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Orientation of non-spherical particles in an axisymmetric random flow

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The dynamics of non-spherical rigid particles immersed in an axisymmetric random flow is studied analytically. The motion of the particles is described by Jeffery's equation; the random flow is Gaussian and has short correlation time. The stationary probability density function of orientations is calculated exactly. Four regimes are identified depending on the statistical anisotropy of the flow and on the geometrical shape of the particle. If $\boldsymbol{\lambda}$ is the axis of symmetry of the flow, the four regimes are: rotation about $\boldsymbol{\lambda}$, tumbling motion between $\boldsymbol{\lambda}$ and $-\boldsymbol{\lambda}$, combination of rotation and tumbling, and preferential alignment with a direction oblique to $\boldsymbol{\lambda}$.

Key words: multiphase and particle-laden flows, suspensions

1. Introduction

Non-spherical solid particles suspended in a moving fluid rotate and orient themselves under the action of the velocity gradient. Even at low concentrations, the orientational dynamics of non-spherical particles can influence the rheological properties of a suspension, namely the intrinsic viscosity and the normal stress coefficients (Bird *et al.* 1977; Larson 1999). This phenomenon has diverse practical applications. In the turbulent regime, for instance, the injection of rod-like polymers in a Newtonian fluid can produce a considerable reduction of the turbulent drag, with this effect being routinely exploited to reduce energy losses in pipelines (Gyr & Bewersdorff 1995). The study of the orientation of non-spherical particles in a fluid flow also has numerous applications in the natural sciences. Amongst them it is worth mentioning the swimming motion of certain biological micro-organisms (Saintillan & Shelley 2008; Koch & Subramanian 2011) and the formation of ice crystals in clouds (Chen & Lamb 1994). This latter phenomenon plays a crucial role in processes such as rain initiation and radiative transfer.

The starting point for understanding the properties of a dilute suspension is the motion of an isolated particle in a given flow field. Analytical results on the dynamics of a non-spherical particle have been obtained for various laminar flows, both steady and unsteady. Jeffery (1922) derived the equations of motion for an inertialess ellipsoid in a steady uniform shear flow at low Reynolds number. For a spheroid (i.e. an ellipsoid of revolution), Jeffery showed that the axis of symmetry of the particle performs a periodic motion on a closed orbit. Bretherton (1962) subsequently extended

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Jeffery's analysis to particles of a more general shape: he demonstrated that, except for certain very long particles, the dynamics of any body of revolution transported by a low-Reynolds-number shear is equivalent to that of a spheroid with an effective aspect ratio. Amongst bodies of revolution, rigid dumbbells and rods have been systematically investigated as prototypes of elongated particles (Bird, Warner & Evans 1971; Doi & Edwards 1986).

In Jeffery's (1922) and Bretherton's (1962) derivations, not only are fluid and particle inertia disregarded, but also Brownian fluctuations due to the collisions of the molecules of the fluid with the particles. Nevertheless, if a particle is sufficiently small, molecular diffusion does influence its orientational dynamics, as was shown by Leal & Hinch (1971) and Hinch & Leal (1972). The review article by Brenner (1974) collects analytical results on the motion of rigid neutrally buoyant bodies of revolution subject to a uniform velocity gradient and to Brownian fluctuations – a more recent review on this problem can be found in Petrie (1999). Finally, even in simple laminar flows, the orientation of particles can form complex patterns; this behaviour was studied by Szeri (1993) in the context of the theory of dynamical systems.

In turbulent flows and in some chaotic flows, the velocity gradient exhibits small-scale fluctuations. Thus, if a particle is larger than the Kolmogorov scale ℓ_K, the velocity gradient can vary appreciably over the size of the particle and Jeffery's assumption of a spatially uniform velocity gradient does not apply. The probability of orientations of particles larger than ℓ_K has been measured experimentally by Parsheh, Brown & Aidun (2005), Parsa *et al.* (2011), Zimmermann *et al.* (2011*a*,*b*) and Parsa *et al.* (2012) (in these experiments, the size of the particles ranged from a few ℓ_K to two orders of magnitude greater than ℓ_K). Jeffery's approach, however, remains applicable to turbulent flows if the particles are much smaller than ℓ_K, since below this scale viscosity smooths out the velocity fluctuations and hence the velocity gradient can be assumed as spatially uniform. The orientation dynamics of such small particles has been studied experimentally in atmospheric flows (Krushkal & Gallily 1988; Newsom & Bruce 1998) and in water-tunnel flows (Bernstein & Shapiro 1994), as well as numerically in turbulent channel flows (Zhang *et al.* 2001; Mortensen *et al.* 2008*a*,*b*), in isotropic turbulence (Shin & Koch 2005; Pumir & Wilkinson 2011; Parsa *et al.* 2012), and in chaotic velocity fields (Wilkinson, Bezuglyy & Mehlig 2009). Theoretical studies were mainly concerned with the derivation of model equations for the probability of orientations, in which turbulent fluctuations were described by an effective isotropic diffusion term (e.g. Krushkal & Gallily 1988; Olson & Kerekes 1998; Shin & Koch 2005). In contrast with the case of laminar flows, few analytical results seem to exist for the probability distribution of orientations in turbulent or chaotic flows. Turitsyn (2007) examined the tumbling motion of rod-like polymers in a random flow resulting from the superposition of a mean shear and of short-correlated isotropic fluctuations. Wilkinson & Kennard (2012) recently studied the alignment of rods with vorticity in an isotropic random flow.

Here, the probability density function (p.d.f.) of orientations is derived exactly for non-spherical particles transported by a homogeneous random flow with axisymmetric statistics. The particles are general bodies of revolution possessing fore–aft symmetry. The axisymmetry of the flow means that the velocity field is statistically invariant under arbitrary rotations of the axes about a given direction as well as under reflections in planes containing that direction or normal to it (Batchelor 1946; Chandrasekhar 1950). Axisymmetry is the simplest form of statistical anisotropy (Biferale & Procaccia 2005; Chang, Bewley & Bodenschatz 2012), and is found in rotating, stratified, or wind-tunnel turbulence (Lindborg 1995). Furthermore, the random flow is assumed to be Gaussian and to have zero correlation time. The assumption of temporal decorrelation is adequate when the correlation time of the flow is short compared to the characteristic time scale of material-line-element stretching. This assumption, albeit restrictive, allows a fully analytical solution of the problem.

The evolution equation for the orientation vector of a non-spherical rigid particle is introduced in § 2. Section 3 is devoted to the derivation of the convection–diffusion equation for the p.d.f. of the orientation angle. The steady-state orientation dynamics is studied in § 4. Some conclusions are drawn in § 5.

2. Orientation dynamics

The particles considered here are rigid bodies of revolution possessing fore–aft symmetry (although in the literature such particles are commonly referred to as 'axisymmetric', this terminology will be avoided here so as not to generate confusion; the term 'axisymmetry' will be reserved for the statistical invariance of the velocity field). The particles are of uniform composition and are suspended in a Newtonian fluid of the same density. Furthermore, the inertia of the particles as well as hydrodynamic particle–particle interactions are disregarded, and no externally imposed force or couple influences the dynamics. In particular, it is appropriate to disregard hydrodynamic interactions when the suspension is sufficiently dilute.

The undisturbed motion of the fluid is described by the velocity field $\boldsymbol{v}(\boldsymbol{x}, t)$. The size of the particles is assumed to be small compared to the typical length over which the velocity gradient $\nabla\boldsymbol{v} = (\partial_j v_i)_{1\leqslant i,j\leqslant 3}$ changes ($\partial_j \equiv \partial/\partial x_j$). For turbulent flows, this means that the particles are smaller than the Kolmogorov scale, and thus the Reynolds number at the scale of a particle is less than 1. Given their small size, the particles also experience Brownian collisions with the molecules of the fluid.

In a sufficiently dilute suspension, attention can be restricted to the dynamics of a single isolated particle. The configuration of a body of revolution is determined by the position of its centre of mass, $\boldsymbol{r}_c(t)$, and by the orientation of its axis of revolution, which is specified by a unit vector $\boldsymbol{N}(t)$ parallel to the axis itself. As the particles are subject to Brownian fluctuations, their dynamics is random even in a laminar flow. Consider first a deterministic velocity field or a given realization of a random velocity field. On the above assumptions, the centre of mass moves according to the following equation (e.g. Doi & Edwards 1986):

$$\dot{\boldsymbol{r}}_c(t) = \boldsymbol{v}(\boldsymbol{r}_c(t), t) + \sqrt{D_T}\,\boldsymbol{\zeta}(t), \tag{2.1}$$

where $D_T > 0$ is the translational diffusion coefficient and $\boldsymbol{\zeta}(t)$ is three-dimensional white noise, i.e. a Gaussian stochastic process with

$$\langle\boldsymbol{\zeta}(t)\rangle = 0 \quad \text{and} \quad \langle\zeta_i(t+\tau)\zeta_j(t)\rangle = \delta_{ij}\delta(\tau) \tag{2.2}$$

for all $t, \tau > 0$ and $i, j = 1, 2, 3$. The orientation vector satisfies the following stochastic differential equation (summation over repeated indexes is implied):

$$\dot{N}_i = \kappa_{ij}(t)N_j - \kappa_{pq}(t)\frac{N_p N_q}{|\boldsymbol{N}|^2}N_i + \sqrt{D_R}\,\varSigma_{ij}(\boldsymbol{N}) \circ \xi_j(t), \quad |\boldsymbol{N}(0)| = 1, \tag{2.3}$$

where

$$\boldsymbol{\kappa}(t) = \boldsymbol{\varOmega}(t) + \gamma\,\boldsymbol{E}(t) \tag{2.4}$$

with

$$\boldsymbol{\Omega}(t) = \frac{\mathbf{G}(t) - \mathbf{G}^{\mathrm{T}}(t)}{2}, \quad \mathbf{E}(t) = \frac{\mathbf{G}(t) + \mathbf{G}^{\mathrm{T}}(t)}{2}, \tag{2.5}$$

and $\mathbf{G}(t) = \nabla \boldsymbol{v}(\boldsymbol{r}_c(t), t)$. Thus, $\boldsymbol{\Omega}(t)$ and $\mathbf{E}(t)$ are the vorticity tensor and the rate-of-strain tensor evaluated at $\boldsymbol{r}_c(t)$, and $\boldsymbol{\kappa}(t)$ is an effective Lagrangian velocity gradient. The scalar constant γ describes the strain efficiency in orienting the particle and depends on the geometrical shape of the particle itself. For $|\gamma| < 1$, the evolution equation can be mapped into that of a spheroid with aspect ratio equal to $\sqrt{(1+\gamma)/(1-\gamma)}$ (Bretherton 1962). Prolate spheroids are obtained for $0 < \gamma < 1$, oblate spheroids for $-1 < \gamma < 0$. Special cases are: spheres ($\gamma = 0$), rigid dumbbells ($\gamma = 1$), rods ($\gamma = 1$), and disks ($\gamma = -1$). Moreover, Bretherton (1962) demonstrated the existence of geometrical shapes for which $|\gamma| > 1$; he provided an example consisting of two non-spherical bodies of revolution connected by a long and thin rigid rod (see figure 5 in Bretherton 1962), even though he observed that the required length and thickness may be unrealistic for a rigid particle.

