Kinetics of ensembles with variable charges

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Kinetics of particle ensembles with variable charges is investigated. It is shown that the energy of such ensembles is not conserved in the interparticle collisions. The case when the equilibrium charge depends on the particle coordinate is studied, and the collision integral describing the momentum and energy transfer in collisions is derived. Solution of the resulting kinetic equation shows that the system is unstable—the mean thermal energy exhibits explosion-like growth, diverging at a finite time.

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I. INTRODUCTION

Many problems in physical kinetics are treated using Hamiltonian description. *Non-Hamiltonian kinetics*, on the other hand, is far less well developed—for the simple reason that such systems have not received as much attention (although they are ubiquitous) and have usually been dealt with by separating time scales and reverting back to the Hamiltonian formalism.

One of the remarkable features distinguishing complex (dusty) plasmas from usual (multispecies) plasmas is that charges on the grains are not constant, but fluctuate in time around some equilibrium value which, in turn, is some function of spatial coordinates [1–6]. The fluctuation (charging) time scale and the mean charge generally depend on the charging mechanism operating in a plasma (see, e.g., [3,5]). Ensembles of particles with variable charges are non-Hamiltonian systems and, therefore, the use of thermodynamic potentials to describe them is not really valid. An appropriate way to investigate their evolution is to use the kinetic approach.

Both in laboratory and space plasmas, the time scales associated with random fluctuations and establishing the equilibrium charge are normally many orders of magnitudes shorter than the time scales related to the grain dynamics [1,3,7–9]. In many cases this fact allows us to separate the charging kinetics from the kinetics of the grains themselves (e.g., [2,4]), and consider dust grains as an ensemble of particles with *given* characteristics of charge variation.

II. KINETICS

Kinetics of ensembles with variable charges have not been studied systematically so far, and the main objective of this paper is to demonstrate some extraordinary features peculiar to such systems. In order to highlight properties of ensembles with variable charges, let us consider the simplest case when no external fields are present. Then the particle kinetics is solely determined by the mutual collisions. The kinetic equation is

$$\dot{f} = \mathrm{St} f, \tag{1}$$

where $\mathrm{St} f$ denotes the collision integral. The major peculiarity of particles with variable charges is that the *collisions do not conserve the energy*, because charge variations cause fluctuations of the interparticle forces during the collisions. For instance, when the equilibrium charge of a particle is a function of coordinates, $q(\mathbf{r})$, then the charge variations are associated with the particle motion and characterized by the gradient ∇q. For a pair of particles interacting within the range λ (screening length), the average displacement during the interaction is also of the order of $\sim\lambda$. Therefore, the relative variation of the particle energy caused by a collision is $|\delta E|/E \sim |\nabla q/q|\lambda$. Note that random variations of charges associated with temporal fluctuations of, e.g., local plasma density/temperature, UV radiation, etc. [3], also contribute to the energy variations [10].

A. Collision integral

In this paper we investigate the kinetics of particles with a *spatial dependence of the equilibrium charge*—the charge variations are due to the particle motion, random fluctuations are neglected. The latter formally corresponds to the limit of infinitely short charging time [3,7,8] (the case of charge fluctuations related to dust discreteness, i.e., finite charging time, will be considered in future work [10]). In order to simplify the calculations, yet keeping the physical essence of the effect, we will consider the kinetics of a small (gaseous) fraction of "light" particles [with spatially dependent charge $q(\mathbf{r})$] added to "heavy" particles (with charge Q). This situation, when particles have very different masses, is quite normal for complex plasma experiments (e.g., the "Plasma Kristall Experiment-Nefedov" experiment onboard International Space Station [11], where the particle sizes differ by factor of 2, or the follow-up experiment "PK-3 Plus,"where this range is about 20), and is ubiquitous in space dusty plasmas (where the grains range from tenths of microns up to meters in size, e.g., [12]).

