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Short-time self-diffusion, collective diffusion and effective viscosity of dilute hard sphere magnetic suspensions

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The virial corrections to short-time self- and collective diffusion coefficients as well as the effective viscosity are calculated for suspensions of hard spheres with the same radii and constant (blocked within the particle) magnetization modelled by a point dipole. Analytic, integral formulae derived from basic principles of statistical mechanics are provided for both cases – in the absence and in the presence of an external magnetic field. In the former case the diffusion and viscosity coefficients are evaluated numerically as a function of the strength of magnetic interactions between the particles and it is reported that the translational collective diffusion coefficient is significantly greater than all the other coefficients. In the presence of an external magnetic field the coefficients become anisotropic and are evaluated in the asymptotic regime of weak interparticle magnetic interactions.

Key words: colloids, low-Reynolds-number flows, magnetic fluids

1. Introduction

Suspensions of magnetic nanoparticles and their macroscopic properties have been a subject of intense investigations in the past few decades due to the large variety of applications, both biomedical and technical. The action of the magnetic field on the suspended magnetic nanoparticles opens the possibility for control of the self-diffusion processes (studied by e.g. Pusey & van Megen 1983; Cichocki & Felderhof 1988; van Megen & Underwood 1989, and many others) and likewise the process of sedimentation, i.e. the collective movement of a suspension under a given force field or light scattering (cf. Batchelor 1972; Cebula *et al.* 1981; Kops-Werkhoven & Fijnaut 1981, 1982; Segre, Behrend & Pusey 1995). However, arguably the most important applications of ferrofluids result from the possibility of controlling their effective viscosity with an externally applied magnetic field. Here we study the short-time, or equivalently high-frequency, transport coefficients, in which case the transport

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processes do not affect the probability distribution of particles, which is the same as the thermal equilibrium distribution.

The potentially relevant biomedical applications include magnetic hyperthermia and thermoablation in cancer treatment, which, besides stopping tumour growth by raising its temperature, reduce the side effects on healthy tissue (cf. Pankhurst *et al.* 2003; Trahms 2009). The temperature rise at least partly results from dissipation of the magnetic energy of the external field alternating at high frequency ($\sim 10^2$ kHz) into viscous heating of the nanoparticles. Another interesting application is magnetorelaxometry, which uses alternating magnetic fields to monitor biological binding reactions and is often described by the Smoluchowski model (Sonntag & Strenge 1988), where binding results from diffusion-driven collisions of particles. The response of the magnetic suspension to the alternating magnetic field depends strongly on the effective viscosity (see the review by Trahms 2009). On the other hand, there are also technical applications of ferrofluids where high-frequency magnetic fields are used and the effective viscosity, in particular the high-frequency effective viscosity, plays a crucial role in the dynamics. These involve worm-like locomotion systems, where an elastic capsule filled with a ferrofluid travels in an external magnetic field alternating at frequencies up to kHz or locomotion of magnetic fluid layers driven by travelling external magnetic field waves (cf. Zimmermann *et al.* 2007; Trahms 2009). Another example includes magnetofluidic dampers in which the oscillatory magnetic field is used to control the viscosity of a ferrofluid inside a damper (cf. Bayat *et al.* 2009).

To the best knowledge of the authors a systematic study of the high-frequency transport coefficients of ferrofluids relying on basic principles of statistical mechanics has never been done. There have been numerous studies into the long-time transport coefficients for ferrofluids, such as, for example, Buevich, Zubarev & Ivanov (1989), Morozov (1993), Bacri *et al.* (1995), Morozov (1996) and Pshenichnikov, Elfimova & Ivanov (2011) based on the idea of transport driven by the chemical potential gradient. Here we determine the self- and collective diffusion coefficients as well as the effective viscosity at short times for diluted magnetic suspensions within the scope of the virial expansion and without any phenomenological input to the theory. We consider a dilute suspension of spherical particles subject to a quasiperiodic flow or alternating magnetic field. The period of oscillation T of the flow and/or the magnetic field is less than the time necessary for a particle to diffuse across its diameter, i.e. $T < (2a)^2/D_0$, where $D_0 = k_B T/6\pi\eta a$ is the single-particle diffusion mobility coefficient, η is the ambient-fluid viscosity, and $2a$ is the particle diameter, so that the contribution from Brownian motions is negligible. The Reynolds number based on the size L of the system (characteristic length scale of the flow) and the characteristic velocity u_c is assumed to be low compared with unity – that is, $Re = u_c L/\nu \ll 1$, where ν is the kinematic viscosity, i.e. we consider the Stokes limit. The flow and the magnetic field are quasisteady in the sense that their period T is small compared with the typical time L/u_c, so that the fluid performs small oscillations and the particle positions and orientations are practically frozen. It follows that the nonlinear term in the Navier–Stokes equation is negligible compared to the time derivative of the velocity field. However, the period T is large compared with the typical time L^2/ν for the diffusion of vorticity of the flow, so that the quasisteady Stokes equations apply; in other words, the velocity time derivative is much smaller than the viscous term and can be neglected. In summary, we consider the range $L^2/\nu \ll T \ll L/u_c$, which is consistent with $Re \ll 1$ and $T < (2a)^2/D_0$. Under these assumptions we evaluate the so-called short-time transport coefficients. In the literature they are often referred to as high-frequency transport coefficients.

