Mpemba effect in molecular gases under nonlinear drag

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We look into the Mpemba effect—the initially hotter sample cools sooner—in a molecular gas with nonlinear viscous drag. Specifically, the gas particles interact among them via elastic collisions and also with a background fluid at equilibrium. Thus, within the framework of kinetic theory, our gas is described by an Enskog–Fokker–Planck equation. The analysis is carried out in the first Sonine approximation, in which the evolution of the temperature is coupled to that of the excess kurtosis. This coupling leads to the emergence of the Mpemba effect, which is observed in an early stage of the relaxation and when the initial temperatures of the two samples are close enough. This allows for the development of a simple theory, linearizing the temperature evolution around a reference temperature—namely the initial temperature closer to the asymptotic equilibrium value. The linear theory provides a semiquantitative description of the effect, including expressions for the crossover time and the maximum temperature difference. We also discuss the limitations of our linearized theory.

I. INTRODUCTION

One of the signatures of nonequilibrium systems is the presence of memory effects. A system displays memory when its time evolution from a given initial state is not uniquely determined by the initial values of its macroscopic—or hydrodynamic variables. In other words, the system evolution depends on how it has been previously being aged: memory effects are thus intimately related to aging, which has been typically associated with glassy behavior. Notwithstanding this, in addition to being investigated in models for glasses, it has been found in many different physical systems: granular fluids, dense granular matter, ferroelectrics, disordered mechanical systems, and frictional interfaces, to name just a few.

The Mpemba effect is a counterintuitive memory phenomenon: given two samples of fluid, the one that is initially hotter may cool more rapidly. Therefore, the curves describing the time evolution of the temperature for the two samples cross each other at a certain time $t_c$, and the curve for the initially hotter sample stays below the other one for longer times, $t > t_c$. It is important to characterize the range of values of the relevant physical quantities that allow for the emergence of the Mpemba effect: in general, the difference of initial temperatures must be small enough.

Although first reported in the case of water, its existence for that liquid is still controversial. As a proof of concept, the feasibility of the Mpemba effect has recently been reported in granular gases. Therein, collisional inelasticity couples the evolution of the (granular) temperature to other quantities—such as the kurtosis or the rotational-to-translational energy ratio—monitoring the nonequilibrium nature of the velocity distribution function (VDF), even in homogeneous and isotropic states.

In this work, we show that the Mpemba effect is also present in homogeneous and isotropic states of molecular gases—i.e., with elastic collisions—driven by an external drag force with a velocity-dependent friction coefficient. The particles of our system are supposed to be hard spheres, for the sake of simplicity, surrounded by a background fluid in equilibrium. Gas particles collide among them, these collisions being modeled by a Boltzmann–Enskog collision term in the evolution equation for the VDF.

Gas particles also interact with the background fluid. This interaction translates into two forces: (i) a macroscopic, deterministic, nonlinear drag force, and (ii) a stochastic force. The intensity of the latter follows from the fluctuation-dissipation theorem, which ensures that the gas VDF tends to a Maxwellian with the temperature of the background fluid in the long-time limit. In the evolution equation, the interaction between the gas and the fluid is described by a Fokker–Planck term: Therefore, the VDF obeys an Enskog–Fokker–Planck equation with nonlinear drag.

The framework of our work is thus nonlinear Brownian motion, but the Brownian particles are no longer independent since they interact through instantaneous hard collisions. If the particles of the background fluid, with mass $m_{bf}$, are much lighter than the Brownian particles, with mass $m$, the drag force is usually assumed to be linear in the velocity of the particles, $F = -m\zeta v$. In fact, this is the leading behavior found when an expansion in powers of $m_{bf}/m$ is performed, which leads to linear Brownian motion. Nevertheless, the drag force becomes nonlinear when higher order terms in the expansion are brought to bear. Specifically, the drag force can be written as $F = -m\zeta (v)v$ and there appears a velocity-
dependent drag coefficient $\zeta(v)$, with $\zeta(v = 0) = \zeta_0$. Therein, the first correction in $\zeta_0$ introduces a quadratic dependence on $v$. In some situations, nonlinearities in the drag coefficient have quite strong physical implications.

The main goal of this paper is to study the Mpemba effect in the kinetic theory framework we have just described, i.e., the Enskog–Fokker–Planck equation with nonlinear drag. To meet this end, we work in the first Sonine approximation, in which the time evolution of the temperature is coupled to that of the excess kurtosis. This coupling, which is absent in the case $\zeta(v) = \zeta_0$, is responsible for the emergence of the Mpemba effect. The value of the excess kurtosis is assumed to be small in the Sonine approximation, which entails that the initial temperatures of the samples must be close to each other and the Mpemba crossover takes place in the early stage of the relaxation. This allows us to linearize the problem and derive analytical expressions for the relevant physical quantities that characterize the Mpemba effect, like the crossing time in the temperature evolution, the maximum value of the initial temperature difference, and the magnitude of the effect.

The plan of the paper is as follows. We put forward the model and the kinetic description in Sec. III where the equations for the velocity moments are also derived. Section III is devoted to the Sonine approximation: The infinite hierarchy for the velocity moments is closed by expanding the VDF in Laguerre polynomials, retaining only the first nontrivial cumulant, namely the excess kurtosis. The Mpemba effect is analyzed in Sec. IV, where we develop a linearized model, investigate the crossover time, construct the phase diagram in the space of parameters, quantify the magnitude of the effect, and finally study the accuracy of the linearized theory. Finally, Sec. VI presents the main conclusions of our work.

