownian motion: Superdiffusion and subdiffusion

Anna Bodrova,1 Awadhesh Kumar Dubey,2 Sanjay Puri,2 and Nikolai Brilliantov3

1Faculty of Physics, M.V.Lomonosov Moscow State University, Moscow, 119991, Russia
2School of Physical Sciences, Jawaharlal Nehru University, New Delhi, 110067, India.
3Department of Mathematics, University of Leicester, Leicester LE1 7RH, United Kingdom

(Dated: May 2, 2014)

Brownian motion in a granular gas in a homogeneous cooling state is studied theoretically and by means of molecular dynamics. We use the simplest first-principle model for the impact-velocity dependent restitution coefficient, as it follows for the model of viscoelastic spheres. We reveal that for a wide range of initial conditions the ratio of granular temperatures of Brownian and bath particles demonstrates complicated non-monotonous behavior, which results in transition between different regimes of Brownian dynamics: It starts from the ballistic motion, switches later to superballistic one and turns at still later times into subdiffusion; eventually normal diffusion is achieved. Our theory agrees very well with the MD results, although extreme computational costs prevented to detect the final diffusion regime. Qualitatively, the reported intermediate diffusion regimes are generic for granular gases with any realistic dependence of the restitution coefficient on the impact velocity.

Introduction. Brownian motion is a fundamental process in nature which takes place on very different time and length scales [1]. In its classical formulation it implies a random motion of a big (Brownian) particle driven by thermal motion of much smaller bath particles. The mean-square displacement of Brownian particles grows with time as \( \langle R^2 \rangle \sim t^3 \), with \( \beta = 1 \) for normal diffusion. Anomalous diffusion with \( \beta > 1 \) (superdiffusion) and \( \beta < 1 \) (subdiffusion) has been found in a variety of systems [2], ranging from supercooled and glass forming liquids, e.g. [3], to surfaces [4], biological [5] and granular [6] systems; usually a few diffusion regimes with a crossover between them are observed. Mechanisms of anomalous diffusion are seemingly very different – macro-molecular crowding in solutions and cells [3], caging in glassy liquids [5], trapping in a random potential [4], etc. Nevertheless, one can possibly infer an important common feature – anomalous diffusion always arises due to interplay of different basic dynamics in a system: For instance, a (fast) hopping dynamics in glass forming liquids interferes with a (slow) dynamics of structural rearrangements, resulting in subdiffusion. Moreover, anomalous diffusion in most of the systems manifests as an intermediate regime, which transforms asymptotically into normal diffusion, e.g. [3,4].

In the present Letter we report intermediate anomalous diffusion in granular systems, where particles interact with dissipative forces. Although the microscopic mechanism of this phenomenon is novel and related to inelastic collisions, it again results, as it is shown below, from the interplay of different dynamics represented by evolution of two granular temperatures.

Granular Brownian motion demonstrates a large variety of new surprising phenomena, among which is the violation of energy equipartition in a mixture of massive and light particles [7,8]: Both granular tempera-

\[ \varphi = \frac{\langle B \rangle}{\langle m_B \rangle} \]

tures of Brownian (\( T_B \)) and bath (\( T \)) particles decrease in a force-free granular gas, while their ratio rapidly relaxes to a steady-state value \( \langle B/T \rangle_{s.s.} > 1 \). Moreover, \( \langle T_B/T \rangle_{s.s.} \) increases with increasing inelasticity and the mass ratio \( m_B/m \) of Brownian (\( m_B \)) and bath (\( m \)) particles [7,8]. It has been also shown that Brownian motion in these systems may change qualitatively – from diffusive to a ballistic one, depending on the steady state value of \( \varphi = \langle B \rangle/m_B/\langle m_B \rangle \) [10]. Surprisingly, one can treat this phenomena as a "phase transition" with the "order parameter" \( \varphi \) [10], which however is not observed as a real process, since the steady state \( \varphi \) is constant.