Due to the small concentration of light particles (and also their relatively small charges), one can neglect their mutual collisions and consider only pair collisions with the heavy particles. Under these approximations the collision integral becomes linear. Moreover, if the relative variation of the particle energy in collisions is small—namely, if the following inequality for the charge gradient is satisfied,

$$\epsilon \equiv |\nabla q/q|\lambda \ll 1 \qquad (2)$$

(i.e., spatial scale of the charge variation is much larger than the screening length), then one can divide the collision integral into two parts—corresponding to the variation of the absolute value of momentum, and of the momentum direction—and present the first part in the differential Fokker-Planck form, expanding over the smallness parameter ϵ. For simplicity we neglect the motion of the large particles. Then the collision integral is [13,14]

$$\mathrm{St}\,f = \frac{1}{p^2}\frac{\partial}{\partial p}\left[\mathcal{A}p^2 f + \frac{\partial}{\partial p}(\mathcal{B}p^2 f)\right] + N\frac{p}{m}\int [f(p,\theta') - f(p,\theta)]\,d\sigma. \qquad (3)$$

The charge (of light particles) is assumed to have constant gradient $\boldsymbol{\nabla} q$; the distribution function is symmetric with respect to the gradient, and thus depends on two variables: the absolute value of the momentum p and the angle θ between the gradient and the momentum vector. The differential cross section $d\sigma$ corresponds to the scattering between θ and θ', with $\cos\theta' = \cos\theta\cos\alpha + \sin\theta\sin\alpha\cos(\varphi-\varphi')$ expressed in terms of the scattering angle α [14]. As usual, the Fokker-Planck coefficients [13], $\mathcal{A} = -\langle\delta p\rangle/\delta t$ and $\mathcal{B} = \langle(\delta p)^2\rangle/2\delta t$, are averaged over many collisions occurring within δt. Variation (of the absolute value) of momentum in a collision, δp, is related to the energy variation via $\delta E \simeq p\,\delta p/m$. From Eq. (A2) we obtain $\delta p/p \sim \epsilon(\cos\theta + \cos\theta')S(\alpha)$, which yields after averaging the coefficients

$$\mathcal{A} = -\frac{\epsilon p^2}{m\ell}c_1\cos\theta,$$

$$\mathcal{B} = \frac{\epsilon^2 p^3}{2m\ell}(c_2\cos^2\theta + c_3). \qquad (4)$$

It is convenient to introduce the free path $\ell = (N\sigma_{\rm tr})^{-1}$ expressed in terms of the momentum transfer cross section, $\sigma_{\rm tr} = \int(1-\cos\alpha)d\sigma$, and the concentration of large particles, N. The free path is generally a function of p (e.g., the screened Coulomb interaction often reduces to the hard spheres limit, with a weak logarithmic dependence on p for the free path [15]). The Fokker-Planck coefficients are determined by the dimensionless "scattering constants:"

$$c_1 = \int (1+\cos\alpha)S(\alpha)d\sigma/\sigma_{\rm tr},$$

$$c_2 = \tfrac{1}{2}\int (1 + 4\cos\alpha + 3\cos^2\alpha)S^2(\alpha)d\sigma/\sigma_{\rm tr},$$

$$c_3 = \tfrac{1}{2}\int \sin^2\alpha\, S^2(\alpha)\,d\sigma/\sigma_{\rm tr}.$$

B. Polynomial expansion

We expand the distribution function into a series of Legendre polynomials and keep the first two terms:

$$f(p,\theta,t) = f_0(p,t) + f_1(p,t)\cos\theta. \qquad (5)$$

Substituting Eqs. (4) and (5) in Eq. (3), after the usual procedure [14] we derive from Eq. (1) the following expressions for the symmetric and asymmetric parts of the distribution function:

$$\dot{f}_0 = \left(\frac{1}{3}c_2 + c_3\right)\frac{\epsilon^2}{mp^2}\frac{\partial^2}{\partial p^2}\left(\frac{p^5}{\ell}f_0\right) - \frac{1}{3}c_1\frac{\epsilon}{mp^2}\frac{\partial}{\partial p}\left(\frac{p^4}{\ell}f_1\right),$$

$$\dot{f}_1 = -\frac{p}{m\ell}f_1 - c_1\frac{\epsilon}{mp^2}\frac{\partial}{\partial p}\left(\frac{p^4}{\ell}f_0\right). \qquad (6)$$