In (2.3), the random vector $\boldsymbol{\xi}(t)$ is three-dimensional white noise and hence has the same properties as $\boldsymbol{\zeta}(t)$, but is statistically independent of it. The matrix $\boldsymbol{\Sigma}(\boldsymbol{n})$ has the following form:

$$\boldsymbol{\Sigma}(\boldsymbol{n}) = \mathbf{I} - \boldsymbol{n}\boldsymbol{n}/|\boldsymbol{n}|^2 \tag{2.6}$$

and $D_R > 0$ is the rotary diffusion coefficient. By using the Cauchy–Schwarz inequality, it is easy to check that $\boldsymbol{\Sigma}(\boldsymbol{n})$ is positive semi-definite. Finally, the symbol $\circ$ indicates that the stochastic term in (2.3) is understood in the Stratonovich sense.

Equation (2.3) is Jeffery's equation for the orientation vector of a body of revolution with the addition of a stochastic term modelling Brownian fluctuations. The stochastic term is chosen in such a way as to produce isotropic diffusion of $\boldsymbol{N}(t)$ on the unit sphere, so that $|\boldsymbol{N}(t)|$ is preserved in time (see appendix A for more details). It is worth remarking that a Brownian term of the same form has been used to model the turbulent fluctuations of the velocity gradient (Krushkal & Gallily 1988; Olson & Kerekes 1998; Shin & Koch 2005) or to describe particle–particle interactions both in semi-dilute suspensions (Doi & Edwards 1986) and in concentrated suspensions (Doi & Edwards 1978; Kuzuu & Doi 1980).

Equation (2.3) can be generalized to the case of a homogeneous axisymmetric random flow. The velocity field transporting the particle is Gaussian and has zero mean and correlation:

$$\langle v_i(\boldsymbol{x}+\boldsymbol{r}, t+\tau) v_j(\boldsymbol{x}, t)\rangle = Q_{ij}(\boldsymbol{r})\delta(\tau), \quad i, j = 1, 2, 3. \tag{2.7}$$

The form of the correlation guarantees that $\boldsymbol{v}(\boldsymbol{x}, t)$ is statistically homogeneous in space. Additionally, the velocity field is assumed to be incompressible ($\nabla \cdot \boldsymbol{v} = 0$) and statistically axisymmetric with respect to the direction specified by the unit vector $\boldsymbol{\lambda}$. The tensor $\mathbf{Q}(\boldsymbol{r})$ must then take the form:

$$Q_{ij}(\boldsymbol{r}) = Ar_ir_j + B\delta_{ij} + C\lambda_i\lambda_j + D(r_i\lambda_j + \lambda_i r_j), \tag{2.8}$$

where A, B and C are smooth functions of r^2 and $\mu^2 r^2$ with $r = |\boldsymbol{r}|$ and $\mu = (\boldsymbol{r} \cdot \boldsymbol{\lambda})/r$, while D is a function of r^2 and μr, being odd in μr (Batchelor 1946; Chandrasekhar 1950). As a consequence of incompressibility, the functions A, B, C, D are not independent and satisfy certain differential relations (Batchelor 1946; Chandrasekhar 1950). Moreover, the expansion of these functions for small values of r is (Batchelor 1946, p. 486):

$$A = a + O\left(r^2\right), \tag{2.9a}$$

$$B = B_0 - \tfrac{1}{2}\left(4a + d - b\mu^2\right) r^2 + O\left(r^4\right), \tag{2.9b}$$

$$C = C_0 - \tfrac{1}{2}(b + c + 5d - c\mu^2)r^2 + O\left(r^4\right), \tag{2.9c}$$

$$D = \mu r\left[d + O\left(r^2\right)\right], \tag{2.9d}$$

where a, b, c, d and B_0, C_0 are real constants. The velocity field defined above is an axisymmetric generalization of the isotropic random flow introduced by Kraichnan (1968) in the context of passive turbulent transport. The same axisymmetric velocity field was used by Shaqfeh & Koch (1992) to study polymer stretching in flows through random beds of fibres.

The velocity gradient is also Gaussian and zero-mean. The single-point two-time correlation of the components of $\nabla \boldsymbol{v}$ can be derived from (2.7) by using the statistical homogeneity of the velocity field:

$$\langle \partial_j v_i(\boldsymbol{x}, t+\tau)\partial_q v_p(\boldsymbol{x}, t)\rangle = \Gamma_{ijpq}\delta(\tau), \quad i, j, p, q = 1, 2, 3, \tag{2.10}$$

where

$$\Gamma_{ijpq} \equiv 2\int_0^\infty \langle \partial_j v_i(\boldsymbol{x}, t+\tau)\partial_q v_p(\boldsymbol{x}, t)\rangle \,\mathrm{d}\tau \tag{2.11}$$

is the time integral of the correlation of the components of $\nabla \boldsymbol{v}$ and satisfies:

$$\Gamma_{ijpq} = -\left.\frac{\partial^2 Q_{ip}}{\partial r_j \partial r_q}\right|_{r=0}. \tag{2.12}$$

Thus, $\nabla \boldsymbol{v}$ is a tensorial white noise with diffusion tensor $\boldsymbol{\Gamma}$. In particular, Γ_{ijij} (no summation) gives the amplitude of the fluctuations of $\partial_j v_i$. Substituting (2.8) and (2.9) into (2.12) yields (see (5.12) in Batchelor 1946):

$$\begin{aligned}\Gamma_{ijpq} = {} & (d+4a)\delta_{jq}\delta_{ip} - a(\delta_{pq}\delta_{ij} + \delta_{iq}\delta_{pj}) + (b+c+5d)\delta_{jq}\lambda_i\lambda_p - b\delta_{ip}\lambda_j\lambda_q \\ & - d[(\delta_{pq}\lambda_i + \delta_{iq}\lambda_p)\lambda_j + (\delta_{pj}\lambda_i + \delta_{ij}\lambda_p)\lambda_q] - c\lambda_i\lambda_p\lambda_j\lambda_q.\end{aligned} \tag{2.13}$$

For the sake of simplicity, $\boldsymbol{\lambda}$ is taken in the direction of the third axis, i.e. $\boldsymbol{\lambda} = (0, 0, 1)$. Then, the coefficients a, b, c, d are related to the time integrals of the correlations of the components of $\nabla \boldsymbol{v}$ (see (2.11)) as follows:

$$a = \tfrac{1}{2}\left(\Gamma_{1212} - \Gamma_{1111}\right), \quad d = \Gamma_{3333} - \Gamma_{1111}, \tag{2.14a}$$

$$b = \Gamma_{1212} - \Gamma_{1313}, \quad 2a + b + c + 4d = \Gamma_{3131} - \Gamma_{3333}. \tag{2.14b}$$

Note that the two-time correlation of the components of the vorticity $\boldsymbol{\omega} = \nabla \times \boldsymbol{v}$ can be expressed in terms of a, c and d (Batchelor 1946, p. 490):

$$\langle \omega_1(\boldsymbol{x}, t+\tau)\omega_1(\boldsymbol{x}, t)\rangle = \langle \omega_2(\boldsymbol{x}, t+\tau)\omega_2(\boldsymbol{x}, t)\rangle = (10a + c + 9d)\delta(\tau) \tag{2.15}$$

and

$$\langle \omega_3(\boldsymbol{x}, t+\tau)\omega_3(\boldsymbol{x}, t)\rangle = (10a + 2d)\delta(\tau). \tag{2.16}$$

Moreover, the coefficients a, b, c, d in (2.13) are not free; they are constrained by the following inequalities (see appendix C):

$$\left.\begin{array}{ll} a+d>0 & \text{if } a \geqslant 0, \\ 5a+d>0 & \text{if } a<0, \end{array}\right\} \tag{2.17a}$$

$$4a-b+d>0, \quad 4a+b+c+6d>0, \tag{2.17b}$$

$$15a^2-b^2-(b-d)(c+5d)+2a(2c+13d)>0. \tag{2.17c}$$

If $a>0$ and $b=c=d=0$, then Γ_{ijpq} gives the single-point correlation of the gradient of an isotropic velocity field (Robertson 1940). Thus, a determines the intensity of the isotropic part of $\nabla \boldsymbol{v}$, while b, c, d control the statistical anisotropy of the flow.

Note that an alternative representation of the tensor $\boldsymbol{Q}$ was proposed by Lindborg (1995). The relation between (2.13) and Lindborg's representation is given in appendix B.

The orientation dynamics of the particle depends on the velocity gradient evaluated at $\boldsymbol{r}_c(t)$, which was denoted as $\boldsymbol{G}(t)$ in § 2. By virtue of the δ-correlation in time and the statistical homogeneity of the flow, $\boldsymbol{G}(t)$ has the same temporal statistics as $\nabla \boldsymbol{v}(\boldsymbol{x}, t)$ for any given $\boldsymbol{x}$ (Falkovich, Gawędzki & Vergassola 2001) (the presence of white noise in (2.1) does not modify the statistics of $\boldsymbol{G}(t)$). The components of $\boldsymbol{\kappa}(t)$, defined in (2.4), are a linear combination of the components of $\boldsymbol{G}(t)$, and consequently $\boldsymbol{\kappa}(t)$ is a Gaussian process with zero mean and correlation:

$$\langle \kappa_{ij}(t+\tau)\kappa_{pq}(t)\rangle = K_{ijpq}\delta(\tau) \tag{2.18}$$

with

$$K_{ijpq} = \tfrac{1}{4}[\Gamma_{ijpq}-\Gamma_{ijqp}-\Gamma_{jipq}+\Gamma_{jiqp}+2\gamma(\Gamma_{ijpq}-\Gamma_{jiqp}) \\ +\gamma^2(\Gamma_{ijpq}+\Gamma_{ijqp}+\Gamma_{jipq}+\Gamma_{jiqp})]. \tag{2.19}$$

The form of K_{ijpq} can be derived by substituting (2.4) and (2.5) into the left-hand side of (2.18) and by using (2.10). The tensor K_{ijpq} is positive semi-definite, for it is the covariance of a second-order Gaussian tensor. Thus, in the evolution equation for the orientation vector, $\boldsymbol{\kappa}(t)$ plays the role of a multiplicative tensorial white noise. As $\boldsymbol{\kappa}(t)$ can be thought of as an approximation of a real noise process in the limit of zero correlation time, the corresponding terms in (2.3) must be interpreted in the Stratonovich sense (e.g. Kloeden & Platen 1992, p. 227). The stochastic differential equation for $\boldsymbol{N}(t)$ can then be rewritten as follows:

$$\dot{N}_i = M_{ipq}(\boldsymbol{N}) \circ \kappa_{pq}(t) + \sqrt{D_R}\, \Sigma_{ij}(\boldsymbol{N}) \circ \xi_j(t), \tag{2.20}$$

where $M_{ipq}(\boldsymbol{n}) = (\delta_{ip}\delta_{jk}-\delta_{ik}\delta_{jp})n_j n_k n_q/|\boldsymbol{n}|^2$ and the initial condition $\boldsymbol{N}(0)$ is such that $|\boldsymbol{N}(0)|=1$.