II. ENSKOG–FOKKER–PLANCK EQUATION. MOMENT EQUATIONS

Let us consider a $d$-dimensional system of elastic hard spheres of mass $m$ and diameter $\sigma$ in a uniform and isotropic fluidized state. The spheres are assumed to be suspended in a background fluid in equilibrium, so that they are subject to a nonlinear drag force $F = -m\zeta(v)v$ plus a white-noise stochastic force with a nonlinear variance $m^2\xi^2(v)$. Under those conditions, the corresponding Enskog–Fokker–Planck equation reads

$$\partial_t f(v) - \frac{\partial}{\partial v} \left[ \zeta(v)v + \frac{\xi^2(v)}{2} \frac{\partial}{\partial v} \right] f(v) = J[v|f, f].$$

Here, $f(v)$ is the one-body VDF and

$$J[v_1|f, f] = \sigma^{d-1} g(\sigma^+) \int dv_2 \int d\sigma \Theta(v_{12} \cdot \sigma) v_{12} \cdot \sigma \times [f(v'_1) f(v'_2) - f(v_1) f(v_2)].$$

is the Boltzmann–Enskog collision operator, where $g(\sigma^+)$ is the contact value of the pair correlation function, $\Theta(\cdot)$ is the Heaviside step function, $v_{12} = v_1 - v_2$ is the relative velocity, and the primes denote postcollisional velocities.

The velocity-dependent coefficients $\zeta(v)$ and $\xi(v)$ are related by the condition that Eq. (2.1) admits as a stationary solution the equilibrium VDF

$$f_s(v) = n \left( \frac{m}{2\pi k_B T_s} \right)^{d/2} e^{-mv^2/2k_BT_s},$$

where $k_B$ is the Boltzmann constant and $T_s$ is the equilibrium temperature of the background fluid, which acts as a thermostat. This yields the fluctuation-dissipation relation

$$\xi^2(v) = \frac{2k_BT_s}{m} \zeta(v).$$

Equation (2.1) can then be rewritten as

$$\partial_t f(v) - \frac{\partial}{\partial v} \cdot \zeta(v)(v + \frac{k_BT_s}{m} \frac{\partial}{\partial v}) f(v) = J[v|f, f],$$

which describes the Brownian motion of an ensemble of particles of mass $m$ moving in the background fluid. These Brownian particles are not independent, their interaction being incorporated through the collision term. In this work, as the simplest nonlinear model, we consider the quadratic dependence of the drag coefficient on the velocity derived in Refs. 31–33. Thus, we restrict ourselves to

$$\zeta(v) = \zeta_0 \left( 1 + \gamma \frac{mv^2}{k_BT_s} \right),$$

where $\gamma > 0$ is a dimensionless parameter measuring the degree of nonlinearity of the drag force. When both Brownian particles and background fluid particles are three-dimensional hard spheres, it has been shown that $\gamma = m_{sd}/10m$. See also Appendix A.

By taking velocity moments in Eq. (2.9), the evolution equation for the temperature

$$T = \frac{m}{k_BT} \langle v^2 \rangle = \frac{m}{k_BT} \int dv v^2 f(v),$$

where $n = \int dv f(v)$ is the number density, is obtained as

$$\frac{\dot{T}}{T} = 2(T_s - T) \left[ 1 + \gamma (d + 2) \frac{T}{T_s} \right] - 2\gamma (d + 2) \frac{T^2}{T_s} a_2,$$

in which we have introduced the excess kurtosis

$$a_2 = \frac{d}{d + 2} \left( \langle v^d \rangle \right)^2 - 1.$$

In the particular case of a linear drag, $\gamma = 0$, the solution to Eq. (2.9) is simply $T(t) = T_0 + (T(0) - T_s) e^{-2\gamma t}$. 

However, in the case of nonlinear drag, $\gamma > 0$, the evolution of the temperature is coupled to that of the excess kurtosis. Imagine that $T(0) > T_s$; the larger the value of $a_2(0)$, the larger the initial cooling rate is and the sooner the temperature is expected to reach the thermostat value $T_s$. This property can give rise to a Mpemba phenomenon, as reported in the case of granular fluids. Similarly, the inverse Mpemba effect, in which the cooler phenomenon, as reported in the case of granular fluids, may also be expected for $T(0) < T_s$.

Since the evolution equation (2.8) involves the excess kurtosis $a_2(t)$, we need to consider its evolution equation. This in turn involves sixth-degree moments, and so on, giving rise to an infinite hierarchy of moment equations. To derive this hierarchy, let us introduce the dimensionless VDF $\phi(c)$ as

$$ f(v) = \frac{n}{v_T^3(t)}\phi(c), \quad c = \frac{v}{v_T(t)}, \quad (2.10) $$

where $v_T(t) \equiv \sqrt{2k_BT(t)/m}$ is the thermal velocity. Then, the kinetic equation (2.8) becomes

$$ \partial_t \phi(c) - \frac{\partial}{\partial c} \left[ \frac{T}{2T} c + \zeta_0 \left( 1 + \frac{2T}{T_s} c^2 \right) \right] \phi(c) = \nu_s \sqrt{\frac{T}{T_s}} I[c|\phi, \phi], \quad (2.11) $$

in which we have defined $\nu_s \equiv g(\sigma^+)n\sigma d^{-1} \sqrt{2k_BT_s/m}$, which is basically the collision frequency at the steady state, and the dimensionless collision operator

$$ I[c_1|\phi, \phi] = \int d\sigma^2 \int d\sigma \Theta(c_{12} \cdot \hat{c}) c_{12} \cdot \hat{c} \left[ \phi(c_1')\phi(c_2') - \phi(c_1)\phi(c_2) \right]. \quad (2.12) $$

Also, we have employed the property

$$ \frac{v_T^n}{n} \partial_t f(v) = \partial_t \phi(c) - \frac{T}{2T} \frac{\partial}{\partial c} \left[ \phi(c) \right]. \quad (2.13) $$

Multiplying both sides of Eq. (2.11) by $c^\ell$, integrating over $c$, and making use of Eq. (2.8), we obtain the hierarchy of equations for the moments $M_\ell \equiv \langle c^\ell \rangle$,

$$ M_\ell = \ell \zeta_0 \left\{ \left[ \gamma(\ell - 2) + \gamma(d + 2) \frac{T}{T_s} (1 + a_2) - \frac{T}{T_s} \right] M_{\ell-2} - \gamma \frac{2T}{T_s} M_{\ell+2} + \frac{\ell + d - 2}{2} \frac{T}{T_s} M_{\ell-2} \right\} - \nu_s \sqrt{\frac{T}{T_s}} \mu_\ell, \quad (2.14) $$