One can see from Eqs. (6) that there is a hierarchy of time scales characterizing the evolution of f_0 and f_1: The time scale for the asymmetric part is equal to the collision-free time $\tau_{\rm coll} = m\ell/p$. The symmetric part evolves at much longer time scales $\sim\epsilon^{-2}\tau_{\rm coll}$. This allows us to decouple Eqs. (6). From the second equation we derive

$$f_1(p,t) \simeq -c_1\frac{\epsilon\ell}{p^3}\frac{\partial}{\partial p}\left(\frac{p^4}{\ell}f_0\right)(1 - e^{-pt/m\ell}). \qquad (7)$$

Equation (7) demonstrates that after a few collisions (at $t \gtrsim \tau_{\rm coll}$) the asymmetric part rapidly converges to the quasistationary solution determined by the kinetics of the "slow" symmetric part. Substituting this asymptotic solution for f_1 into the first equation (6), we finally derive

$$\dot{f}_0 \simeq \left(\frac{1}{3}c_1^2 + \frac{1}{3}c_2 + c_3\right)\frac{\epsilon^2}{mp^2}\frac{\partial^2}{\partial p^2}\left(\frac{p^5}{\ell}f_0\right) - \frac{1}{3}c_1^2\frac{\epsilon^2}{mp^2}\frac{\partial}{\partial p}\left(\frac{p^4}{\ell}f_0\right). \qquad (8)$$

C. Analysis of moments: Energy growth

Although Eq. (8) can be solved analytically, it is much more useful to analyze the kinetics of the derived equation in terms of p-moments. The zero moment yields the conservation of the particle density, $4\pi\int p^2 f_0\,dp = 1$ (we normalized the distribution function to unity). The second moment corresponds to the mean kinetic energy $\mathcal{E} = 4\pi\int(p^2/2m)p^2 f_0\,dp$. We obtain

$$\dot{\mathcal{E}} = \epsilon^2\left(\frac{2}{3}c_1^2 + \frac{1}{3}c_2 + c_3\right)\left(\frac{4\pi}{m^2}\right)\int (p^5/\ell)f_0 dp > 0. \qquad (9)$$

The derived equation shows that nonlinear ($\propto\epsilon^2$) terms in the Fokker-Planck expansion cause a *growth of mean energy* irrespective of the initial state, i.e., the system is always unstable.

It is remarkable that the most common type of the dissipation—the friction caused by collisions with neutral gas molecules—can be easily incorporated in our calculations in the Fokker-Planck form [13,14], with the following additional term to the collision integral (3):

$$\mathrm{St}_n f = \gamma \frac{\partial}{\partial \mathbf{p}} \left(\mathbf{p} f + m T_n \frac{\partial f}{\partial \mathbf{p}} \right),$$

where T_n is the temperature of the molecules and γ is the frictional damping rate in the corresponding Langevin equation. The friction yields the additional term, $-2\gamma(\mathcal{E}-\frac{3}{2}T_n)$, in Eq. (9) and, hence, sets up a threshold for the instability. To derive this threshold, we evaluate the integral in Eq. (9). From the Hölder inequality we obtain $\int p^5 f_0\, dp \geqslant (\int p^4 f_0 dp)^{3/2} (\int p^2 f_0 dp)^{1/2} \propto \mathcal{E}^{3/2}$. Normalizing the energy by the neutral gas temperature, $\tilde{\mathcal{E}} = \mathcal{E}/T_n$, and assuming for simplicity $\ell =$const, we derive the following equation for the *lower energy bound*:

$$\dot{\tilde{\mathcal{E}}} \geqslant 2\sqrt{2}\epsilon^2 (\tfrac{2}{3}c_1 + \tfrac{1}{3}c_2 + c_3) \langle \tau_{\mathrm{coll}} \rangle^{-1} \tilde{\mathcal{E}}^{3/2} - 2\gamma (\tilde{\mathcal{E}} - \tfrac{3}{2}), \tag{10}$$

where $\langle \tau_{\mathrm{coll}} \rangle = \ell / v_T$ is the "mean" collision-free time, with $v_T = \sqrt{T_n/m}$. From the requirement $\dot{\tilde{\mathcal{E}}} > 0$ for any $\tilde{\mathcal{E}}$ we obtain the sufficient condition for the instability,

$$\gamma \langle \tau_{\mathrm{coll}} \rangle < \tfrac{3}{2} \epsilon^2 (2c_1 + c_2 + 3c_3). \tag{11}$$

The solution of Eq. (10) diverges at a finite (critical) time, $\tilde{\mathcal{E}} \propto (1 - t/t_{\mathrm{cr}})^{-2}$, provided Eq. (11) is satisfied. The critical time scales as $t_{\mathrm{cr}} \sim \epsilon^{-2} \langle \tau_{\mathrm{coll}} \rangle$. Thus, we have an explosionlike energy growth. We should note, however, that at sufficiently high energies the free path is no longer a constant or a weak (logarithmic) function of momentum [15]—it starts increasing with p, and once $\ell(p)$ grows faster than linearly, the instability is saturated.