3. Convection–diffusion equation for the probability density function of the orientation angle

As $\boldsymbol{v}(\boldsymbol{x}, t)$ is statistically invariant under spatial translations, the p.d.f. of $\boldsymbol{N}(t)$ taking the value $\boldsymbol{n}=(n_1, n_2, n_3)$ at time t is independent of $\boldsymbol{r}_c$ and is thus denoted by $f(\boldsymbol{n}; t)$. The Itô equation equivalent to (2.20) is

$$\dot{N}_i = \beta_i(\boldsymbol{N}) + M_{ipq}(\boldsymbol{N})\kappa_{pq}(t) + \sqrt{D_R}\, \Sigma_{ij}(\boldsymbol{N})\xi_j(t) \tag{3.1}$$

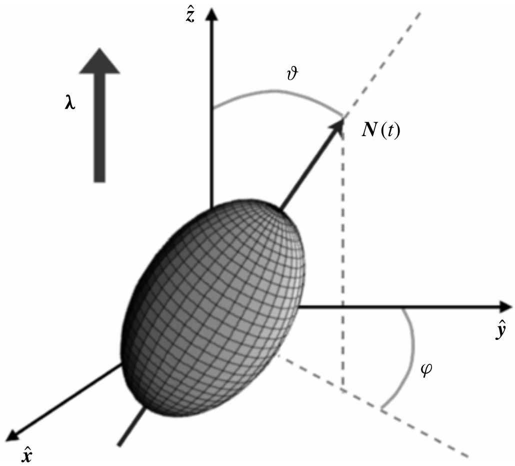

FIGURE 1. Orientation of a non-spherical particle.

with

$$\beta_i(\boldsymbol{n}) = \frac{1}{2} K_{mnpq} M_{jpq}(\boldsymbol{n}) \frac{\partial}{\partial n_j} M_{imn}(\boldsymbol{n}) + \frac{D_R}{2} \Sigma_{jk}(\boldsymbol{n}) \frac{\partial}{\partial n_j} \Sigma_{ik}(\boldsymbol{n}). \tag{3.2}$$

Consequently, $f(\boldsymbol{n}; t)$ satisfies the Fokker–Planck equation:

$$\frac{\partial f}{\partial t} = -\frac{\partial}{\partial n_i} [\beta_i(\boldsymbol{n}) f] + \frac{1}{2} \frac{\partial^2}{\partial n_i \partial n_j} [\alpha_{ij}(\boldsymbol{n}) f] \tag{3.3}$$

with

$$\begin{aligned} \alpha_{ij}(\boldsymbol{n}) &= K_{mnpq} M_{imn}(\boldsymbol{n}) M_{jpq}(\boldsymbol{n}) + D_R \Sigma_{ik}(\boldsymbol{n}) \Sigma_{jk}(\boldsymbol{n}) \\ &= K_{mnpq} M_{imn}(\boldsymbol{n}) M_{jpq}(\boldsymbol{n}) + D_R \Sigma_{ij}(\boldsymbol{n}), \end{aligned} \tag{3.4}$$

where the last equality follows from (A 3) in appendix A. Equations (3.1) and (3.3) can be derived from (2.20) by using the formal rules $\kappa_{ij}(t)\,\mathrm{d}t = O(\sqrt{\mathrm{d}t})$ and $\kappa_{ij}(t)\,\mathrm{d}t\,\kappa_{pq}(t)\,\mathrm{d}t = K_{ijpq}\,\mathrm{d}t$ and by proceeding as in the case of a vectorial white noise (see Gardiner 1983 and the appendix in Falkovich *et al.* 2001). The diffusion tensor $\boldsymbol{\alpha}$ is positive semi-definite as a consequence of the positive semi-definiteness of $\boldsymbol{K}$ and $\boldsymbol{\Sigma}$.

To study the orientation dynamics of a non-spherical particle, it is convenient to move from Cartesian coordinates (n_1, n_2, n_3) to spherical coordinates (n, ϑ, φ) according to the usual transformations:

$$n = \sqrt{n_1^2 + n_2^2 + n_3^2}, \quad \vartheta = \arctan\left(\sqrt{n_1^2 + n_2^2}/n_3\right), \quad \varphi = \arctan(n_2/n_1) \tag{3.5}$$

with $0 \leqslant n$, $0 \leqslant \vartheta \leqslant \pi$, and $0 \leqslant \varphi < 2\pi$ (figure 1). On account of the fixed length of the orientation vector, the probability density function of orientations must take the form $f(n, \vartheta, \varphi; t) = \psi(\vartheta, \varphi; t)\delta(n - L)$ with $L = 1$. Thus, $\psi(\vartheta, \varphi; t) \sin\vartheta\,\mathrm{d}\vartheta\,\mathrm{d}\varphi$ is the probability of the particle being oriented at time t within an elementary solid angle $\sin\vartheta\, \mathrm{d}\vartheta\, \mathrm{d}\varphi$ of (ϑ, φ). In addition, the following normalization holds:

$$\int_0^{\pi}\int_0^{2\pi} \psi(\vartheta, \varphi; t) \sin\vartheta\, \mathrm{d}\vartheta\, \mathrm{d}\varphi = 1 \quad \forall\, t \geqslant 0. \tag{3.6}$$

The function $\psi(\vartheta, \varphi; t)$ satisfies the convection–diffusion equation (see appendix D for the derivation):

$$\frac{\partial\psi}{\partial t} = \frac{1}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\left\{\sin\vartheta\left[-\mathscr{W}_\vartheta(\vartheta)\psi + \frac{1}{2}\mathscr{D}_{\vartheta\vartheta}(\vartheta)\frac{\partial\psi}{\partial\vartheta}\right]\right\} + \frac{\mathscr{D}_{\varphi\varphi}(\vartheta)}{2\sin^2(\vartheta)}\frac{\partial^2\psi}{\partial\varphi^2}, \tag{3.7}$$

where

$$\mathscr{W}_\vartheta(\vartheta) = \tfrac{1}{4}\sin(2\vartheta)[2\nu_2 - 3\nu_4 - 3\nu_3\sin^2(\vartheta)] \tag{3.8}$$

and

$$\mathscr{D}_{\vartheta\vartheta}(\vartheta) = 2\nu_1 + \nu_4\sin^2(\vartheta) + \nu_3\sin^4(\vartheta), \tag{3.9a}$$

$$\mathscr{D}_{\varphi\varphi}(\vartheta) = 2\nu_1 + \nu_5\sin^2(\vartheta) \tag{3.9b}$$

with

$$\nu_1 = \tfrac{1}{8}\left[2(5+3\gamma^2)a - 4\gamma b + (1-\gamma)^2 c + (9 - 10\gamma + 5\gamma^2)d + 4D_R'\right], \tag{3.10a}$$

$$\nu_2 = \tfrac{3}{4}\gamma\,[2b + (1-\gamma)c + (5-\gamma)d], \tag{3.10b}$$

$$\nu_3 = c\gamma^2, \tag{3.10c}$$

$$\nu_4 = \gamma[2b + (1-\gamma)c + 5d], \tag{3.10d}$$

$$\nu_5 = \tfrac{1}{4}\left[4b\gamma - (1-\gamma)^2 c - (1-\gamma)(7-3\gamma)d\right], \tag{3.10e}$$

and $D_R' = D_R/L^2$ with $L = 1$ (the numerical values of D_R' and D_R coincide, but their physical dimensions are different).

Inequalities (2.17) guarantee that $\mathscr{D}_{\vartheta\vartheta}(\vartheta)$ and $\mathscr{D}_{\varphi\varphi}(\vartheta)$ are strictly positive (appendix C). The contribution to (3.7) due to the isotropic part of $\boldsymbol{\Gamma}$ is a diffusion term with diffusion coefficient proportional to a; this contribution is of the same form as that coming from $\boldsymbol{\xi}(t)$. The isotropic component of the flow and the Brownian fluctuations therefore have the same effect on the orientation statistics of the particle. Note that $\mathscr{W}_\vartheta(\vartheta)$ plays the role of an apparent drift velocity; its explicit expression is:

$$\mathscr{W}_\vartheta(\vartheta) = \tfrac{3}{8}\gamma\,\sin(2\vartheta)[c\gamma\cos(2\vartheta) - 2b - c - (5+\gamma)d]. \tag{3.11}$$

Also observe that $\mathscr{W}_\vartheta(\vartheta)$ does not depend on a, and hence the convective term in (3.7) vanishes for an isotropic flow.

As ϑ and φ are angular variables, the boundary conditions for $\psi(\vartheta, \varphi; t)$ are periodic:

$$\psi(\vartheta, \varphi; t) = \psi(\vartheta + 2\pi, \varphi; t) \quad \text{and} \quad \psi(\vartheta, \varphi; t) = \psi(\vartheta, \varphi + 2\pi; t) \tag{3.12}$$

for all ϑ, φ, and t. The long-time properties of $\psi(\vartheta, \varphi; t)$ can be deduced from (3.7). If the partial derivative with respect to time is dropped, then (3.7) is invariant under the transformations $\vartheta \leftrightarrow 2\pi - \vartheta$ (reflections with respect to planes containing $\boldsymbol{\lambda}$) and $\vartheta \leftrightarrow \pi - \vartheta$ (reflections with respect to planes orthogonal to $\boldsymbol{\lambda}$). Furthermore, the coefficients $\mathscr{W}_\vartheta$, $\mathscr{D}_{\vartheta\vartheta}$ and $\mathscr{D}_{\varphi\varphi}$ do not depend on φ (invariance under rotations about $\boldsymbol{\lambda}$). These properties of (3.7) are a natural consequence of the statistical axisymmetry of the velocity field and translate into analogous properties of the stationary p.d.f. of orientations.

The invariance of (3.7) under rotations about $\boldsymbol{\lambda}$ can be used to derive a one-dimensional convection–diffusion equation for the marginal p.d.f.: $\widehat{\psi}(\vartheta; t) \equiv \int_0^{2\pi} \psi(\vartheta, \varphi; t)\, \mathrm{d}\varphi$. Indeed, integrating (3.7) with respect to φ from 0 to 2π and making use of (3.12) yields:

$$\frac{\partial \widehat{\psi}}{\partial t} = \frac{1}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\left\{\sin\vartheta\left[-\mathscr{W}_\vartheta(\vartheta)\widehat{\psi} + \frac{1}{2}\mathscr{D}_{\vartheta\vartheta}(\vartheta)\frac{\partial\widehat{\psi}}{\partial\vartheta}\right]\right\}. \tag{3.13}$$

The solution of the above equation must be normalized and periodic: $\widehat{\psi}(\vartheta; t) = \widehat{\psi}(\vartheta + 2\pi; t)$ for all ϑ, t. A direct consequence of (3.13) is that, along the trajectory of the particle, the time evolution of $\vartheta(t)$ is decoupled from that of $\varphi(t)$ and is described by the Itô stochastic ordinary differential equation (see appendix D):

$$\dot{\vartheta}(t) = \mathscr{B}_\vartheta(\vartheta(t)) + \sqrt{\mathscr{D}_{\vartheta\vartheta}(\vartheta(t))}\,\eta(t), \quad 0 \leqslant \vartheta(t) \leqslant \pi, \tag{3.14}$$

where $\eta(t)$ is white noise and

$$\mathscr{B}_\vartheta(\vartheta) = \mathscr{W}_\vartheta(\vartheta) + \frac{1}{2\sin\vartheta}\frac{\partial}{\partial\vartheta}[\mathscr{D}_{\vartheta\vartheta}(\vartheta)\sin\vartheta]. \tag{3.15}$$

4. Stationary statistics of the orientation angle

It was argued in § 3 that the stationary solution of (3.7), ψ_{st}, does not depend on φ. Therefore, ψ_{st} solves the equation:

$$\frac{\mathrm{d}}{\mathrm{d}\vartheta}\left\{\sin\vartheta\left[-\mathscr{W}_\vartheta(\vartheta)\psi_{st} + \frac{1}{2}\mathscr{D}_{\vartheta\vartheta}(\vartheta)\frac{\mathrm{d}\psi_{st}}{\mathrm{d}\vartheta}\right]\right\} = 0. \tag{4.1}$$

Thanks to the periodic boundary conditions (3.12), $\psi_{st}(\vartheta)$ can be calculated by equating the term in square brackets in (4.1) to zero (see appendix D), whence:

$$\psi_{st}(\vartheta) = \mathscr{N}\exp\left[2\int_{\vartheta_0}^{\vartheta}\frac{\mathscr{W}_\vartheta(s)}{\mathscr{D}_{\vartheta\vartheta}(s)}\,\mathrm{d}s\right], \tag{4.2}$$

where $0 \leqslant \vartheta_0 \leqslant \pi$ and $\mathscr{N}$ is a positive normalization constant such that

$$2\pi\int_0^\pi \psi_{st}(\vartheta)\sin\vartheta\,\mathrm{d}\vartheta = 1. \tag{4.3}$$

In (4.2), the choice of ϑ_0 is in fact unimportant, since it only modifies the value of $\mathscr{N}$.