Here, we have introduced the collisional moments $\mu_\ell$ as

$$ \mu_\ell = - \int dc c^\ell I[c|\phi, \phi]. \quad (2.15) $$

Since, by definition, $M_0 = 1$, $M_2 = d/2$, and $M_4 = d(d + 2)(1 + a_2)/4$, it is easy to check that, as it should be, $M_0 = \dot{M}_2 = 0$ (note that $\mu_0 = \mu_2 = 0$). Next, setting $\ell = 4$ in Eq. (2.14), we get

$$ \dot{a}_2 = 8 \zeta_0 \gamma \frac{4T}{T_s} \left\{ \frac{2T}{T_s} (1 + a_2) + (d + 2)(1 + a_2)^2 - (d + 4) \times (1 + 3a_2 - a_3) \right\} - \gamma \frac{4T}{T_s} a_2 - \frac{4\nu_s}{d(d + 2)} \sqrt{\frac{T}{T_s}} \mu_4, \quad (2.16) $$

where we have introduced the sixth-degree cumulant $a_3$ by $M_6 = d(d + 2)(d + 4)(1 + 3a_2 - a_3)/8$.

Some comments are in order. First, note that two time scales compete in Eqs. (2.14) and (2.16). The inverse of the drag coefficient for low velocities, $\zeta_0^{-1}$, dictates the time scale over which particles feel the action of the background fluid. Meanwhile, the characteristic time for particle-particle collisions is the inverse of the stationary collision frequency, $\nu_s^{-1}$. Second, Equations (2.8), (2.11), and (2.16) are formally exact within the Enskog–Fokker–Planck description, but they do not make a closed finite set. Not only does $M_\ell$ explicitly involve a higher-degree moment $M_{\ell+2}$, but also the collisional moment $\mu_\ell$ is a nonlinear functional of the full VDF $\phi(c)$. An approximate closure is needed to deal with a finite set of equations.

### III. SONINE APPROXIMATION

For isotropic states, the reduced VDF $\phi(c)$ can be expanded in a complete set of orthogonal polynomials as

$$ \phi(c) = e^{-c^2} \left[ 1 + \sum_{\ell=2}^{\infty} a_\ell L_\ell^{(\nu)}(c^2) \right], \quad (3.1) $$

where $L_\ell^{(\nu)}(x)$ are generalized Laguerre (or Sonine) polynomials. In the first Sonine approximation, all terms beyond $\ell = 2$ in Eq. (3.1) are dropped, i.e.,

$$ \phi(c) \approx e^{-c^2} \left[ 1 + a_2 \left\{ c^2 - \frac{d + 2}{2} c^2 + \frac{d(d + 2)}{8} \right\} \right]. \quad (3.2) $$

Inserting Eq. (3.2) into Eq. (2.16) with $\ell = 2$ and neglecting terms quadratic in $a_2$, one obtains $\Gamma(d/2)\mu_4 \approx \sqrt{2}(d - 1)\pi^{d/2} / 4 a_2^{d/2 - 1}$. Therefore, Eq. (2.16) becomes

$$ \dot{a}_2 = 8 \zeta_0 \gamma \left\{ \frac{4T}{T_s} - 8 \gamma + 4\gamma(d + 8) \frac{T}{T_s} \right\} + \tau_s^{-1} \left( \frac{8(d - 1)}{d(d + 2)} \sqrt{\frac{T}{T_s}} \right) a_2, \quad (3.3) $$

where we have introduced the mean free time at the steady state

$$ \tau_s = \sqrt{\Gamma(d/2)\pi^{1-d}/2 - 1}, \quad (3.4) $$
and, for consistency, the terms $a_2^2$ and $a_3$ have been neglected.

Equations 2.16 and 3.3 make a closed set to investigate the existence of the Mpemba effect. First, we define dimensionless temperature and time by

$$\theta = \frac{T}{T_s}, \quad t^* = \frac{t}{t_s}.$$  \hspace{1cm} (3.5)

The latter approximately measures the accumulated number of collisions per particle up to time $t$. With these variables, Eqs. 2.16 and 3.3 can be rewritten as

$$\dot{\theta} = 2(1 - \theta) [1 + \gamma/(d + 2)\theta] - 2\gamma(d + 2)\theta^2 a_2,$$  \hspace{1cm} (3.6a)

$$\frac{\partial}{\partial t} = 8\gamma(1 - \theta) - \left[\frac{4}{\theta} - 8\gamma + 4\gamma(d + 8)\theta\right] a_2,$$  \hspace{1cm} (3.6b)

where we have introduced a dimensionless low-velocity drag coefficient as

$$\zeta_0^* = \zeta_0 t_s.$$  \hspace{1cm} (3.7)

Now, the dot over $\theta$ and $a_2$ denotes a derivative with respect to $t^*$.

Equations 3.6 constitute our starting point for the analysis of the Mpemba effect, to be carried out in the next section. In the dimensionless variables we are using, there are only two relevant parameters: (i) $\gamma$, which measures the strength of the nonlinearity in the drag, and (ii) $\zeta_0^*$, which compares the characteristic times for collisions, $t_s$, and for the viscous drag, $\zeta_0 t_s$. Note that the regime $\zeta_0^* \ll 1$ ($\zeta_0^* \gg 1$) means that the viscous drag acts over a much longer (shorter) time scale than collisions do.

IV. MPEMBA EFFECT

A. Linearized model

Let us imagine two initial states $A$ and $B$ with $\{(\theta(0), a_2(0)) = \{\theta_A^0, a_{2A}^0\}$ and $\{\theta_B^0, a_{2B}^0\}$, respectively. The corresponding solutions to Eqs. 3.6 are denoted by $\{\theta_A(t^*), a_{2A}(t^*)\}$ and $\{\theta_B(t^*), a_{2B}(t^*)\}$. Without loss of generality, we assume that $\theta_A^0 > \theta_B^0$.