To date, the most theoretical studies of Brownian motion are done with the assumption of a constant restitution coefficient \( \varepsilon = gT/g \), where \( g \) and \( gT \) are, respectively, the normal components of the relative inter-particle velocities before and after a collision. The simplifying hypothesis of constant \( \varepsilon \), however, contradicts experimental results, which indicate that \( \varepsilon \) does depend on \( g \) and that \( \varepsilon(g) \) is a decreasing function of the impact velocity [11]; it is not also consistent with a rigorous theoretical analysis [7]. The impact-velocity dependence of the restitution coefficient may, however, crucially change the behavior of a granular system, e.g. [12]. In particular, it has been shown that in a gas of viscoelastic particles, where the impact-velocity dependence of \( \varepsilon \) stems from the simplest first-principle model [13], clusters and vortices manifest as transient phenomena [14], in a sharp contrast with the case of a constant \( \varepsilon \) [13]. Furthermore, in a mixture of massive and light particles, the temperature ratio \( T_B/T \), as well as \( \varphi \), evolve with time in a HCS [12], again in contrast with the case of \( \varepsilon = \text{const} \). Therefore, one can expect that in a granular gas of viscoelastic particles, where \( \varphi \) varies with time, the "phase transition" between different regimes of Brownian motion may occur during the gas evolution as a real physical process.
We analyze granular Brownian motion in a gas of viscoelastic particles in a homogeneous cooling state (HCS) — the basic state of a force free granular gas by means of molecular dynamics (MD) and theoretically. We have revealed a sequence of new diffusive regimes for a Brownian particle — superballistic motion (superdiffusion), which follows after ballistic one and transforms then to subdiffusion. Eventually, normal diffusion is observed.

**MD simulations.** We perform event-driven simulations \[17\] for a force-free system, using \(N = 64000\) bath particles of mass \(m\) and diameter \(\sigma\) and one Brownian particle of mass \(m_B \gg m\) and diameter \(\sigma_B\) (for simplicity we take \(\sigma_B = \sigma\)) in a cube of length \(L = 130\sigma\) with periodic boundary conditions. The number density of the bath particles was \(n = N\pi \sigma^3/(6L^3) \simeq 0.015\). The reported results correspond to averages over 20 or 50 independent runs, depending on the initial conditions.

In our simulations we use the restitution coefficient, as it follows for the model of viscoelastic particles \[13, 18\]:

\[
\varepsilon = 1 + \sum_{k=1}^{\infty} C_k \delta^{k/2} (2u)^{k/20} w^{k/10}.
\]

Here, \(C_k\)'s are numerical coefficients, which have been computed up to \(k = 20\) \[18\] and \(\delta\) quantifies the dissipative interactions. Eq. \((1)\) describes both types of collisions — between Brownian particle and a bath particle and between the bath particles themselves. In the former case \(w = |(\vec{v}_{Bi} \cdot \hat{c})|\), where \(\vec{v}_{Bi} = \vec{v}_B - \vec{v}_i\) being the relative velocity of Brownian \((\vec{v}_B)\) and \(i\)-th bath \((\vec{v}_i)\) particle, \(v_T = \sqrt{2T/m}\) is the thermal velocity of the bath particles and \(\hat{c}\) is the unit vector, joining particles' centers at the collision instant.

In the latter case \(w = |(\vec{v}_{ij} \cdot \hat{c})|\), where \(\vec{v}_{ij} = \vec{v}_i - \vec{v}_j\) being the relative velocity of the colliding \(i\)-th and \(j\)-th bath particles. For both types of collisions we use for simplicity the same \(\delta = 0.1\). The quantity \(u(t) = T(t)/T(0)\) is the dimensionless temperature of the bath at time \(t\), and granular temperatures are defined as usual, \(3T/2 = (m\vec{v}^2/2)\) and \(3T_B/2 = \langle m_B \vec{v}_B^2/2\rangle\). The results of the MD simulation are presented in Fig. \[1\].