Only the case $\nu_3 \neq 0$ is considered here; the case $\nu_3 = 0$ is examined in appendix E, even though no new physical regimes emerge when ν_3 is equal to zero. For $\nu_3 \neq 0$, the integral on the right-hand side of (4.2) can be calculated explicitly (see appendix D). The final result is:

$$\psi_{st}(\vartheta) = \frac{\mathscr{N}\chi(\vartheta)}{[\mathscr{D}_{\vartheta\vartheta}(\vartheta)]^{3/4}}, \tag{4.4}$$

where

$$\chi(\vartheta)=\begin{cases}\exp\left\{\dfrac{2}{\sqrt{\Delta}}\left(\nu_2-\dfrac{3}{4}\nu_4\right)\arctan\left[\dfrac{\nu_4+2\nu_3\sin^2(\vartheta)}{\sqrt{\Delta}}\right]\right\} & (\Delta>0)\\ \exp\left\{\left(\dfrac{3}{4}\nu_4-\nu_2\right)\dfrac{2}{\nu_4+2\nu_3\sin^2(\vartheta)}\right\} & (\Delta=0)\\ \left|\dfrac{\sqrt{-\Delta}+\nu_4+2\nu_3\sin^2(\vartheta)}{\sqrt{-\Delta}-\nu_4-2\nu_3\sin^2(\vartheta)}\right|^{(3\nu_4/4-\nu_2)/\sqrt{-\Delta}} & (\Delta<0)\end{cases} \tag{4.5}$$

with $\Delta=8\nu_1\nu_3-\nu_4^2$. It is shown in appendix C that $\psi_{st}(\vartheta)$ is bounded for all values of a, b, c, d and γ. The stationary p.d.f. of orientations satisfies $\psi_{st}(2\pi-\vartheta)=\psi_{st}(\vartheta)$ and $\psi_{st}(\pi-\vartheta)=\psi_{st}(\vartheta)$. These properties are a consequence of the statistical symmetries of the carrier flow, as was noted after (3.12).

Since Δ can be written as $\Delta=a^2F(\gamma, b/a, c/a, d/a, D'_R/a)$, $\psi_{st}(\vartheta)$ only depends on the ratios b/a, c/a, d/a, D'_R/a (and on γ). The same conclusion could have been reached by rescaling t by a^{-1} in (3.7).

For a spherical particle (i.e. $\gamma=0$), ν_2 and ν_4 vanish. Consequently, $\chi(\vartheta)=1$, $\mathscr{D}_{\vartheta\vartheta}(\vartheta)=\text{const.}$, and hence $\psi_{st}(\vartheta)=(4\pi)^{-1}$, in accordance with the fact that all orientations are equally probable for a sphere. Similarly, if a or D'_R is much greater than b, c, d, i.e. if the isotropic component of the velocity field or the Brownian fluctuations prevail over the anisotropic component of the flow, then $\nu_1\gg\nu_i$, $i=2,\dots,5$ and hence $\psi_{st}(\vartheta)$ weakly depends on ϑ regardless of the shape of the particle. In the following, therefore, γ is assumed to be non-zero and b, c, d are of the same order of magnitude as a and D'_R or greater.

The behaviour of $\psi_{st}(\vartheta)$ can be deduced from that of its first derivative. By using (3.11), (4.1) and (4.2), it is easy to see that, for all values of Δ,

$$\frac{\mathrm{d}}{\mathrm{d}\vartheta}\psi_{st}(\vartheta)=\mathscr{W}_\vartheta(\vartheta)h(\vartheta)=\frac{3}{8}\gamma\sin(2\vartheta)[c\gamma\cos(2\vartheta)-\sigma]h(\vartheta), \tag{4.6}$$

where $h(\vartheta)\equiv 2\psi_{st}(\vartheta)/\mathscr{D}_{\vartheta\vartheta}(\vartheta)$ is strictly positive for all $0\leqslant\vartheta\leqslant\pi$ (see (C 7*a*) and (C 8*a*)) and

$$\sigma\equiv 2b+c+(5+\gamma)d. \tag{4.7}$$

Thus, the extrema of ψ_{st} are the zeros of the function $\mathscr{W}_\vartheta$ determining the apparent drift velocity in the convection–diffusion equation (3.7). Note that σ can be rewritten as $\sigma=(\Gamma_{3131}-\Gamma_{1313})+\gamma(\Gamma_{3333}-\Gamma_{1111})$, where the term in the first parentheses describes the anisotropy of the vorticity tensor and that in the second is related to the anisotropy of the strain tensor, with the ratio of the two terms being given by γ.

Four regimes are identified depending on the properties of the axisymmetric flow and on the geometrical shape of the particle.

(i) *Rotation about the axis of symmetry of the flow*. For the following values of the parameters:

$$|\sigma|>|c\gamma| \quad \text{and} \quad \gamma\sigma<0, \tag{4.8}$$

the function ψ_{st} only has three extrema at $\vartheta=0,\pi/2,\pi$ (indeed the equation $c\gamma\cos(2\vartheta)=\sigma$ has no solution). More precisely, $\psi_{st}(\vartheta)$ has a maximum at $\pi/2$ and two minima at 0 and π (figure 2). Thus, in this regime a non-spherical particles rotates about the direction $\boldsymbol{\lambda}$; the level of alignment with the plane

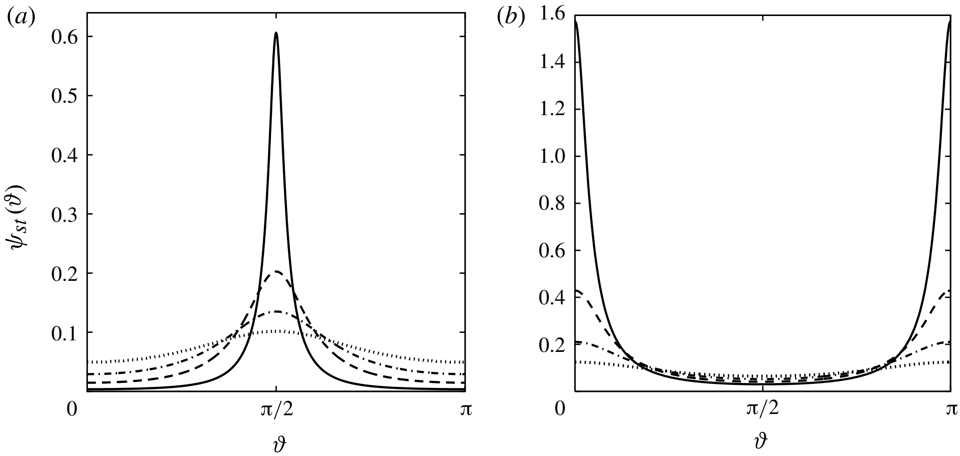

FIGURE 2. Stationary p.d.f. of the orientation angle ϑ. The normalization coefficient $\mathscr{N}$ has been computed numerically according to (4.3). (*a*) Rotation about $\boldsymbol{\lambda}$: $b/a = 4.8$, $c/a = 55$, $d/a = 0.85$, $D'_R/a = 10^{-2}$ and $\gamma = -1$ (solid line), $\gamma = -0.75$ (dashed line), $\gamma = -0.5$ (dot-dashed line), $\gamma = -0.25$ (dotted line). (*b*) Tumbling motion between $\boldsymbol{\lambda}$ and $-\boldsymbol{\lambda}$: $b/a = 4.75$, $c/a = 10$, $d/a = 0.9$, $D'_R/a = 10^{-2}$ and $\gamma = 1$ (solid line), $\gamma = 0.75$ (dashed line), $\gamma = 0.5$ (dot-dashed line), $\gamma = 0.25$ (dotted line).

perpendicular to $\boldsymbol{\lambda}$ decreases as the degree of anisotropy of the flow vanishes or the shape of the particle approaches a spherical one (figure 2).

(ii) *Tumbling motion*. In the following regime:

$$|\sigma| > |c\gamma| \quad \text{and} \quad \gamma\sigma > 0, \tag{4.9}$$

ψ_{st} has three extrema: a minimum at $\pi/2$ and two maxima at 0 and at π (figure 2). The particle tumbles between the direction parallel to $\boldsymbol{\lambda}$ and that antiparallel to $\boldsymbol{\lambda}$. The probability of the orientation angle ϑ being in the neighbourhood of 0 or π depends on the anisotropy degree of the flow and on the shape of the particle (figure 2).

(iii) *Preferential alignment with a direction oblique to the axis of symmetry of the flow*. If

$$|\sigma| < |c\gamma| \quad \text{and} \quad c > 0, \tag{4.10}$$

then ψ_{st} has three minima at $0, \pi/2, \pi$ and two maxima at $\vartheta_\star$ and $\pi - \vartheta_\star$, where $0 < \vartheta_\star < \pi/2$ is such that

$$\sin\vartheta_\star = \sqrt{\frac{1}{2}\left(1 - \frac{\sigma}{c\gamma}\right)}. \tag{4.11}$$

The particle, therefore, spends most of the time at an angle $\vartheta_\star$ (or $\pi - \vartheta_\star$) with respect to $\boldsymbol{\lambda}$ (figure 3).

(iv) *Combination of rotation and tumbling*. For

$$|\sigma| < |c\gamma| \quad \text{and} \quad c < 0, \tag{4.12}$$

the function ψ_{st} has three maxima at $0, \pi/2, \pi$ and two minima at $\vartheta_\star$ and $\pi - \vartheta_\star$ with $\vartheta_\star$ defined in (4.11) (see figure 3). In this regime, the particle preferentially

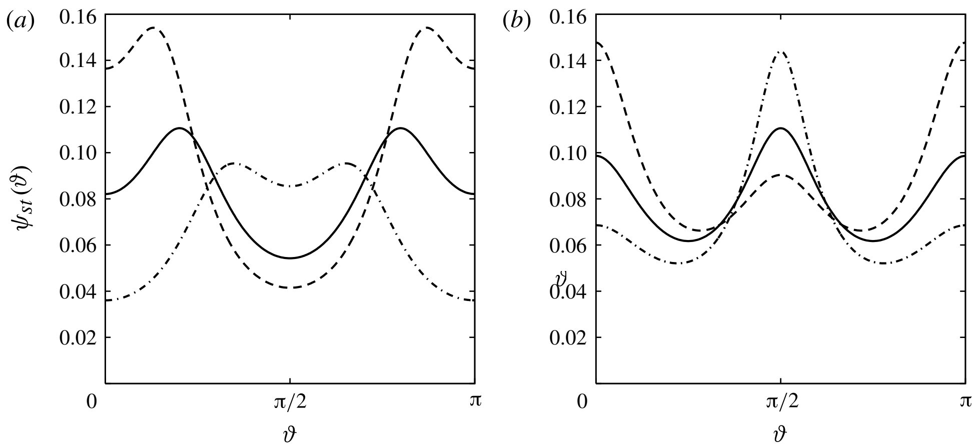

FIGURE 3. Stationary p.d.f. of the orientation angle ϑ. The p.d.f. has been normalized numerically according to (4.3). (*a*) Preferential alignment with a direction oblique to $\boldsymbol{\lambda}$: $\gamma = 1$, $D'_R/a = 10^{-2}$, $b/a = -6$, $d/a = -0.9$ and $c/a = 55$ (dashed line), $c/a = 25$ (solid line), $c/a = 11$ (dot-dashed line). (*b*) Combination of rotation and tumbling: $\gamma = 1$, $D'_R/a = 10^{-2}$, $c/a = -10$, $d/a = 1$ and $b/a = 1.1$ (dot-dashed line), $b/a = 2$ (solid line), $b/a = 3$ (dashed line).

lies either in the plane perpendicular to $\boldsymbol{\lambda}$, in the direction parallel to $\boldsymbol{\lambda}$, or in the direction antiparallel to it.