Below, we show that both the Mpemba effect and its inverse version are expected to emerge when the initially hotter sample has the larger value of the excess kurtosis. First, we analyze the case in which both initial temperatures are higher than the stationary one, $\theta_A^0 > \theta_B^0 > 1$, and the system cools down to reach the steady state. The Mpemba effect is present when $\theta_A(t^*)$ relaxes more rapidly than $\theta_B(t^*)$, which calls for the existence of a crossover time $t^*_c$ such that $\theta_A(t^*_c) = \theta_B(t^*_c)$. Since the cooling rate increases with $a_2$, the condition $a_{2A}^0 > a_{2B}^0$ seems to be necessary for the Mpemba effect to emerge.

Second, we look into the case in which both temperatures are lower than the stationary one, $1 > \theta_A^0 > \theta_B^0$, and the system heats up. The inverse Mpemba effect appears if $\theta_A(t^*)$ relaxes more slowly than $\theta_B(t^*)$, which again needs that $a_{2A}^0 > a_{2B}^0$.

In general, the nonlinear dependence on $\theta$ of the set of equations 3.6 impedes a fully analytical treatment. However, the excess kurtosis is supposed to be small in the Sonine approximation, which has allowed us to neglect nonlinear terms in $a_2$. Since $a_2$ is the quantity controlling the appearance of the Mpemba effect, its smallness implies that both initial temperatures, $\theta_A^0$ and $\theta_B^0$, cannot be very far from each other for the Mpemba effect to emerge. In addition, the crossing of the curves $\theta_A(t^*)$ and $\theta_B(t^*)$ must take place in the early stage of evolution.

Following the discussion above, we write $\theta(t^*) = \theta_r + \Psi(t^*)$, where $\theta_r \approx \theta_A^0 \approx \theta_B^0$ is a certain reference temperature, and linearize Eqs. 3.6 with respect to $\Psi(t^*)$ and $a_2(t^*)$. The detailed solution of this linearization procedure is carried out in Appendix B; here we present the results relevant for the analysis of the Mpemba effect. The time evolution is controlled by the matrix $\Lambda$ with elements

$$\Lambda_{11} = 2\zeta_0^*[1 + \gamma/(d + 2)(2\theta_r - 1)],$$  \hspace{1cm} (4.1a)

$$\Lambda_{12} = 2\zeta_0^*(d + 2)\theta_r^2,$$  \hspace{1cm} (4.1b)

$$\Lambda_{21} = 8\zeta_0^*\gamma,$$  \hspace{1cm} (4.1c)

$$\Lambda_{22} = \zeta_0^* \left[\frac{4}{\theta} - 8\gamma + 4\gamma(d + 8)\theta_r\right] + \frac{8(d - 1)}{d(d + 2)} \sqrt{\theta_r},$$  \hspace{1cm} (4.1d)

and eigenvalues

$$\lambda_{\pm} = \frac{\Lambda_{11} + \Lambda_{22} \pm \delta_\lambda}{2}, \quad \delta_\lambda \equiv \sqrt{(\Lambda_{11} - \Lambda_{22})^2 + 4\Lambda_{12}\Lambda_{21}}.$$  \hspace{1cm} (4.2)

Let us consider the differences $\Delta \theta(t^*) \equiv \theta_A(t^*) - \theta_B(t^*)$ and $\Delta a_2(t^*) \equiv a_{2A}(t^*) - a_{2B}(t^*)$ between the time evolutions corresponding to the two different initial states $A$ and $B$. Within the linearized theory, these differences are given by (see Appendix B)

$$\Delta \theta(t^*) = \frac{\lambda - \Lambda_{11}}{\delta_\lambda} \Delta \theta_0 - \frac{\Lambda_{12}}{\delta_\lambda} \Delta a_2^0 e^{-\lambda_r t^*},$$

$$- \frac{\lambda - \Lambda_{11}}{\delta_\lambda} \Delta \theta_0 - \frac{\Lambda_{12}}{\delta_\lambda} \Delta a_2^0 e^{-\lambda_r l^*},$$  \hspace{1cm} (4.3a)

$$\Delta a_2(t^*) = \frac{\lambda - \Lambda_{22}}{\delta_\lambda} \Delta a_2^0 - \frac{\Lambda_{21}}{\delta_\lambda} \Delta \theta_0 e^{-\lambda_r t^*},$$

$$- \frac{\lambda - \Lambda_{22}}{\delta_\lambda} \Delta a_2^0 - \frac{\Lambda_{21}}{\delta_\lambda} \Delta \theta_0 e^{-\lambda_r l^*}.$$  \hspace{1cm} (4.3b)

Note that both $\Delta \theta$ and $\Delta a_2$ vanish in the long-time limit.
FIG. 1. First stage in the evolution of $\theta_A(t^*)$ and $\theta_B(t^*)$ for $d = 3$, $\zeta^* = 1$, and $\gamma = 0.1$. The initial states are (a) $(\theta_A^0, a_{2A}^0) = (10, 0.5)$, $(\theta_B^0, a_{2B}^0) = (9, -0.35)$, (b) $(\theta_A^0, a_{2A}^0) = (2, 0.5)$, $(\theta_B^0, a_{2B}^0) = (1.8, -0.35)$, (c) $(\theta_A^0, a_{2A}^0) = (1.1, 0.5)$, $(\theta_B^0, a_{2B}^0) = (1.05, -0.35)$, and (d) $(\theta_A^0, a_{2A}^0) = (0.9, 0.5)$, $(\theta_B^0, a_{2B}^0) = (0.85, -0.35)$. Circles correspond to the numerical solutions of Eqs. (4.3), whereas solid lines correspond to the linearized model, given by Eqs. (B4). The Mpemba effect is neatly observed in panels (a)–(c), and the linearized theory with $\theta_r = \theta_B^0$ gives a correct account thereof—although it deviates from the numerical solution of Eqs. (4.3) as time increases in panels (a) and (b), for which their initial temperatures are not close to the steady state. The inverse Mpemba effect is depicted in panel (d) and the linear theory also describes it correctly, but now we have chosen $\theta_r = \theta_A^0$.