Equation (11) allows us to evaluate the physical conditions when the charge gradients should cause the energy growth. For instance, let us consider typical laboratory experiments with microsize particles of temperature ~ 1 eV and density $\sim 10^3$ cm^{-3} embedded into a plasma with the screening length $\lambda \sim 1$ mm. The resulting mean collision free time is $\langle \tau_{\mathrm{coll}} \rangle \sim 3 \times 10^{-2}$ s [15]. Assuming the spatial scale of the "natural" charge gradients in the presheath region about $\simeq 5$ mm, i.e., five times larger than the screening length [16], we get for the smallness parameter $\epsilon^2 \sim 3 \times 10^{-2}$ and, hence, the instability condition is $\gamma \lesssim 1$ s^{-1}. This corresponds to fairly realistic pressures about 1 Pa or less. "Artificial" charge gradients at much shorter spatial scales can be easily created by means of, e.g., external UV radiation [17,18] partially focused on the dust clouds, and then the heating should be possible at much higher pressures. In space plasmas (e.g., in the interstellar clouds, where the charge gradients can be created by inhomogeneous cosmic radiation [1]) the condition to trigger the heating might be substantially relaxed, due to much lower ambient pressure.

Important remark: The total energy consists of the thermal and drift parts, $\mathcal{E} = \mathcal{T} + \mathcal{K}$, with $\mathcal{K} = \langle \mathbf{p} \rangle^2 / 2m$ (due to symmetry, the drift is along the charge gradient). After a few collisions, the mean drift momentum $\langle \mathbf{p} \rangle \propto \int p^3 f_1 dp$ converges to the quasi-stationary value [see Eq. (7)]. In the case $\ell =$const the drift asymptotically vanishes: For instance, assuming the Boltzmann distribution for f_0 we derive using Eq. (7) that $\langle \mathbf{p} \rangle \propto t^{-4}$ at $t \gtrsim \langle \tau_{\mathrm{coll}} \rangle$. Therefore, $\mathcal{K} \to 0$ and, hence, $\mathcal{E} \to \mathcal{T}$, i.e., the particles are completely thermalized due to collisions. If the free path is a function of momentum, $\ell = \ell(p)$, then $\langle \mathbf{p} \rangle$ converges with time to some nonzero value $\propto \epsilon \int (\ell_p' / \ell) p^4 f_0\, dp$. Assuming logarithmic (or power) dependence for $\ell(p)$ we obtain: $\ell_p' / \ell \propto p^{-1}$, and, hence, the mean drift momentum tends to $\propto \epsilon \int p^3 f_0\, dp \leqslant \epsilon (\int p^2 f_0\, dp)^{1/2} (\int p^4 f_0\, dp)^{1/2} \sim \epsilon \sqrt{\mathcal{E}}$. Therefore, the drift part of the energy remains relatively small, $\mathcal{K} \propto \epsilon^2 \mathcal{E}$, i.e., most of the energy is always transmitted into particle heating.

D. Applicability

In our calculations we neglected the explicit dependence of the solution on spatial coordinates. In fact, spatial charge inhomogeneity, $q(\mathbf{r})$, implies the spatial dependence of the cross section in the collision integral (3) and results in the appearance of an additional term $(\mathbf{p}/m) \boldsymbol{\nabla} f$ in Eq. (1). This inhomogeneity, however, is weak and is determined by the smallness parameter ϵ. Expanding $\mathrm{St} f$ into a series over ϵ and assuming a homogeneous initial distribution $f|_{t=0}$, the resulting corrections to "homogeneous" solution (6) are of higher order in ϵ. Hence, they were not considered further. Also, we neglected the motion of heavy particles, which formally corresponds to a zero light-to-heavy particle mass ratio, $m/M \to 0$. In fact, the kinetics of light particles is not altered significantly provided the rate of energy exchange, which is $\sim (m/M) \langle \tau_{\mathrm{coll}} \rangle^{-1}$ [14], is less than the energy growth rate, $\sim \epsilon^2 \langle \tau_{\mathrm{coll}} \rangle^{-1}$. This implies that the condition $m/M \lesssim \epsilon^2$ must be satisfied. Important remark: The studied above example with ensembles of light particles was chosen solely to simplify the algebra; it is not crucial. On the other hand, the very fact that the particle energy is not conserved in mutual collisions is a *universal* intrinsic feature peculiar to any ensemble with variable charges. The mechanism of the energy growth should therefore be generic and apply to different ensembles as well (e.g., "monodisperse" particles).