Note that whereas sufficiently elongated or flattened spheroids can be strongly aligned in regimes (i) and (ii), the ability of the flow to orient particles is weaker in regimes (iii) and (iv) (figure 3). In these regimes, strong alignment can be obtained only for $|\gamma| > 1$ (figure 4). However, Bretherton (1962) observed that particles with $|\gamma| > 1$ may be unrealistic, albeit conceivable from a purely geometrical point of view.

The above classification holds for all $\gamma \neq 0$ and for all a, b, c, d satisfying inequalities (2.17) (with $c \neq 0$). Naturally, the coefficient a controlling the intensity of the isotropic component of the flow does not play any role in the classification; the dynamical regime is selected by b, c, d, and by the shape coefficient γ. The way the anisotropy parameters and the shape coefficient enter conditions (4.8), (4.9), (4.10) and (4.12) is rather involved. There are cases, however, which can be easily interpreted from a physical point of view.

Consider for example the case $b = d = 0$, $c \neq 0$, $a > 0$, and $|\gamma| \leqslant 1$. Then, the condition $|\sigma| > |c\gamma|$ is trivially satisfied and the sign of γc determines whether a particle rotates around $\boldsymbol{\lambda}$ ($\gamma c < 0$) or tumbles ($\gamma c > 0$). This behaviour can be easily understood by noting that if $c > 0$, the flow differs from an isotropic flow only in that $\Gamma_{3131} - \Gamma_{3333}$ is greater in value, i.e. the difference between the amplitude of the fluctuations of $\partial_1 v_3$ and that of the fluctuations of $\partial_3 v_3$ is greater than it would be in the isotropic case (recall that v_3 is the component of the velocity in the direction of $\boldsymbol{\lambda}$, while v_1 and v_2 are the components orthogonal to $\boldsymbol{\lambda}$). Therefore, the orientation vector of an elongated particle ($0 < \gamma \leqslant 1$ and hence $\gamma c > 0$) is attracted towards the direction parallel to $\boldsymbol{\lambda}$ or towards that antiparallel to $\boldsymbol{\lambda}$ and thus performs a tumbling motion between the two directions; the orientation vector of a flattened particle ($-1 \leqslant \gamma < 0$ and hence $\gamma c < 0$) is attracted to the plane orthogonal to $\boldsymbol{\lambda}$ and thus predominantly rotates in that plane. The situation is reversed for $c < 0$, since

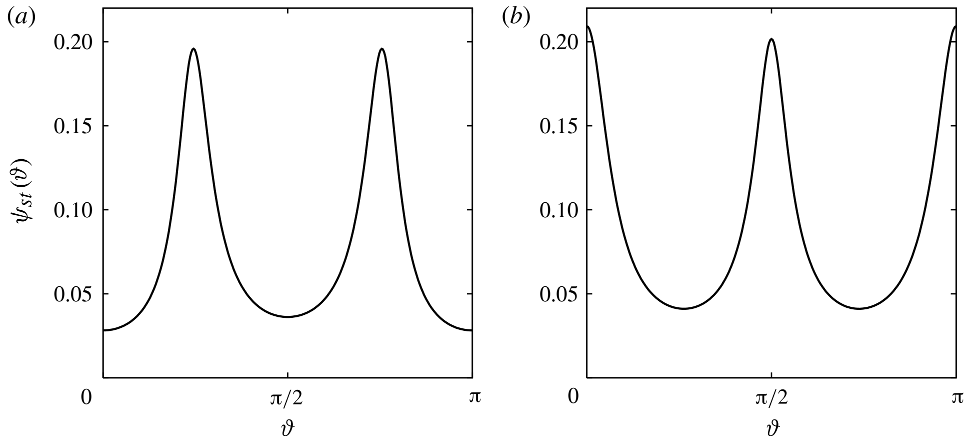

FIGURE 4. Stationary p.d.f. of the orientation angle ϑ for $|\gamma| > 1$. (*a*) Preferential alignment with a direction oblique to $\boldsymbol{\lambda}$: $\gamma = 5$, $D'_R/a = 10^{-2}$, $b/a = -6$, $c/a = 25$, $d/a = -0.9$. (*b*) Combination of rotation and tumbling: $\gamma = 5$, $D'_R/a = 10^{-2}$, $b/a = 2$, $c/a = -10$, $d/a = 1$.

in this case the difference between the amplitude of the fluctuations of $\partial_1 v_3$ and the amplitude of the fluctuations of $\partial_3 v_3$ is less than in an isotropic flow. Other examples can be examined in a similar way, but the analysis may be less intuitive when b, c, d are all non-zero.

For $d = 0$ (i.e. when the fluctuations of $\partial_1 v_1$, $\partial_2 v_2$, and $\partial_3 v_3$ have the same amplitude), the study of the dependence of $\psi_{st}(\vartheta)$ upon the shape of the particles can be developed further. This case is examined below.

4.1. *The case $d = 0$*

If $d = 0$, then $\nu_2 = 3\nu_4/4$ and the form of $\psi_{st}(\vartheta)$ simplifies considerably (see (4.4) and (4.5)):

$$\psi_{st}(\vartheta) = \mathscr{N} \, [\mathscr{D}_{\vartheta\vartheta}(\vartheta)]^{-3/4} \, . \tag{4.13}$$

Furthermore, σ does not depend on γ; the conditions $|\sigma| \gtrless |c\gamma|$ are thus independent of the sign of γ, i.e. of whether a particle is elongated or flattened. This property of the $d = 0$ case has the following implications (which once again hold for any $\gamma \neq 0$).

(*a*) If for a given σ the particles with shape coefficient $\widehat{\gamma}$ are in regime (i) (i.e. they rotate in the plane orthogonal to $\boldsymbol{\lambda}$), then for the same σ the particles with shape coefficient $-\widehat{\gamma}$ are in regime (ii) (i.e. they tumble). Similarly, if the particles with shape coefficient $\widehat{\gamma}$ are in regime (ii), then the particles with shape coefficient $-\widehat{\gamma}$ are in regime (i). Thus, for $d = 0$, elongated and flattened particles behave in an opposite way if $|\sigma| > |c\gamma|$.

(*b*) If for a given flow the particles with shape coefficient $\widehat{\gamma}$ are in regime (i) (i.e. they rotate about $\boldsymbol{\lambda}$), then in such a flow all particles with $|\gamma| < |\widehat{\gamma}|$ and $\mathrm{sgn}(\gamma) = \mathrm{sgn}(\widehat{\gamma})$ are in the same regime. In other words, if a prolate spheroid rotates about $\boldsymbol{\lambda}$, then all prolate spheroids having smaller aspect ratio rotate around $\boldsymbol{\lambda}$. Oblate spheroids behave analogously. Moreover, if the particles with shape coefficient $\widehat{\gamma}$ are in regime (ii) (i.e. they tumble), then all particles with $|\gamma| < |\widehat{\gamma}|$ and $\mathrm{sgn}(\gamma) = \mathrm{sgn}(\widehat{\gamma})$ also tumble.

(*c*) In contrast with regimes (i) and (ii), regimes (iii) and (iv) are independent of the sign of γ, i.e. of whether a particle is elongated or flattened; nevertheless, $\vartheta_\star$ changes if the sign of γ changes.

(*d*) If the particles with shape coefficient $\widehat{\gamma}$ are in regime (iii) (respectively in regime (iv)), then all particles with $|\gamma| > |\widehat{\gamma}|$ are in regime (iii) (respectively in regime (iv)). This means, for instance, that if a prolate spheroid preferentially orients itself in a direction oblique $\boldsymbol{\lambda}$, the same holds for all prolate spheroids with greater aspect ratio.

The above properties hold for $d = 0$, but in general do not extend to the case $d \neq 0$. For example, if $b/a = -4.4$, $c/a = 0.5$, $d/a = 1.5$, then particles tumble for $1 \geqslant \gamma > 0.8$, they have a preferential orientation for $0.8 > \gamma > 0.4$, they rotate for $0.4 > \gamma > 0$, and they tumble again for $0 > \gamma \geqslant -1$.

5. Conclusions

Axisymmetric turbulence arises as one of the simplest frameworks in which to study the orientation dynamics of non-spherical particles. On the assumptions of Gaussianity and short correlation in time, it has been shown analytically that the dynamics of a non-spherical particle immersed in a random axisymmetric flow exhibits four regimes: rotation around the axis of symmetry of the flow, tumbling, a combination of rotation and tumbling, and preferential alignment with a direction oblique to the axis of symmetry of the flow. The regime is selected by the form of the anisotropic component of the flow and by the geometrical shape of the particle. If the flow is weakly anisotropic or if the particle is almost spherical, the mathematical description of $\psi_{st}(\vartheta)$ in terms of minima and maxima remains formally valid, but the above physical classification loses its meaning, since $\psi_{st}(\vartheta)$ does not deviate appreciably from the uniform distribution.

The orientational dynamics of a rigid rod in the flow resulting from the superposition of a uniform shear and of an isotropic short-correlated random noise is characterized by a tumbling motion in the plane of the shear (Puliafito & Turistyn 2005; Turitsyn 2007). Such tumbling motion, however, differs from that experienced by non-spherical particles in the axisymmetric random flow (see regime (ii) in § 4). In the presence of a strong mean shear, the tumbling dynamics of a rod consists of aperiodic transitions between two unstable states: the one aligned with the direction of the shear and that anti-aligned with it. When a fluctuation takes the rod away from the aligned or anti-aligned state and moves it into the unstable region of the flow the mean shear makes the rod flip. By contrast, in the axisymmetric case, large deviations of the orientation of the particle from the axis of symmetry of the flow do not necessarily result into sudden flips of the particle (figure 5). Simply, the orientation vector of the particle fluctuates randomly, but the orientations aligned and anti-aligned with the axis of symmetry of the flow are much more probable than the other orientations. Thus, there can be excursions of $\vartheta(t)$ from $\vartheta \approx 0$ to $\vartheta \approx \pi/2$ followed by a return to $\vartheta \approx 0$ (figure 5). The probability of such events would be extremely small in the presence of a strong mean shear.