B. Mpemba crossover

The accuracy of the linearized theory developed above for describing the Mpemba effect—and also the inverse Mpemba effect—is illustrated in Fig. 1. The linear theory remains valid even when the system is initially far from the steady state: the analytical expressions of the linearized theory, given by Eqs. (4.4), predict the crossover of the curves correctly but start to deviate from the “exact” numerical integration as time grows. For all these plots, an optimal choice for $\theta_r$ is the initial temperature of the sample that is closer to the steady state.

Let us now restrict ourselves to a situation in which the Mpemba effect is present. Thus, there exists a crossover time such that $\Delta \theta(t^*_c) = 0$. According to Eq. (4.3b), it is given by

$$t^*_c = \frac{1}{\delta \lambda} \ln \frac{\Lambda_{12} - (\lambda_\gamma - \Lambda_{11}) R^0}{\Lambda_{12} - (\lambda_\gamma - \Lambda_{11}) R^0}, \quad R^0 = \frac{\Delta \theta^0}{\Delta a^0_2}. \quad (4.4)$$

The crossover time $t^*_c$ depends on the initial preparation only through the reference temperature $\theta_r$. This implies that $\theta_A \approx \theta^0_A \approx \theta^0_B$ and the ratio $R^0$ in this simplified description, for given values of $\zeta^*$ and $\gamma$. Note that we have chosen $\Delta \theta^0 > 0$ and, for the Mpemba effect to exist, we need that $\Delta a^0_2 > 0$, i.e., we have that $R^0 > 0$.

Figure 2 displays $t^*_c$ as a function of $R^0$ for some illustrative cases. Different panels correspond to different values of the reference temperature. In all of them, the crossover time $t^*_c$ vanishes in the limit as $R^0 \to 0$ and grows with $R^0$. Figure 2 also includes the values of the crossover time obtained from the numerical solution of Eqs. (2.4) for $a_{2A}^0 = 0.5$ and $a_{2B}^0 = -0.35$ with $\theta_B^0 = \theta_r$ in panels (a)–(c) and $\theta_A^0 = \theta_r$ in panel (d). It is observed that the agreement with Eq. (4.3) improves as $\gamma$ increases and $R^0$ decreases. Also, Eq. (4.3b) overestimates the crossover time for the direct Mpemba effect with initial temperatures far from that of the thermostat, while it tends to overestimate $t^*_c$ for the inverse Mpemba effect or when the initial temperatures are close to the thermostat one.

Equation (4.3b) shows that $t^*_c$ diverges in the limit as $R^0 \to R_{th}$, where $R_{th}$ is a threshold value for the ratio,
trolled by the strength of the drag nonlinearity $\gamma$.

Thus, the Mpemba effect disappears if $R^0 \geq R^0_{th}$; in this region $t^*_c$, as defined by Eq. (4.4), ceases to be a real number. In fact, if we define

$$\beta \equiv \frac{\lambda_- - \Lambda_{11}}{\lambda_+ - \Lambda_{11}} = 1 - \frac{\delta_\lambda}{\lambda_+ - \Lambda_{11}},$$

we can rewrite $t^*_c$ as

$$t^*_c = \frac{1}{\delta_\lambda} \ln \frac{1 - \beta R^0 / R^0_{th}}{1 - R^0 / R^0_{th}}.$$  \hspace{1cm} (4.7)

The emergence of the Mpemba effect is basically controlled by the strength of the drag nonlinearity $\gamma$. As expected on a physical basis, the Mpemba crossover takes place earlier as $\gamma$ increases. Throughout Fig. 2 the curves for $\gamma = 0.1$ lie above those for $\gamma = 0.2$. Thus, the smaller $\gamma$ is, the smaller the threshold value $R^0_{th}$ we find. Recall that the drag becomes linear in the limit as $\gamma \to 0^+$, for which the temperature obeys a closed first-order differential equation—independently of the value of the excess kurtosis, and the Mpemba effect is no longer present.

C. Phase diagram

A phase diagram in the $(\gamma, R^0)$ plane can be constructed, as illustrated in Fig. 3. The line $R^0 = R^0_{th}$ separates the regions in which the Mpemba effect is present ($R^0 < R^0_{th}$) and absent ($R^0 > R^0_{th}$), where $R^0_{th}$ is defined in Eq. (4.4). The range $0 < R^0 < R^0_{th}$ for which the Mpemba effect emerges increases with $\gamma$, $\zeta^*_0$, and $\theta_r$. It must be remarked that $\theta_r < 1$ corresponds to the inverse Mpemba effect, in which the system relaxes to equilibrium from below the steady temperature $T_s$.

The threshold values $R^0_{th}$ obtained from the numerical solution of Eqs. (3.6) for $a^0_{2, A} = 0.5$ and $a^0_{2, B} = -0.35$ are also shown in Fig. 3. The agreement with the simplified model is quite good, especially in panel (a), for which $\zeta^*_0 < 1$. In panel (b), for which $\zeta^*_0 \geq 1$, the linearized theory still gives a semi-quantitative picture and, notably, successfully captures the weak influence of both $\gamma$ and $\zeta^*_0$ on $R^0_{th}$ if $\zeta^*_0 \geq 1$. In any case, the linear model overestimates (underestimates) $R^0_{th}$ for $\theta_r = 10$ and 2 ($\theta_r = 1.05$ and 0.5), as anticipated from Fig. 2.

Interestingly, the maximum ratio relative to the reference temperature, $\theta_r^{-1} R^0_{th}$, keeps increasing with increasing $\theta_r$. At fixed $\theta_r$, the upper bound of $R^0_{th}$ corresponds...
to the limit $\gamma \to \infty$, which is independent of $\zeta_0^*$, namely

$$\lim_{\gamma \to \infty} R^0_{th} = \frac{2(d + 2) \theta_0 / (d - 2 + 12 \theta_0)}{1 + \sqrt{1 + 16(d + 2) \theta_0^2 / (d - 2 + 12 \theta_0)^2}}$$

(4.8)

Notwithstanding, in our modeling we are only retaining the first correction, quadratic in the velocities, in the drag coefficient $\zeta(v)$. Therefore, from a physical point of view, $\gamma$ is expected not to be very large; otherwise, higher order terms in the velocity should be incorporated into the drag coefficient.