**Theory.** Since concentration of Brownian particles is much smaller than that of the bath particles, we assume, that Brownian particles do not affect evolution of the bath \[2\]. In a force-free gas \(T\) gradually decreases, due to dissipative collisions, with the cooling coefficient \(\xi(t) = -(dT/dt)/T(t)\). As temperature \(T\) tends to zero, \(\varepsilon\) tends to unity, and all collisions tend to be elastic \[\Xi\].

Evolution of Brownian particles may be described, using the Boltzmann equation, \(f_B(\vec{v}_B, t) = I(f_B, f)\) for the velocity distribution function, \(f_B(\vec{v}_B, t)\), e.g. \[\Xi\]; the collision integral \(I(f_B, f)\) accounts for the alteration of \(f_B(\vec{v}_B, t)\) in pair-wise collisions with the collision rule,

\[
\vec{v}_B' = \vec{v}_B - (1 + \varepsilon) \mu (\vec{v}_B \cdot \hat{c}) \hat{c}.
\]

Here \(\vec{v}_B'\) is the velocity of a Brownian particle after a collision and \(\mu \equiv m_B/m\). Solving the Boltzmann equation with the standard technique, e.g. \[2\], one obtains the distribution function \(f_B\). Brownian dynamics may be described with very compact notations if the pseudo-Liouville operator \(\hat{L}\) is used \[\Xi\]:

\[
\dot{\vec{v}}_B = \hat{L} \vec{v}_B = \hat{L}_0 \vec{v}_B + \sum_i \hat{T}_{Bi} \vec{v}_B,
\]

where \(\hat{L}_0 = \vec{v}_B \cdot \nabla \vec{r}_B\) describes the free streaming and \(\hat{T}_{Bi} = \sigma_B^2 \int d\Theta (-(\vec{v}_B \cdot \hat{c}))[\vec{v}_B \cdot \hat{c}] \delta (\vec{r}_B - \vec{e}_0) (\vec{b}_B_i - 1)\) the binary collisions of particles. The radius-vector \(\vec{r}_B\) in the above equation joins centers of Brownian and bath particle, \(\sigma_0 = (\sigma_B + \sigma)/2\) and the operator \(\hat{b}_{Bi}\) acts on the velocities, as \(\vec{b}_B, \vec{v}_B = \vec{v}_B'\) with the collision rule \[\Xi\].

Taking into account, that \(\hat{R}_B(t) = \int_0^t \vec{v}_B(t_1) dt_1\), the mean-square displacement of Brownian particles reads in terms of the velocity autocorrelation function:

\[
\langle R^2_B(t) \rangle = \int_0^t dt_1 \int_0^t dt_2 \langle \vec{v}_B(t_1) \vec{v}_B(t_2) \rangle.
\]

We introduce the reduced time \(\tau_B\), defined as

\[
d\tau_B = dt \sqrt{T_B(t)/T_B(0)/\tau_c(0)},
\]

where \(\tau_c^{-1}(t) = 4\sqrt{\pi\sigma^2 g_2(\sigma) m \sqrt{T(t)/m}}\) is the mean collision time of bath particles, with \(g_2(\sigma)\) being the contact value of the pair correlation function \[\Xi\]. In this time scale one deals with the reduced velocities, \(\vec{v}_B = \vec{v}_B/\sqrt{2T/m}\), and granular temperature of Brownian particles remains constant. Hence the decay of the (reduced) velocity correlation function occurs similarly, as in equilibrium gases: A particle loses memory of its initial velocity in random collisions; in dilute gases the velocity correlation function is exponential \[19\], therefore we approximate

\[
\langle \vec{c}_B(\tau_B(t_1)) \vec{c}_B(\tau_B(t_2)) \rangle = \langle \vec{c}_B^2 \rangle \exp \left( -\frac{\tau_B(t_2) - \tau_B(t_1)}{\tau_c(1)} \right),
\]

where \(\langle \vec{c}_B^2 \rangle = 3/2\) and \(\tau_c(1)\) is the (reduced) velocity relaxation time. Exploiting the above form of the correlation function, we obtain the mean-square displacement:

\[
\langle R^2_B(t) \rangle = 6 \int_0^t dt_1 D_B(t_1) \left[1 - \exp \left( -\frac{\tau_B(t) - \tau_B(t_1)}{\tau_c(1)} \right) \right].
\]