The orientational dynamics in the laminar uniform shear (Jeffery 1922; Bretherton 1962) and that in the axisymmetric random flow depend on the geometrical shape of particles in a different way. In the former case, the motion of a rod or of a disk represents a degenerate case of the dynamics (Jeffery 1922); furthermore, the orientational dynamics is qualitatively different for $|\gamma| < 1$ and $|\gamma| > 1$

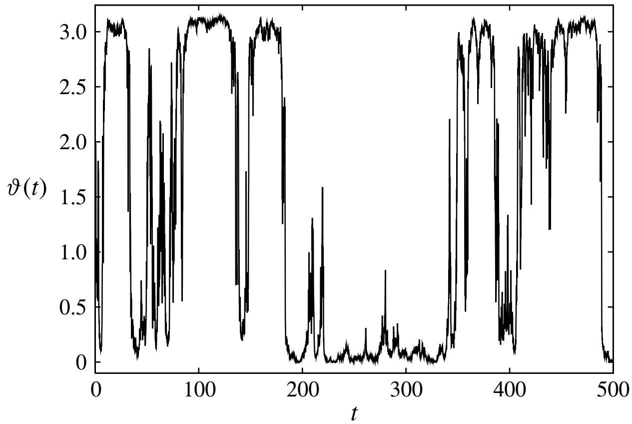

FIGURE 5. A typical time evolution of $\vartheta(t)$ in the tumbling regime ($\gamma = 1$, $b/a = 4.75$, $c/a = 10$, $d/a = 0.9$, $D'_R/a = 10^{-2}$); $\vartheta(t)$ has been computed by numerically integrating (3.14).

(Bretherton 1962). In the latter case, the dynamics of particles changes smoothly as a function of the shape coefficient.

It is moreover worth remarking that the function χ defined in (4.5) also determines the probability distribution of Jeffery's orbits in the presence of weak Brownian fluctuations (Leal & Hinch 1971). There does not seem to be, however, a simple relation between the orientation dynamics in the uniform shear and that in the axisymmetric random flow.

Finally, the results presented here also hold for elastic dumbbells or for slightly deformable spheroids, since for such particles the dynamics of the size and that of the orientation are decoupled (Olbricht, Rallison & Leal 1982). The study of highly deformable particles requires more detailed models that also account for the shape dynamics (e.g. Minale 2010) and is the topic of future work.

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Appendix A. Rotary diffusion

Rotary diffusion is introduced in the dynamics of $\boldsymbol{N}(t)$ by assuming that it produces isotropic Brownian fluctuations of the direction of $\boldsymbol{N}(t)$, but not of its length. Mathematically, this is obtained by adding a Laplacian term to the equation for $f(\boldsymbol{n}; t)$ which acts only on the orientation of $\boldsymbol{n}$. For a deterministic flow, $f(\boldsymbol{n}; t)$ thus satisfies the following equation (e.g. Leal & Hinch 1971; Hinch & Leal 1972; Brenner 1974; Bird *et al.* 1977; Doi & Edwards 1986):

$$\partial_t f = -\widetilde{\nabla} \cdot [\boldsymbol{w}(\boldsymbol{n}, t) f] + \frac{D_R}{2} \widetilde{\nabla}^2 f, \tag{A 1}$$

where

$$\widetilde{\nabla} \equiv \boldsymbol{\Sigma}(\boldsymbol{n}) \cdot \nabla_{\boldsymbol{n}} \tag{A 2}$$

with $\boldsymbol{\Sigma}(\boldsymbol{n}) = \boldsymbol{I} - \boldsymbol{nn}/|\boldsymbol{n}|^2$ and $\nabla_{\boldsymbol{n}} = (\partial/\partial n_1, \partial/\partial n_2, \partial/\partial n_3)$. The differential operator $\widetilde{\nabla}$ is the angular part of the gradient or, in other words, the restriction of the gradient to the sphere of radius $|\boldsymbol{n}|$. In (A 1), $\widetilde{\nabla}^2 \equiv \widetilde{\nabla} \cdot \widetilde{\nabla}$ and $\boldsymbol{w}(\boldsymbol{n}, t) = \boldsymbol{\kappa}(t) \cdot \boldsymbol{n} - [\boldsymbol{\kappa}(t){:}\boldsymbol{nn}]\boldsymbol{n}/|\boldsymbol{n}|^2$.

The matrix $\boldsymbol{\Sigma}$ satisfies:

$$\boldsymbol{\Sigma} = \boldsymbol{\Sigma}^{\mathrm{T}} = \boldsymbol{\Sigma}^2, \quad \boldsymbol{n} \times [\nabla_{\boldsymbol{n}} \cdot \boldsymbol{\Sigma}(\boldsymbol{n})] = \boldsymbol{0}, \quad \boldsymbol{\Sigma}(\boldsymbol{n}) \cdot \boldsymbol{n} = \boldsymbol{0}. \tag{A 3}$$

By using properties (A 3) and $\boldsymbol{w}(\boldsymbol{n}, t) \cdot \boldsymbol{n} = 0$, it is possible to rewrite (A 1) as follows:

$$\frac{\partial f}{\partial t} = -\frac{\partial}{\partial n_i}[w_i(\boldsymbol{n}, t)f] + \frac{D_R}{2}\frac{\partial}{\partial n_i}\Sigma_{ik}(\boldsymbol{n})\frac{\partial}{\partial n_j}\Sigma_{jk}(\boldsymbol{n})f. \tag{A 4}$$

Equation (2.3) is the Stratonovich stochastic differential equation associated with (A 4).

It is worth noting that given that the orientation vector has unit length, $f(\boldsymbol{n}; t)$ must take the form $f(\boldsymbol{n}; t) = F(\boldsymbol{n}; t)\delta(|\boldsymbol{n}| - 1)$ with $\partial F/\partial n = 0$. It would therefore be more natural to consider the evolution equation for $F(\boldsymbol{n}; t)$ instead of that for $f(\boldsymbol{n}; t)$; this is indeed the usual approach in the literature (Leal & Hinch 1971; Hinch & Leal 1972; Brenner 1974; Bird *et al.* 1977; Doi & Edwards 1986). In the present context, however, it is more convenient to formulate the problem in Cartesian coordinates and to move to angular variables afterwards.

Appendix B. Representation of the velocity correlation tensor of a homogeneous axisymmetric random flow

Consider the scalar variable $z = \mu r$ and the orthogonal unit vectors $\mathfrak{e}^{(1)} = (\boldsymbol{\lambda} \times \boldsymbol{r})/\rho$, $\mathfrak{e}^{(2)} = \mathfrak{e}^{(1)} \times \boldsymbol{\lambda}$ with $\rho = |\boldsymbol{\lambda} \times \boldsymbol{r}|$. Then, an alternative representation of the tensor $\boldsymbol{Q}$ defined in (2.7) is (Lindborg 1995):

$$\begin{aligned} Q_{ij}(\boldsymbol{r}) = R_1(\rho, z)\lambda_i\lambda_j + R_2(\rho, z)\mathfrak{e}_i^{(2)}\mathfrak{e}_j^{(2)} + R_3(\rho, z)\mathfrak{e}_i^{(1)}\mathfrak{e}_j^{(1)} \\ + R_4(\rho, z)(\lambda_i\mathfrak{e}_j^{(2)} + \lambda_j\mathfrak{e}_i^{(2)}), \end{aligned} \tag{B 1}$$

where up to second-order in ρ and z (see Lindborg 1995, p. 189):

$$R_1(\rho, z) = R_1(0, 0) - \mathfrak{a}_1\rho^2 - \mathfrak{b}_1 z^2 + \cdots, \tag{B 2a}$$

$$R_2(\rho, z) = R_2(0, 0) - \mathfrak{a}_2\rho^2 - \mathfrak{b}_2 z^2 + \cdots, \tag{B 2b}$$

$$R_3(\rho, z) = R_2(0, 0) - (3\mathfrak{a}_2 - \mathfrak{b}_1)\rho^2 - \mathfrak{b}_2 z^2 + \cdots, \tag{B 2c}$$

$$R_4(\rho, z) = \mathfrak{b}_1\rho z + \cdots. \tag{B 2d}$$

In the above equations, $\mathfrak{a}_1$, $\mathfrak{a}_2$, $\mathfrak{b}_1$ and $\mathfrak{b}_2$ are real constants. The functions R_1, R_2, R_3, R_4 are related to the functions A, B, C, D in (2.8) through the following identities (Lindborg 1995, p. 185):

$$R_1 = \mu^2 r^2 A + B + C + 2r\mu D, \quad R_2 = r^2(1 - \mu^2)A + B, \tag{B 3a}$$

$$R_3 = B, \quad R_4 = \mu r^2\sqrt{1 - \mu^2}\,A + r\sqrt{1 - \mu^2}\,D. \tag{B 3b}$$

The relations between the parameters $\mathfrak{a}_1$, $\mathfrak{a}_2$, $\mathfrak{b}_1$, $\mathfrak{b}_2$ and the parameters a, b, c, d used in the text are obtained by substituting (2.9) into (B3), by making the change of variables $r = (\rho^2 + z^2)^{1/2}$ and $\mu = z(\rho^2 + z^2)^{-1/2}$, and by comparing the resulting expressions for R_1, R_2, R_3, R_4 with the expansions given in (B2). It follows that

$$a = 2\mathfrak{a}_2 - \mathfrak{b}_1, \qquad b = 2(3\mathfrak{a}_2 - \mathfrak{b}_1) - 2\mathfrak{b}_2, \tag{B 4a}$$

$$c = 2(\mathfrak{a}_1 - \mathfrak{a}_2 - 3\mathfrak{b}_1 + \mathfrak{b}_2), \qquad d = 2(\mathfrak{b}_1 - \mathfrak{a}_2). \tag{B 4b}$$

In particular, the parameter σ defined in (4.7) is written:

$$\sigma = 2[(\mathfrak{a}_1 - \mathfrak{b}_2) + \gamma(\mathfrak{b}_1 - \mathfrak{a}_2]. \tag{B 5}$$

Moreover, by inverting (B4) and using inequalities (2.17), it is possible to show that $\mathfrak{a}_1$, $\mathfrak{a}_2$, $\mathfrak{b}_1$ and $\mathfrak{b}_2$ are strictly positive.

Appendix C. Positive semi-definiteness of the covariance tensor

The fourth-order tensor $\boldsymbol{\Gamma}$ is the covariance of a Gaussian second-order tensor, and must therefore be positive semi-definite, i.e.

$$\sum_{1\leqslant i,j,p,q\leqslant 3} U_{ij}\Gamma_{ijpq}U_{pq} \geqslant 0 \tag{C 1}$$

for all second-order tensors $\boldsymbol{U}$. Inequality (C 1) can be reinterpreted within the theory of positive semi-definite second-order tensors (e.g. Moakher 2008). Consider an invertible map ℓ which assigns to each pair of indices (i, j), $1 \leqslant i, j \leqslant 3$, a single index $\ell(i, j)$ ranging from 1 to 9. By means of the map ℓ, $\boldsymbol{U}$ can be regarded as a nine-dimensional vector; likewise, $\boldsymbol{\Gamma}$ can be regarded as a symmetric 9×9 second-order tensor, whose symmetry follows from $\Gamma_{ijpq} = \Gamma_{pqij}$. Accordingly, (C 1) can be rewritten in the following form:

$$\sum_{1\leqslant \ell(i,j),\ell(p,q)\leqslant 9} U_{\ell(i,j)}\Gamma_{\ell(i,j)\ell(p,q)}U_{\ell(p,q)} \geqslant 0. \tag{C 2}$$

Inequality (C 2) is the definition of positive semi-definiteness for second-order tensors. The theory of such tensors says that a necessary and sufficient condition for an Hermitian second-order tensor to be positive semi-definite is that all the principal minors of the tensor are non-negative (Gantmacher 1959, p. 307). When this condition is applied to $\Gamma_{\ell(i,j)\ell(p,q)}$ it yields the following inequalities:

$$\left.\begin{array}{ll} a + d \geqslant 0 & \text{if } a \geqslant 0, \\ 5a + d \geqslant 0 & \text{if } a < 0, \end{array}\right\} \tag{C 3}$$

and

$$\varpi_1 \equiv 4a - b + d \geqslant 0, \qquad \varpi_2 \equiv 4a + b + c + 6d \geqslant 0, \tag{C 4}$$

$$\varpi_3 \equiv 15a^2 - b^2 - (b - d)(c + 5d) + 2a(2c + 13d) \geqslant 0. \tag{C 5}$$

In this paper, the above inequalities are assumed to hold strictly; (2.17) follows from this assumption.