D. Magnitude of the Mpemba effect

When the Mpemba effect is present, the temperature difference $\Delta \theta$ vanishes at the crossover time $t_\ast^\ast$. Since $\Delta \theta$ also vanishes in the long time limit as $t^\ast \to \infty$, there must exist a certain time $t_m^\ast > t_\ast^\ast$ where $|\Delta \theta(t^\ast)|$ reaches a local maximum. Therefore, one has that $|\Delta \theta(t^\ast)| \leq |\Delta \theta(t_m^\ast)|$ for any time $t^\ast > t_\ast^\ast$.

The above discussion can be used to introduce a quantitative measure of the magnitude of the Mpemba effect. Let us define

$$M_p \equiv |\Delta \theta(t_m^\ast)|$$

(4.9)
as a quantitative measure of its magnitude. From Eqs. (4.13) one finds

$$t_m^\ast = t_\ast^\ast + \frac{1}{\delta \lambda} \ln \frac{\lambda_+}{\lambda_-},$$

(4.10a)

$$\frac{M_p}{\Delta a_2^0} = \Lambda_{12} \left( \frac{1 - R^0 / R_{th}^0}{\lambda_+} \right)^{\frac{2}{\delta \lambda}} \left( 1 - \frac{\lambda_-}{1 - \beta R^0 / R_{th}^0} \right)^{\frac{2}{\delta \lambda}}.$$  

(4.10b)

Thus, the Mpemba magnitude $M_p$ depends on the initial differences $\Delta \theta_0$ and $\Delta a_2^0$ by a simple scaling law: the ratio $M_p / \Delta a_2^0$ is a function of the ratio $R^0 \equiv \Delta \theta_0 / \Delta a_2^0$. Figure 4 shows that the larger the ratio $R^0$ is, the smaller $M_p / \Delta a_2^0$ becomes. Of course, $M_p$ vanishes as $R^0$ approaches its threshold value $R_{th}^0$, as readily seen in Eq. (4.10b). Comparison with the values of $M_p$ obtained from the numerical solution of Eqs. (3.6) shows that the simple linearized model is qualitatively correct in capturing the dependence of the order of magnitude of $M_p$ on the parameters of the problem. In agreement with what was observed in Figs. 2 and 3, the linearized model tends to overestimate (underestimate) $M_p$ for the direct (inverse) Mpemba effect. Figure 4(c) shows that the prediction of $M_p$ is especially accurate if the initial temperatures are close to the equilibrium one: in that case, $M_p$ is slightly overestimated for small $R^0$, while it is slightly underestimated as $R^0$ approaches its threshold value $R_{th}^0$.

E. Reliability of the linear theory

The linear theory we have presented does not apply for all times, unless the reference temperature $\theta_r = \theta_\ast = 1$ and both initial temperatures $\theta_A^0$ and $\theta_B^0$ are close to the steady state. Nevertheless, as already stated before, our linear theory is not the standard linearization around the steady state but an approximate scheme to obtain a good approximation to the actual time evolution of the system in the early stage of its evolution, where the Mpemba effect is expected to come about. This means that our linear approach does have some limitations, as observed in Figs. 2–4. According to them, the linearized model becomes more accurate as $|\theta_r - 1|$ and $\zeta_0^*$ decrease.

While a complete account of the range of validity of the linear approximation is outside the scope of our paper, quite simple arguments can be presented in the limit of weak nonlinearity, $\gamma \to 0^+$. The behavior of the threshold $R_{th}^0$—below which the Mpemba effect is found—depends on those of the numerator, $\Lambda_{12}$, and its denominator, $\lambda_+ - \Lambda_{11}$. On the one hand, Eq. (4.11) tells us
that the former is linear in $\gamma$, $\Lambda_{12} = O(\gamma)$. On the other hand, the behavior of $\lambda_+ - \Lambda_{11}$ for small $\gamma$ depends on the sign of the function

$$
\varphi(\theta_r, \zeta_0^*) \equiv \frac{4(d-1)}{d(d+2)\zeta_0^*} \theta_r^{3/2} - \theta_r + 2.
$$

Specifically, it can be readily shown that

$$
\lambda_+ - \Lambda_{11} \approx \begin{cases} 
\frac{2\zeta_0^* \varphi(\theta_r, \zeta_0^*)}{\theta_r}, & \varphi(\theta_r, \zeta_0^*) > 0, \\
-\frac{8\zeta_0^*(d+2)\theta_r^3}{\varphi(\theta_r, \zeta_0^*)}, & \varphi(\theta_r, \zeta_0^*) < 0.
\end{cases}
\quad (4.12)
$$

and thus

$$
R_{th}^0 \sim \begin{cases} 
\frac{(d+2)\theta_r^3}{\varphi(\theta_r, \zeta_0^*)}, & \varphi(\theta_r, \zeta_0^*) > 0, \\
-\frac{1}{4\theta_r} \frac{\varphi(\theta_r, \zeta_0^*)}{\gamma}, & \varphi(\theta_r, \zeta_0^*) < 0.
\end{cases}
\quad (4.13)
$$

In the limit as $\gamma \to 0^+$, the drag becomes linear, the temperature obeys a closed equation and no Mpemba effect can be present in the system. This is consistent with the behavior found for the threshold $R_{th}^0$ when $\varphi(\theta_r, \zeta_0^*) > 0$: therein, $R_{th}^0 \to 0$. However, $R_{th}^0 \to \infty$ when $\varphi(\theta_r, \zeta_0^*) < 0$: this is an unphysical result that makes us conclude that the simplified linear model ceases to be reliable if $\varphi(\theta_r, \zeta_0^*) < 0$ and $\gamma \ll 1$. The locus $\varphi(\theta_r, \zeta_0^*) = 0$ is plotted in Fig. 5.