Here \(D_B = T_B(t)/\tau_c(0)/m\) is the time-dependent diffusion coefficient of a Brownian particle, expressed in terms of \(\tau_c(t) = \tau_c(0)/\sqrt{T_B(t)/T_B(0)}\) — the velocity relaxation time in laboratory time units. The reduced relaxation time \(\tau_c(1)/t\) in the exponential function in Eq. \[\Xi\] may be easily found from its time derivative at \(t_1 \to t_2\), yielding \[\Xi, 20\]

\[
\tau_c^{-1} = -(N - 1) \frac{\langle \vec{v}_{T B} \vec{v}_{T B} \rangle}{\langle \vec{v}_B^2 \rangle} - \frac{1}{2} \xi_B,
\]

with the cooling coefficient of Brownian particles

\[
\xi_B = -\frac{1}{T_B} \frac{dT_B}{dt} = -(N - 1) \frac{\langle \vec{T}_{B B} \vec{v}_B^2 \rangle}{\langle \vec{v}_B^2 \rangle}.
\]
Performing the averaging in Eqs. (5) and (6), we ignore, for simplicity, deviations of the velocity distribution function from the Maxwellian [12]; it may be however shown that their impact on the calculated quantities is negligible. Hence, we obtain the velocity relaxation time

$$\frac{\tau_{vB}^{-1}(t)}{\tau_{vB}^{-1}(0)} = \sqrt{\frac{8u}{9}} \left( 1 + \frac{1}{2} \sum_{i=2}^{\infty} A_i B_i \right)$$

and the cooling coefficient of Brownian particles:

$$\xi_B(t) = 2\tau_{vB}^{-1}(0) \sqrt{\frac{8u}{9}} \left( 1 + \frac{1}{2} \sum_{i=2}^{\infty} A_i B_i \right),$$

where $A_i = 4C_i + \sum_{j+k=i} C_j C_k$ are pure numbers and

$$B_i(t) = \delta^{i/2} \langle u(t) \rangle^{i/20} \left( \frac{20i + i^2}{800} \right) \Gamma \left( \frac{i}{20} \right)$$

with $\Gamma(x)$ being the Gamma-function.

The temperature of the Brownian particles $T_B$ can be found from the equation $dT_B(t)/dt = -\xi_B(t)T_B(t)$, while the temperature $T(t)$ and the mean-squared displacement $\langle R^2(t) \rangle$ of the bath particles – from the above results using the substitute $m_B \to m$ and $\sigma_B \to \sigma$.

**Results and discussion.** The theoretical predictions are compared with the simulation data in Fig. 1. First we consider the case of the initial energy equipartition, $T_B(0) = T(0) = 400/3$. The time dependence of the ratio $T_B/T$ demonstrates here a complicated non-monotonous behavior: It initially increases, reaches a maximum, and then decreases, tending eventually to unity, Fig. 1(b). This is in a sharp contrast with the case of a constant $\varepsilon$, where $T_B/T$ rapidly relaxes to a steady-state value. One can explain this effect as follows. From the collision rules [see, e.g., Eq. (2)], the ratio of energy losses of Brownian ($\Delta E_B$) and bath ($\Delta E$) particles at a collision, scales as $\Delta E_B/\Delta E \sim v_B/v_i \sim \sqrt{(T_B m_B)/(T m_B)}$. Initially $\Delta E_B \ll \Delta E$, since $T_B = T$ and $m_B \gg m$, therefore Brownian particles cool down significantly slower than the bath particles and the temperature ratio $T_B/T$ increases. One can call this effect as "retarded cooling" , which may be also understood as a tendency of a system to reach the ratio of $T_B/T > 1$, corresponding to the steady-state value $(T_B/T)_{ss} > 1$ of a gas with the respective constant $\varepsilon$. The larger the mass ratio the more pronounced the effect (Fig. 1(b). In the course of time the temperature ratio $T_B/T$ gets so large that $\Delta E_B$ and $\Delta E$ become comparable; the granular temperatures start then to equilibrate. At this stage $T_B/T$ decreases – Brownian particles cool faster than the bath particles and one can call this regime as "accelerated cooling". Again, this effect may be understood, noticing that the ratio of $(T_B/T)_{ss} > 1$ is smaller for larger $\varepsilon$ and that the effective restitution coefficient grows as the gas cools down, see Eq. (11). For very large times $\varepsilon$ tends to unity, the system becomes elastic and the energy equipartition is achieved. This complicated temperature dependence results in transition between different diffusion regimes, which may be most clearly seen using the re-scaled time $\tau_u$, defined as $d\tau_u = dt \sqrt{u(t)}/\tau_i(0)$. In this time scale the average velocity of the bath particles is constant and they move, as in an equilibrium molecular gas, that is, ballistically, $\langle R^2 \rangle \sim \tau_u^2$; for $\tau_u \sim 1$ and diffusively, $\langle R^2 \rangle \sim \tau_u^2$ with $\beta < 1$; 4 - normal diffusion $\langle R^2 \rangle \sim \tau_u$.