E. Numerical calculations

In order to verify the obtained theoretical results we performed 2D molecular dynamics simulations of a Yukawa system of particles with variable charges. The particles were divided into two groups and randomly distributed over a square of size $L = 1$ cm: 5000 "large" (motionless) particles with constant charges $Q = 3 \times 10^4 e$ and 5000 "small" (movable) particles with an initial (isotropic) Maxwellian velocity distribution and charges depending linearly on the z-coordinate, with $q = 3 \times 10^3 e$ at the left boundary $z = 0$. Periodic boundary conditions were chosen for the particle velocities at $z = 0$ and $z = L$. The transverse boundaries, $y = 0$ and $y = L$, were "mirror walls." The screening length for the Yukawa interaction was $\lambda = 30$ μm (about one third of the mean interparticle distance). Figure 1 shows the mean kinetic energy of "small" particles, $\tilde{\mathcal{E}}$ (normalized to the initial temperature), as function of time for different values of the charge gradient ϵ. Of course, the mean energy remains constant without the gradient. For finite ϵ, the energy scales

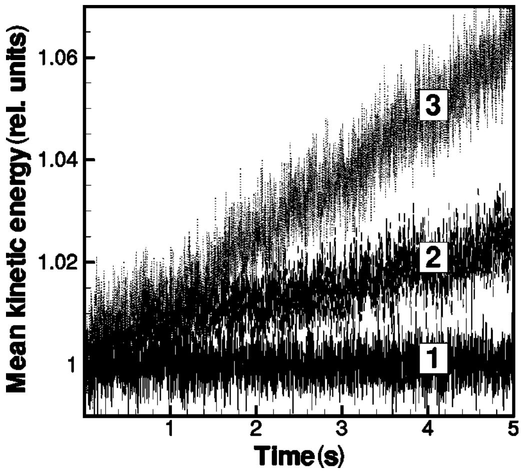

FIG. 1. Mean kinetic energy of particles, $\tilde{\mathcal{E}}$ (normalized to the initial temperature), versus time t. Curves correspond to different values of the dimensionless charge gradient: $\epsilon=0$ (constant charge, 1), $\epsilon=10^{-2}$ (2), and $\epsilon=1.5\times10^{-2}$ (3).

initially as $\tilde{\mathcal{E}}\propto\epsilon^2 t$, in agreement with Eq. (10). The computed magnitude of the energy growth coincides with the theoretical prediction as well [given the combination of the scattering constants $c_{1,2,3}$ in Eq. (10) equal to $\simeq 0.5$]. In agreement with the theory, the drift part of the kinetic energy rapidly decreases in the simulations after a few collisions (at $t \lesssim 10^{-1}$ s for this example) and is negligible at later stages, $\mathcal{K}\lesssim 10^{-2}\mathcal{E}$. Therefore, the plotted curves actually show the thermal part $\mathcal{T}$ of the mean energy.

III. CONCLUSIONS

In conclusion, we investigated the kinetics of particle ensembles with variable charges. Those are non-Hamiltonian systems, and the total energy is not conserved in mutual particle collisions. We focused on the case of inhomogeneous charge distribution—when the equilibrium charge depends on the particle coordinate—and derived the collision integral which describes the momentum and energy transfer in collisions. Solution of the resulting kinetic equation shows that the system is unstable—the mean thermal energy exhibits explosionlike growth, diverging at a finite time. The obtained solutions can be of significant importance for laboratory dusty plasmas as well as for space plasma environments, where inhomogeneous charge distributions are often present. For instance, the instability can cause the dust heating in low-pressure complex plasma experiments and be responsible, e.g., for the melting of plasma crystals, or might operate in protoplanetary disks and, thus, affect kinetics of the planet formation (e.g., [19]), etc.