Some useful inequalities can be derived from (2.17). Here, $\gamma \neq 0$ (the case of a spherical particle is indeed trivial), $D_R' = 0$ (it is easily seen that a positive D_R' does not alter the inequalities below), and $\nu_3 \neq 0$ (the case $\nu_3 = 0$ is treated separately in appendix E).

First, $\mathscr{D}_{\vartheta\vartheta}(\vartheta)$ and $\mathscr{D}_{\varphi\varphi}(\vartheta)$ are positive for all $\gamma \neq 0$ and for all values of a, b, c, d satisfying (2.17). Indeed, the quantities $2\nu_1$ and $2\nu_1 + \nu_3 + \nu_4$ are quadratic polynomials in γ, their discriminant is equal to $-\varpi_3 < 0$, and for $\gamma = 1$ they are equal to $\varpi_1 > 0$

and to $\varpi_2 > 0$, respectively. Moreover, $2\nu_1 + \nu_5$ is a quadratic polynomial in γ with discriminant equal to $-(3a+d)(5a+d) < 0$ and takes the value $4a + d > 0$ for $\gamma = 1$. Hence

$$\nu_1 > 0, \quad 2\nu_1 + \nu_5 > 0, \quad 2\nu_1 + \nu_3 + \nu_4 > 0 \tag{C 6}$$

for all a, b, c, d and γ, and consequently

$$\mathscr{D}_{\vartheta\vartheta}(0) = \mathscr{D}_{\vartheta\vartheta}(\pi) = 2\nu_1 > 0, \tag{C 7a}$$

$$\mathscr{D}_{\varphi\varphi}(0) = \mathscr{D}_{\varphi\varphi}(\pi) = 2\nu_1 > 0, \tag{C 7b}$$

and

$$\mathscr{D}_{\vartheta\vartheta}(\vartheta) \geqslant \nu_4 \sin^2(\vartheta) + (2\nu_1 + \nu_3)\sin^4(\vartheta) \geqslant (2\nu_1 + \nu_3 + \nu_4)\sin^4(\vartheta) > 0, \tag{C 8a}$$

$$\mathscr{D}_{\varphi\varphi}(\vartheta) \geqslant (2\nu_1 + \nu_5)\sin^2(\vartheta) > 0 \tag{C 8b}$$

for all $0 < \vartheta < \pi$.

Secondly, $\psi_{st}(\vartheta)$ is bounded for all a, b, c, d and γ and for all $0 \leqslant \vartheta \leqslant \pi$. For $\Delta > 0$, this property is obvious. For $\Delta < 0$, note that

$$\left(\sqrt{-\Delta} + \nu_4\right)\left(\sqrt{-\Delta} - \nu_4\right) = -\nu_3 P_1(\gamma), \tag{C 9}$$

and

$$\left(\sqrt{-\Delta} + \nu_4 + 2\nu_3\right)\left(\sqrt{-\Delta} - \nu_4 - 2\nu_3\right) = -\nu_3 P_2(\gamma), \tag{C 10}$$

where $P_1(\gamma)$ and $P_2(\gamma)$ are quadratic polynomials in γ such that $P_1(1) = 4\varpi_1 > 0$ and $P_2(1) = 4\varpi_2 > 0$. Furthermore, the discriminants of $P_1(\gamma)$ and $P_2(\gamma)$ are equal to $-16\varpi_3 < 0$. Therefore $P_1(\gamma)$ and $P_2(\gamma)$ are positive for all γ, and the products on the left-hand sides of (C 9) and (C 10) have the same sign as $-\nu_3$.

Also observe that

$$\left(\sqrt{-\Delta} + \nu_4 + 2\nu_3\right)\left(\sqrt{-\Delta} - \nu_4\right) = -\nu_3\left[P_3(\gamma) - 2\sqrt{-\Delta}\right], \tag{C 11}$$

where $P_3(\gamma)$ is a quadratic polynomial satisfying: $P_3^2(\gamma) + 4\Delta = P_1(\gamma)P_2(\gamma) > 0$. Hence $P_3(\gamma) - 2\sqrt{-\Delta} \neq 0$ for all γ. Moreover, for $\gamma = 0$, $P_3(0) - 2\sqrt{-\Delta} = P_3(0) = \varpi_1 + \varpi_2 + 2(a+d) > 0$. As a result $P_3(\gamma) - 2\sqrt{-\Delta} > 0$ for all γ. The left-hand side of (C 11) therefore has the same sign as $-\nu_3$.

Three cases should now be distinguished.

(*a*) $\nu_3 < 0$: in this case, $\sqrt{-\Delta} \pm \nu_4 > 0$ and $\sqrt{-\Delta} - \nu_4 - 2\nu_3 > 0$ (recall that $\Delta = 8\nu_1\nu_3 - \nu_4^2$ with $\nu_1 > 0$), whence $\sqrt{-\Delta} + \nu_4 + 2\nu_3 > 0$ (see (C 10)). Therefore

$$\sqrt{-\Delta} + \nu_4 + 2\nu_3\sin^2(\vartheta) \geqslant \sqrt{-\Delta} + \nu_4 + 2\nu_3 > 0, \tag{C 12a}$$

$$\sqrt{-\Delta} - \nu_4 - 2\nu_3\sin^2(\vartheta) \geqslant \sqrt{-\Delta} - \nu_4 > 0. \tag{C 12b}$$

(*b*) $\nu_3 > 0$ and $\nu_4 > 0$: then $\sqrt{-\Delta} + \nu_4 > 0$ and consequently $\sqrt{-\Delta} - \nu_4 < 0$ (see (C 9)). Hence

$$\sqrt{-\Delta} + \nu_4 + 2\nu_3\sin^2(\vartheta) \geqslant \sqrt{-\Delta} + \nu_4 > 0, \tag{C 13a}$$

$$\sqrt{-\Delta} - \nu_4 - 2\nu_3\sin^2(\vartheta) \leqslant \sqrt{-\Delta} - \nu_4 < 0. \tag{C 13b}$$

(*c*) $\nu_3 > 0$ and $\nu_4 < 0$: for these values of the parameters, (C 10) and (C 11) yield the following relations:

$$\sqrt{-\Delta} + \nu_4 + 2\nu_3\sin^2(\vartheta) \leqslant \sqrt{-\Delta} + \nu_4 + 2\nu_3 < 0, \tag{C 14a}$$

$$\sqrt{-\varDelta} - \nu_4 - 2\nu_3 \sin^2(\vartheta) \geqslant \sqrt{-\varDelta} - \nu_4 - 2\nu_3 > 0. \tag{C 14b}$$

Inequalities (C12)–(C14) guarantee that if $\varDelta < 0$, the function $\chi(\vartheta)$ is bounded for all a, b, c, d, and γ.

For $\varDelta = 8\nu_1\nu_3 - \nu_4^2 = 0$, ν_4 cannot be zero since $\nu_1 > 0$ and $\nu_3 \neq 0$. Moreover, ν_3 and hence c must be positive given that $\nu_1 > 0$. Therefore, for $\nu_4 > 0$ the function $\chi(\vartheta)$ is bounded. The case $\nu_4 < 0$ requires a more detailed analysis. $\varDelta$ can be rewritten thus: $\varDelta = c(6a + 5d)\gamma^2(\gamma^2 - \varrho)$ with

$$\varrho = \frac{(2b + c + 5d)^2 - c(10a + c + 9d)}{c(6a + 5d)}. \tag{C 15}$$

(Note that, for $\gamma = 0$, $\nu_1 = (10a + c + 9d)/8$ and hence $10a + c + 9d > 0$ for all a, c, d as a consequence of (C 6); this is consistent with the positivity of the variance of the components of the vorticity – see (2.15).) Provided that $\varrho > 0$, $\varDelta$ vanishes for $\gamma = \pm\gamma_\star$ with $\gamma_\star = \sqrt{\varrho}$ (the case $\gamma = 0$ is not considered here). Now note that both ν_4 and $2\nu_3 + \nu_4$ are quadratic polynomials in γ. If $\gamma_{\star\star} \equiv 1 + (2b + 5d)/c$ is positive, then

$$\left.\begin{array}{ll} \nu_4 > 0 & \text{if } \gamma \in (0, \gamma_{\star\star}) \\ \nu_4 < 0 & \text{if } \gamma \notin [0, \gamma_{\star\star}] \end{array}\right\} \quad \text{and} \quad \left.\begin{array}{ll} 2\nu_3 + \nu_4 < 0 & \text{if } \gamma \in (-\gamma_{\star\star}, 0) \\ 2\nu_3 + \nu_4 > 0 & \text{if } \gamma \notin [-\gamma_{\star\star}, 0]. \end{array}\right\} \tag{C 16}$$

If $\gamma_{\star\star} < 0$, then

$$\left.\begin{array}{ll} \nu_4 > 0 & \text{if } \gamma \in (-\gamma_{\star\star}, 0) \\ \nu_4 < 0 & \text{if } \gamma \notin [-\gamma_{\star\star}, 0] \end{array}\right\} \quad \text{and} \quad \left.\begin{array}{ll} 2\nu_3 + \nu_4 < 0 & \text{if } \gamma \in (0, \gamma_{\star\star}) \\ 2\nu_3 + \nu_4 > 0 & \text{if } \gamma \notin [0, \gamma_{\star\star}]. \end{array}\right\} \tag{C 17}$$

Hence

$$\nu_4(2\nu_3 + \nu_4) > 0 \quad \forall\, |\gamma| < |\gamma_{\star\star}|. \tag{C 18}$$

Let us now show that $|\gamma_\star| < |\gamma_{\star\star}|$. The quantity $\gamma_{\star\star}^2 - \gamma_\star^2$ is written:

$$\gamma_{\star\star}^2 - \gamma_\star^2 = \frac{P_4(c)}{(6a + 5d)c^2}, \tag{C 19}$$

where $P_4(c)$ is a quadratic polynomial in c whose coefficients depend on a, b, d and such that $P_4(0) = (6a + 5d)\,(2b + 5d)^2 > 0$ and $\lim_{c\to\infty} P_4(c)/c^2 = 4\varpi_1 > 0$. Moreover, $P_4'(c) = 0$ if and only if $c = \widehat{c} \equiv -(2b + 5d)(12a - 2b + 5d)/(8\varpi_1)$, and:

$$P_4(\widehat{c}) = \frac{(2b + 5d)^2[16\varpi_3 + (2b + 5d)^2]}{16\varpi_1} > 0. \tag{C 20}$$

Therefore, $P_4(c) > 0$ for all $c > 0$ and hence $\gamma_{\star\star}^2 > \gamma_\star^2$ for all $c > 0$. As a conclusion, if $\varDelta = 0$, inequality (C 18) holds, and for $\nu_4 < 0$:

$$\nu_4 + 2\nu_3 \sin^2\vartheta \leqslant \nu_4 + 2\nu_3 < 0 \quad \forall\, 0 \leqslant \vartheta \leqslant \pi. \tag{C 21}$$

This result proves that $\chi(\vartheta)$ is bounded also for $\varDelta = 0$ and $\nu_4 < 0$.