FIG. 1: (a) Time dependence of the temperature ratio of Brownian ($T_B$) and bath ($T$) particles. (b) The mean-square displacement of Brownian particles $\langle R^2_B \rangle$ as the function of the re-scaled time $\tau_u$ and the laboratory time $t$ (inset). Lines – theory; symbols – MD data. Numbers indicate different regimes: 1 - ballistic motion, $\langle R^2_B \rangle \sim \tau_u^2$; 2 - superballistic regime (superdiffusion), $\langle R^2_B \rangle \sim \tau_u^2$ with $\beta > 1$; 3 - subdiffusion, $\langle R^2_B \rangle \sim \tau_u^2$ with $\beta < 1$; 4 - normal diffusion $\langle R^2_B \rangle \sim \tau_u$. Brownian motion demonstrates, however, four different regimes – ballistic (regime 1 in Fig. 1(b)), superballistic motion or superdiffusion (regime 2), subdiffusion (regime 3) and normal diffusion (regime 4). Physically, the superballistic motion, which starts after a short ballistic regime corresponds to the "retarded cooling", while the subsequent subdiffusion – to the "accelerated cooling". In the former case Brownian particles get hotter and hotter with respect to the surrounding gas and $\langle R^2_B \rangle$ grows with time faster than for normal diffusion; in the latter case, Brownian particles cool more rapidly than the gas, so that $\langle R^2_B \rangle \sim \tau_u^2$. The larger the mass ratio the more pronounced the effect (Fig. 1(b). In the course of time the temperature ratio $T_B/T$ gets so large that $\Delta E_B$ and $\Delta E$ become comparable; the granular temperatures start then to equilibrate. At this stage $T_B/T$ decreases – Brownian particles cool faster than the bath particles and one can call this regime as "accelerated cooling". Again, this effect may be understood, noticing that the ratio of $(T_B/T)_{ss} > 1$ is smaller for larger $\varepsilon$ and that the effective restitution coefficient grows as the gas cools down, see Eq. (11). For very large times $\varepsilon$ tends to unity, the system becomes elastic and the energy equipartition is achieved. This complicated temperature dependence results in transition between different diffusion regimes, which may be most clearly seen using the re-scaled time $\tau_u$, defined as $d\tau_u = dt \sqrt{u(t)}/\tau_i(0)$. In this time scale the average velocity of the bath particles is constant and they move, as in an equilibrium molecular gas, that is, ballistically, $\langle R^2 \rangle \sim \tau_u^2$; for $\tau_u \sim 1$ and diffusively, $\langle R^2 \rangle \sim \tau_u^2$ with $\beta < 1$; 4 - normal diffusion $\langle R^2 \rangle \sim \tau_u$. Brownian motion demonstrates, however, four different regimes – ballistic (regime 1 in Fig. 1(b)), superballistic motion or superdiffusion (regime 2), subdiffusion (regime 3) and normal diffusion (regime 4). Physically, the superballistic motion, which starts after a short ballistic regime corresponds to the "retarded cooling", while the subsequent subdiffusion – to the "accelerated cooling". In the former case Brownian particles get hotter and hotter with respect to the surrounding gas and $\langle R^2_B \rangle$ grows with time faster than for normal diffusion; in the latter case, Brownian particles cool more rapidly than the gas, so that $\langle R^2_B \rangle \sim \tau_u^2$.
grows with time slower. The duration of the subdiffusion regime increases with the mass ratio $m_B/m$ and may be very long. Asymptotically, at $\tau_B \to \infty$, the system returns to the equipartition, demonstrating the normal diffusion, $\langle R_B^2 \rangle \sim \tau_B$.