The above discussion should not be employed to disregard the linear model directly when $\varphi(\theta_r, \zeta_0^*) < 0$: the linearization can be useful unless $\gamma$ is very small. We can estimate the value of $\gamma$ for which the linear theory is no longer accurate, by asking the estimate for $R_{th}^0$ in (4.13) to be large. This leads to the condition

$$
\gamma \ll \gamma_{\ell} \equiv \frac{|\varphi(\theta_r, \zeta_0^*)|}{4\theta_r}.
\quad (4.14)
$$

We illustrate the above result in Fig. 6 for the three-dimensional case, specifically for $\zeta_0^* = 5$ and $\theta_r = 9$, in which case we have that $\varphi(\theta_r, \zeta_0^*) \approx -4.12$ and $\gamma_{\ell} \approx 0.114$. While the (weak) Mpemba effect predicted by the linearized model with $\gamma = 0.001$ is actually absent, the model succeeds in locating the crossover time if $\gamma = 0.1$, which is quite close to $\gamma_{\ell}$. Note that $\gamma = 0.1$ corresponds to the case in which the mass of the Brownian particles and that of the surrounding fluid are identical, as shown in Appendix A.
are

\[ \theta \]

that controls the failure of the linearized theory.

\[ \gamma \]

namely (a) \[ \zeta \theta \]

FIG. 6. Evolution of \[ \Delta \theta(t^*) = \theta_A(t^*) - \theta_B(t^*) \] for \( d = 3, \zeta_0^* = 5 \). The two panels correspond to small values of \( \gamma \), namely (a) \( \gamma = 0.001 \) and (b) \( \gamma = 0.1 \). The initial states are \( \theta_A^0, a_{2A}^0 = \{10, 0.5\} \), \( \theta_B^0, a_{2B}^0 = \{9, -0.35\} \). Circles correspond to the numerical solutions of Eqs. (3.17), whereas solid lines correspond to the linearized model (1.9) with \( \theta_r = \theta_0^* \). The value of \( \gamma \) in panel (a) verifies the condition (4.14) that controls the failure of the linearized theory.

V. CONCLUSIONS

We have neatly observed the Mpemba effect in a molecular gas with nonlinear drag. For the Mpemba effect—and also for the inverse Mpemba effect, in which the initially cooler sample heats sooner—to emerge the initially hotter sample must have a sufficiently larger value of the excess kurtosis \( \alpha_2 \). The larger \( \alpha_2 \) is, the larger the cooling rate becomes. This behavior is completely analogous to that found in a granular gas of smooth hard spheres\[20\].

Analytical predictions have been obtained, in a wide range of values of the system parameters, within a linearized model. The linearization is carried around a reference temperature—specifically, the initial temperature of the sample that is closer to the equilibrium value, not around the steady temperature. Therefore, our analytical framework is not limited to near-equilibrium situations. Within this scheme, we have found semi-quantitatively accurate expressions for (i) the crossover time, (ii) the maximum value of the initial temperature difference, and (iii) the magnitude of the Mpemba effect. Also, we have looked into the limitations of the linearized model, especially for small values of the parameter \( \gamma \) characterizing the nonlinearity.

This work also opens avenues for further research. It is interesting to consider in more detail some specific limits of the present model, which are physically relevant: (i) small nonlinearity \( \gamma \ll 1 \), which appears naturally as the first correction to the usual linear drag, and (ii) time scale separation between viscous drag and collisions, i.e., either \( \zeta_0^* \ll 1 \) or \( \zeta_0^* \gg 1 \). In both cases, a systematic—mainly perturbative—analytical approach seems to be feasible. Also, it is important to deepen our understanding of aging phenomena in this molecular gas. Specifically, looking into the Kovacs effect\[20,48,49\] which has attracted a lot of attention lately\[48,49,50,61\] is compelling.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.
Appendix A: Hard interaction between Brownian particles and background fluid

In Refs. [31 33] the authors consider the emergence of nonlinear Brownian motion when an ensemble of “heavy” Brownian particles (mass $m$, number density $n$) moves in a bath modeled as a dilute gas of “light” particles (mass $m_{bf}$, number density $n_{bf}$), which is at equilibrium at temperature $T_s$. A velocity-dependent drag coefficient $\zeta(v)$ is obtained as an expansion in powers of the mass ratio $m_{bf}/m$, which is assumed to be small.

The coefficients of the expansion of $\zeta(v)$ are given in terms of integrals that involve the differential cross section for the interaction between the Brownian particles and the bath particles. Explicit expressions for $\zeta(v)$ can be derived when simple potentials are employed for this interaction. For example, all particles are considered to be three-dimensional hard spheres in Ref. [33], both the Brownian particles and the particles in the background dilute gas. With this assumption, it is found that

$$\zeta(v) = \frac{8}{15} n_{bf} S \sqrt{\frac{2 m_{bf}^2}{\pi k_B T_s}} \frac{m + m_{bf}}{m + m_{bf}}.$$  \hspace{1cm} (A1)

Above, $S$ is the total cross section, i.e.,

$$S = \frac{\pi (\sigma + \sigma_{bf})^2}{4},$$  \hspace{1cm} (A2)

where $\sigma$ and $\sigma_{bf}$ are the diameters of the Brownian particles and the background fluid particles, respectively. Equation (A1) is valid up to order $(m_{bf}/m)^{3/2}$ and for not too large velocities, i.e., velocities that lie in the thermal range $mv^2/2k_B T_s = O(1)$.

By comparing Eq. (A2) with Eq. (2.6), we have

$$\zeta_0 = \frac{2}{3} m_{bf} (\sigma + \sigma_{bf})^2 \sqrt{\frac{\pi k_B T_s}{m}} \frac{\sqrt{2mn_{bf}}}{m + m_{bf}},$$  \hspace{1cm} (A3)

which is proportional to $\sqrt{T_s}$, and

$$\gamma = \frac{m_{bf}}{10m}.$$  \hspace{1cm} (A4)

which is expected to be small, $\gamma \lesssim 0.1$—its value for $m_{bf} = m$.