Concerning the energy source providing the instability (divergence of thermal energy) in ensembles with variable charges, we point out that charged grains can be considered as a subsystem of elementary charges (electrons and ions) "bound" to the grains, which along with the subsystem of "free" plasma charges forms a *complete ensemble* of charges. Even if the complete ensemble is in detailed balance and can be considered as a Hamiltonian system, there is a continuous exchange of energy between these two subsystems via the charging processes, so that the charged grains cannot be described by a Hamiltonian function. In addition, there always exist external sources (e.g., inhomogeneous ionization, UV radiation, etc.) which provide the energy influx into the whole system of charges including the subsystem of grains.

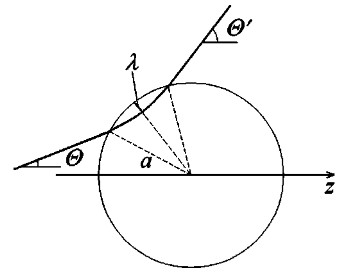

FIG. 2. Sketch of the particle interaction during the collision.

APPENDIX: VARIATION OF ENERGY IN COLLISIONS

Let us consider how the energy of a moving particle changes while it passes by a motionless particle. If the potential distribution around the motionless particle is $\Phi(\mathbf{r})$ and the charge of the moving particle is a certain function of spatial coordinates, $q(\mathbf{r})$, then the variation of the particle energy is

$$\delta E = -\int_C q\boldsymbol{\nabla}\Phi\, d\mathbf{r} = \int_C \Phi\boldsymbol{\nabla}q\, d\mathbf{r}, \tag{A1}$$

where the integration is taken along the trajectory $\mathbf{r}_C$ of the particle. In Eq. (A1) we took into account that $q\Phi$ equals zero at the ends of the path C. Assuming constant charge gradient pointed along the z-axis, we rewrite Eq. (A1) as follows:

$$\delta E = |\boldsymbol{\nabla} q| \int \Phi[\mathbf{r}_C(z)]dz = |\boldsymbol{\nabla} q|\langle\Phi\rangle L_z.$$

Here angle brackets denote the average of $\Phi(\mathbf{r}_C)$, and L_z is the displacement of the particle along the z-axis "during" the collision. Thus, the calculation of δE is essentially a mechanical problem of determining a particle trajectory $\mathbf{r}_C$. Generally speaking, this is a rather complicated task (see, e.g., [20,21]), and for purposes of this paper we just perform a scaling analysis based on asymptotic characteristics of the interaction between highly charged particles.

The electrostatic potential of a charged particle in a plasma is normally of a short range, because the particle is screened. For sufficiently high particle charges the pair interaction is usually reduced to the limit of elastic hard spheres [15,22,23]: For instance, for the screened Coulomb (Yukawa) potential one can approximate the interaction by a collision of a pointlike particle of energy E with a sphere of radius $a \simeq \lambda \ln(qQ/\lambda E)$ and the "elasticity depth" $\sim\lambda$, provided the radius a is much larger than the screening length λ, i.e., $\ln(qQ/\lambda E) \gg 1$. Figure 2 shows the sketch of such an interaction. Assuming scattering within the elasticity depth, the effective length is estimated as $L_z \sim \lambda S(\cos\theta + \cos\theta')$, where S is a function of the scattering angle α. Also, the average electrostatic energy for such interaction is of the order of the kinetic energy, i.e., $q\langle\Phi\rangle \sim E$. Therefore, we have the following estimation for the relative variation of the energy:

$$\delta E/E \sim \epsilon(\cos\theta + \cos\theta')S(\alpha), \tag{A2}$$

where $\epsilon = |\nabla q/q|\lambda \ll 1$ is the smallness parameter introduced in Eq. (2). The energy gain is positive if the particle displacement during the collision is in the direction of the charge gradient, and is negative in the opposite case. Note that such a representation of collisions is not valid at very small scattering angles $\alpha \lesssim \lambda/a \ll 1$. Nevertheless, since the collision cross section does not have singularities at small α, one can use Eq. (A2) to estimate magnitudes of the Fokker–Planck coefficients [Eq. (4)].

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