Appendix D. Stationary probability density function of the orientation angle

The convection–diffusion equation (3.7) and its stationary solution can be obtained by using the general properties of the Fokker–Planck equation (Risken 1989). The starting point is the Fokker–Planck equation (3.3) for $f(\boldsymbol{n}; t)$. Consider the p.d.f. of the angles ϑ and φ:

$$\Psi(\vartheta, \varphi; t) \equiv \psi(\vartheta, \varphi; t) \sin\vartheta, \tag{D 1}$$

which is normalized thus: $\int_0^{\pi}\int_0^{2\pi} \Psi(\vartheta, \varphi; t)\,\mathrm{d}\vartheta\,\mathrm{d}\varphi = 1$ for all $t \geqslant 0$. The p.d.f. $\Psi(\vartheta, \varphi; t)$ satisfies a new Fokker–Planck equation, whose form is derived from (3.3) by using the transformation formulae for the drift and diffusion coefficients under a change of variables (Risken 1989, p. 88), by writing $f(n, \vartheta, \varphi; t) = \psi(\vartheta, \varphi; t)\delta(n - L)$ with $L = 1$, and by integrating the resulting equation with respect to n. The final result is:

$$\frac{\partial \Psi}{\partial t} = -\frac{\partial}{\partial \vartheta}[\mathscr{B}_\vartheta(\vartheta)\Psi] + \frac{1}{2}\frac{\partial^2}{\partial \vartheta^2}[\mathscr{A}_{\vartheta\vartheta}(\vartheta)\Psi] + \frac{1}{2}\mathscr{A}_{\varphi\varphi}(\vartheta)\frac{\partial^2 \Psi}{\partial \varphi^2}, \tag{D 2}$$

where

$$\mathscr{B}_\vartheta(\vartheta) = \beta_i(\boldsymbol{n})\frac{\partial \vartheta}{\partial n_i} + \frac{\alpha_{ij}(\boldsymbol{n})}{2}\frac{\partial^2 \vartheta}{\partial n_i \partial n_j} = \left[\nu_1 + \nu_2 \sin^2(\vartheta) + \nu_3 \sin^4(\vartheta)\right]\cot\vartheta, \tag{D 3a}$$

$$\mathscr{A}_{\vartheta\vartheta}(\vartheta) = \alpha_{ij}(\boldsymbol{n})\frac{\partial \vartheta}{\partial n_i}\frac{\partial \vartheta}{\partial n_j} = 2\nu_1 + \nu_4 \sin^2(\vartheta) + \nu_3 \sin^4(\vartheta), \tag{D 3b}$$

$$\mathscr{A}_{\varphi\varphi}(\vartheta) = \alpha_{ij}(\boldsymbol{n})\frac{\partial \varphi}{\partial n_i}\frac{\partial \varphi}{\partial n_j} = \nu_5 + 2\nu_1 \operatorname{cosec}^2(\vartheta) \tag{D 3c}$$

with ν_k, $k = 1, \ldots, 5$ defined in (3.10). Equations (D3) involve the Jacobian and the Hessian of the transformation from Cartesian to spherical coordinates, which can be calculated from (3.5). The convection–diffusion equation (3.7) follows from (D 1) and (D 2) with:

$$\mathscr{W}_\vartheta(\vartheta) = \mathscr{B}_\vartheta(\vartheta) - \frac{1}{2\sin\vartheta}\frac{\partial}{\partial \vartheta}[\mathscr{A}_{\vartheta\vartheta}(\vartheta)\sin\vartheta], \tag{D 4a}$$

$$\mathscr{D}_{\vartheta\vartheta}(\vartheta) = \mathscr{A}_{\vartheta\vartheta}(\vartheta), \tag{D 4b}$$

$$\mathscr{D}_{\varphi\varphi}(\vartheta) = \mathscr{A}_{\varphi\varphi}(\vartheta)\sin^2(\vartheta). \tag{D 4c}$$

The coefficients $\mathscr{A}_{\vartheta\vartheta}(\vartheta)$ and $\mathscr{A}_{\varphi\varphi}(\vartheta)$ are strictly positive as a consequence of (C7) and (C8). The boundary conditions for $\Psi(\vartheta, \varphi; t)$ are periodic, given that ϑ and φ are angular variables:

$$\Psi(\vartheta, \varphi; t) = \Psi(\vartheta + 2\pi, \varphi; t) \quad \text{and} \quad \Psi(\vartheta, \varphi; t) = \Psi(\vartheta, \varphi + 2\pi; t) \tag{D 5}$$

for all ϑ, φ and t. Moreover, at long times, $\Psi(\vartheta, \varphi; t)$ enjoys the same symmetries as $\psi(\vartheta, \varphi; t)$ as a consequence of the analogous symmetries of (D 2). In particular, the invariance of (D 2) under rotations about $\boldsymbol{\lambda}$ yields a one-dimensional Fokker–Planck equation for $\widehat{\Psi}(\vartheta; t) = \widehat{\psi}(\vartheta; t)\sin\vartheta$ with $\widehat{\psi}(\vartheta; t) = \int_0^{2\pi}\psi(\vartheta, \varphi; t)\,\mathrm{d}\varphi$. This equation is derived by integrating (D 2) with respect to φ from 0 to 2π and by using (D 5):

$$\frac{\partial \widehat{\Psi}}{\partial t} = -\frac{\partial}{\partial \vartheta}[\mathscr{B}_\vartheta(\vartheta)\widehat{\Psi}] + \frac{1}{2}\frac{\partial^2}{\partial \vartheta^2}[\mathscr{A}_{\vartheta\vartheta}(\vartheta)\widehat{\Psi}]. \tag{D 6}$$

The Itô stochastic differential equation associated with (D 6) is:

$$\dot{\vartheta}(t) = \mathscr{B}_\vartheta(\vartheta(t)) + \sqrt{\mathscr{A}_{\vartheta\vartheta}(\vartheta(t))}\,\eta(t), \quad 0 \leqslant \vartheta(t) \leqslant \pi, \tag{D 7}$$

where $\eta(t)$ is white noise. Equations (3.14) and (3.15) follow from (D 4a), (D 4b) and (D 7).

Consider now the functions:

$$\Phi(\vartheta) = \ln\left[\frac{\mathscr{A}_{\vartheta\vartheta}(\vartheta)}{2}\right] - 2\int_{\vartheta_0}^{\vartheta} \frac{\mathscr{B}_{\vartheta}(s)}{\mathscr{A}_{\vartheta\vartheta}(s)}\,\mathrm{d}s \tag{D 8}$$

and

$$g(\vartheta) = 2\int_{\vartheta_0}^{\vartheta} \frac{\mathrm{e}^{\Phi(s)}}{\mathscr{A}_{\vartheta\vartheta}(s)}\,\mathrm{d}s. \tag{D 9}$$

The stationary solution of (D 2) is written (see Risken 1989, p. 98):

$$\Psi_{st}(\vartheta) = \mathscr{N}\mathrm{e}^{-\Phi(\vartheta)} - \mathscr{S}g(\vartheta)\mathrm{e}^{-\Phi(\vartheta)}, \tag{D 10}$$

where $\mathscr{N}$ and $\mathscr{S}$ are constants. Consequently:

$$\psi_{st}(\vartheta) = \mathscr{N}\frac{\mathrm{e}^{-\Phi(\vartheta)}}{\sin\vartheta} - \mathscr{S}g(\vartheta)\frac{\mathrm{e}^{-\Phi(\vartheta)}}{\sin\vartheta}. \tag{D 11}$$

As $\mathscr{A}_{\vartheta\vartheta}(\vartheta)$ and $\mathscr{B}_{\vartheta}(\vartheta)$ are periodic, $\mathrm{e}^{-\Phi(\vartheta)}$ is also periodic. By contrast, $g(\vartheta)$ cannot be periodic given that $\mathscr{A}_{\vartheta\vartheta}(\vartheta) > 0$ for all $0 \leqslant \vartheta \leqslant \pi$ (see (D 4*b*), (C 7*a*), (C 8*a*)) and as a result $g'(\vartheta) > 0$ for all $0 \leqslant \vartheta \leqslant \pi$. Therefore, $\psi_{st}(\vartheta)$ satisfies (3.12) if and only if $\mathscr{S} = 0$, whence

$$\psi_{st}(\vartheta) = \frac{\mathscr{N}}{\mathscr{A}_{\vartheta\vartheta}(\vartheta)\sin\vartheta}\exp\left[2\int_{\vartheta_0}^{\vartheta}\frac{\mathscr{B}_{\vartheta}(s)}{\mathscr{A}_{\vartheta\vartheta}(s)}\,\mathrm{d}s\right]. \tag{D 12}$$

Equation (4.2) follows from (D 4*a*), (D 4*b*) and (D 12). For $\nu_3 \neq 0$, the integral on the right-hand side of (D 12) can be calculated by using the change of variable $y = \sin^2(z)$ and formulae 2.172, 2.175(1), and 2.177(1) in Gradshteyn & Ryzhik (1965). The final result is given in (4.4) and (4.5).

Appendix E. The case $\nu_3 = 0$

If $\nu_3 = 0$ and $\gamma \neq 0$ (i.e. $c = 0$), three cases should be distinguished.

For $\nu_3 = 0$, $\nu_4 \neq 0$, and $\nu_2 \neq 3\nu_4/2$, the integral in (4.2) can be easily calculated by means of the transformation $y = \sin^2(z)$ to yield:

$$\psi_{st}(\vartheta) = \mathscr{N}\left(2\nu_1 + \nu_4\sin^2\vartheta\right)^{(\nu_2/\nu_4)-(3/2)}. \tag{E 1}$$

The stationary p.d.f. is bounded as a consequence of (C 6) with $\nu_3 = 0$. By examining the first derivative of the above function, it can be shown that, depending on the value of the parameters, ψ_{st} has either three or five extrema in the interval $0 \leqslant \vartheta \leqslant \pi$. Therefore, the four regimes identified for $\nu_3 \neq 0$ also describe the dynamics of a non-spherical particle for $\nu_3 = 0$.

For $\nu_3 = \nu_4 = 0$ (i.e. $c = 2b + 5d = 0$), the stationary solution is:

$$\psi_{st}(\vartheta) = \mathscr{N}\exp\left[\frac{\nu_2}{2\nu_1}\sin^2(\vartheta)\right]. \tag{E 2}$$

For $\nu_2/\nu_1 > 0$, ψ_{st} has two minima at 0 and π and one maximum at $\pi/2$, and hence the particle rotates in the plane orthogonal to $\boldsymbol{\lambda}$. For $\nu_2/\nu_1 < 0$, ψ_{st} has two maxima at 0 and π and one minimum at $\pi/2$; therefore the particle tumbles between the direction parallel to $\boldsymbol{\lambda}$ and that antiparallel to $\boldsymbol{\lambda}$.

Finally, for $\nu_3 = 0$ and $\nu_2 = 3\nu_4/2$ (i.e. $c = 2b + (5+\gamma)d = 0$), (E 1) implies that the stationary statistics of orientations is isotropic: $\psi_{st}(\vartheta) = (4\pi)^{-1}$.

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