In the framework developed in this paper, we measure time in terms of the number of collisions of Brownian particles among themselves. Therefore, our evolution equations involve the dimensionless low-velocity drag coefficient $\zeta_0' = \zeta_0 \tau_s$, introduced in Eq. (6.7a). For hard spheres ($d = 3$) in the Boltzmann limit ($g(\sigma^+) = 1$), the characteristic time $\tau_s$ for Brownian-Brownian collisions is

$$\tau_s^{-1} = 2n \sigma^2 \sqrt{\frac{\pi k_B T_s}{m}}.$$  \hspace{1cm} (A5)

Straightforward algebraic manipulation leads to

$$\zeta_0' = \frac{2n_{bf}}{3n} \left(1 + \frac{\sigma_{bf}}{\sigma}\right)^2 \frac{\sqrt{\delta \gamma}}{1 + 10\gamma}.$$  \hspace{1cm} (A6)

Therefore, we have that $\zeta_0'$ depends on three dimensionless quantities: the density ratio $n_{bf}/n$, the diameter ratio $\sigma_{bf}/\sigma$, and the mass ratio $m_{bf}/m$, as measured by $\gamma$. This means that, even in the “natural” heavy Brownian limit $m_{bf}/m \ll 1$ or $\gamma \ll 1$, $\zeta_0'$ varies across a large range of values. For a given problem, its specific value depends on $n_{bf}/n$ and $\sigma_{bf}/\sigma$, but not on the steady temperature: The ratio of time scales associated with the viscous drag and Brownian–Brownian collisions is independent of the temperature of the bath.

The simple expressions for $\gamma$ and $\zeta_0'$, Eqs. (A4) and (A6), hold for hard-sphere interaction in the Boltzmann limit. More complicated behaviors may be found in other situations, but we expect the qualitative picture derived here to be still valid: While the range of $\gamma$ is somehow limited, that of $\zeta_0'$ is not necessarily so. This explains why we have restricted ourselves to $\gamma < 0.25$ throughout the paper but treated $\zeta_0'$ as an independent parameter, which may attain both small and large values.

Appendix B: Solution of the linearized system

Our starting point is the nonlinear system [36], written in the Sonine approximation. First, we introduce the deviation of the temperature from a certain reference temperature $\theta_r$ by

$$\Psi(t^*) = \theta(t^*) - \theta_r.$$  \hspace{1cm} (B1)

Second, we linearize Eq. (3.6) with respect to $\Psi(t^*)$ and $a_2(t^*)$, which leads to

$$\begin{align*}
(\dot{\Psi})_{a_2} &= - \left( \Lambda_{11} \Lambda_{12} \Lambda_{21} \Lambda_{22} \right) (\dot{\Psi})_{a_2} + \left( \Lambda_{11} \Lambda_{12} \Lambda_{21} \Lambda_{22} \right) (\dot{C}_1)_{a_2}, \quad (B2)
\end{align*}$$

where

$$C_1 = 2\zeta_0' (1 - \theta_r) [1 + \gamma (d + 2) \theta_r],$$  \hspace{1cm} (B3a)

$$C_2 = 8\zeta_0' \gamma (1 - \theta_r),$$  \hspace{1cm} (B3b)

and the $\Lambda_{ij}$ have been defined in Eq. (4.14). The solution of the simplified linear model [12] yields

$$\begin{align*}
\Psi(t^*) &= D_1 + \frac{(\lambda_+ - \Lambda_{11}) (\Psi^0 - D_1) - \Lambda_{12} (a_2^0 - D_2)}{\delta \lambda e^{\lambda_+ t^*}} - \frac{(\lambda_- - \Lambda_{11}) (\Psi^0 - D_1) - \Lambda_{12} (a_2^0 - D_2)}{\delta \lambda e^{\lambda_- t^*}}, \quad (B4a)
\end{align*}$$

$$\begin{align*}
a_2(t^*) &= D_2 + \frac{(\lambda_+ - \Lambda_{22}) (a_2^0 - D_2) - \Lambda_{21} (\theta^0 - D_1)}{\delta \lambda e^{\lambda_+ t^*}} - \frac{(\lambda_- - \Lambda_{22}) (a_2^0 - D_2) - \Lambda_{21} (\theta^0 - D_1)}{\delta \lambda e^{\lambda_- t^*}}, \quad (B4b)
\end{align*}$$

where $\lambda_{\pm}$ are the eigenvalues of the matrix $\Lambda$ defined in Eq. (132) and we have defined the parameters

$$\begin{align*}
D_1 &= \frac{\Lambda_{22} C_1 - \Lambda_{12} C_2}{\Lambda_{11} \Lambda_{22} - \Lambda_{12} \Lambda_{21}}, \quad D_2 = \frac{\Lambda_{11} C_2 - \Lambda_{21} C_1}{\Lambda_{11} \Lambda_{22} - \Lambda_{12} \Lambda_{21}}. \quad (B5)
\end{align*}$$
Neither $\Psi$ nor $\theta$ reach their actual equilibrium values in the long-time limit, unless $\theta = 1$. This is not a problem for the analysis carried out in the main text, because this linear theory is only used for the early stage of the time evolution, in which the Mpemba effect may emerge.

Now, let us consider two different initial states: A, with initial values of the temperature and the excess kurtosis $\{\theta_A, d_2^A\}$, and B, with initial values $\{\theta_B, d_2^B\}$. The linear theory makes it possible to write analytical predictions for the differences between their respective time evolutions, i.e., $\Delta \theta(t) = \theta_A(t) - \theta_B(t) = \Psi_A(t^*) - \Psi_B(t^*)$ and $\Delta d_2(t) = d_2^A(t) - d_2^B(t)$. Making use of Eq. (15), we arrive precisely at Eq. (13) in the main text.

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