The respective transition between diffusion regimes is also seen for the laboratory time $t$ (Fig. 1A, inset), where the "normal diffusion" is described by $\langle R_B^2 \rangle \sim t^{1/6}$, as it follows for the gas of viscoelastic particles [7, 20]. If we consider the time scale $\tau_B$, where $T_B$ keeps constant, the superballistic motion does not appear; all three other diffusion regimes, nevertheless, present.

Since superballistic motion is caused by the "retarded cooling", it is not observed if $T_B/T$ does not increase. This happens for initial conditions, when the ratio $T_B/T$ is already large, or more precisely, when $T_B/T$ exceeds some threshold, estimated as the ratio $(T_B/T)_{\text{n.s.}}$, for a gas with a constant $\varepsilon$, equal to the effective restitution coefficient $\varepsilon_{\text{eff}}$ for a gas of viscoelastic particles at $t = 0$. In our case Eq. (1) with $u(0) = 1$ and $\langle w \rangle = 4/\sqrt{2\pi}$ yields $\varepsilon_{\text{eff}}(0) = 0.876$ and $T_B(T)/T(0) = (T_B/T)_{\text{n.s.}} = 213.5$. Hence, this initial temperature ratio delimits two incipient diffusion regimes: For $T_B(0)/T(0) < 213.5$ the ballistic and then superballistic motion takes place, while otherwise only ballistic regime is observed, Fig. 1b. Interestingly, the ballistic motion in the latter case lasts for significantly longer time, than for normal diffusion: this follows from the fact that trajectories of heavy Brownian particles are almost not affected by much colder bath of light particles. The subsequent subdiffusion and normal diffusion persist independently on initial conditions, see Fig. 1b. This is the universal feature of Brownian motion in a gas of viscoelastic particles. As it follows from Fig. 1a a very good agreement between our theory and simulation data is observed for evolution of temperature and mean-square displacement for intermediate ballistic, superballistic and the onset of subdiffusive regimes; due to extreme computational costs we fail to detect numerically the final regime of normal diffusion.

Conclusion. We have studied Brownian motion in a force-free granular gas, composed of particles interacting with an impact-velocity dependent restitution coefficient $\varepsilon(g)$, as it follows from the model of viscoelastic spheres. We have revealed that for a wide range of initial conditions the ratio of granular temperatures of massive and light particles $T_B/T$ demonstrates complicated non-monotonous dependence on time as the system evolves in the homogeneous cooling state. Somewhat similar to other systems with anomalous diffusion, this interplay of different dynamics of Brownian and bath particles, quantified by their temperatures, gives rise to a sequence of intermediate diffusive regimes in granular Brownian motion: At early times the ballistic motion is observed, which alters then to superballistic one (for non-monotonous evolution of $T_B/T$). At still later times the opposite regime of subdiffusion – motion, slower than normal diffusion, always takes place; it lasts for a relatively long time and tends asymptotically to normal diffusion. Qualitatively, these features of granular Brownian motion are not specific to a particular model of $\varepsilon(g)$ but generic for granular systems with any realistic restitution coefficient that increases with decreasing impact velocity.

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