The physical meaning of the assumptions made is as follows. The inequality $L^2/\nu \ll (2a)^2/D_0$ has to be satisfied in a system, so that a time scale T can be chosen satisfying $L^2/\nu \ll T < (2a)^2/D_0$. The inequality $L^2/\nu \ll (2a)^2/D_0$ is equivalent to $\eta \gg \sqrt{k_B T \rho L^2/24\pi a^3}$ and hence the latter can be considered a condition for the applicability of the analysis presented in this paper. This condition is satisfied, for example, for paraffin-based ferrofluids (cf. Hezaveh, Fazlali & Noshadi 2012), in which case the viscosity is in the range $\eta \sim 10$–10^3 Pa s. In general, the short-time transport coefficients are relevant and should be considered in the case of ferrofluids with relatively large magnetic particles, say 10–50 nm, and high viscosities of the carrier fluid η.

Our analysis generalizes the results of Cichocki, Ekiel-Jezewska & Wajnryb (1999, 2003) and Cichocki *et al.* (2002) to the case of ferrofluids. We assume that the particles are 'magnetically hard', i.e. the magnetic moment is 'blocked' within each particle and no Néel relaxation mechanism occurs (which of the two relaxation mechanisms for magnetized particles under the action of an external magnetic field dominates depends mainly on the sizes of particles and the temperature of the suspension). All the particles are also assumed to have magnetic moments of the same magnitude. The liquid in which the magnetic particles are immersed is assumed to be non-magnetic. Furthermore, the well-known effect of forming chain-like agglomerates or clusters by the nanoparticles, which is present when nanoparticles exceed a certain critical size, and in such a case strongly affects the fluid's viscous behaviour (see e.g. Odenbach 2003, 2004; Zubarev & Chirikov 2010), is assumed negligible here. The paper is structured as follows. First, in § 2, we calculate the transport coefficients for suspensions of magnetically interacting nanoparticles, but in the absence of an external magnetic field. The dependence of the transport coefficients on the parameter measuring the strength of the interparticle interactions is established. Section 3 is devoted to the calculation of the transport coefficients under the influence of an external magnetic field. The complete integral formulae are provided and then the asymptotic limit of weak interparticle magnetic interactions, which should be relevant to suspensions of small magnetic nanoparticles (cf. Odenbach 2003, 2004), is studied. In this limit the analytical dependence of the transport coefficients on the magnitude of the external magnetic field is derived. We end with some concluding remarks in § 4.

2. The diffusion coefficients and viscosity in the absence of an external magnetic field

2.1. *Formulae*

The short-time transport coefficients are given as the equilibrium ensemble average of N-particle hydrodynamic mobility matrices. To evaluate this average we assume that the suspension of particles is semidilute, i.e. its volume fraction is not large, and we expand the ensemble average in powers of the volume fraction ϕ, keeping only the lowest powers of ϕ. The coefficients at the order of ϕ and ϕ^2 (virial coefficients) are given as two-particle ensemble averages of the corresponding two-particle mobilities (see Cichocki *et al.* 1999, 2002, 2003). Now the ensemble averages contain the two-particle Boltzmann factors related to the interactions between pairs of particles' magnetic moments; in the case when the external magnetic field is present, studied in § 3, additional Boltzmann factors correspond to the interactions of particles' moments with the external magnetic field.

Let us define the volume fraction for the suspended spherical particles, each of radius a, as

$$\phi = \tfrac{4}{3}\pi a^3 n, \tag{2.1}$$

where $n = N/V$ is the number of particles per unit volume. Cichocki *et al.* (1999, 2003) and Cichocki *et al.* (2002) derived explicit formulae for the virial expansion coefficients of the self-diffusion, collective diffusion and effective viscosity for suspensions of spherical particles interacting only via the hydrodynamic interactions. We exploit their first-order formulae, i.e. linear in the volume fraction ϕ for the self-diffusion and collective diffusion (sedimentation coefficient):

$$D^{qq} = D_0(1 + \mathcal{D}^{qq}\phi + \cdots), \quad K^{qq} = 1 + (\mathcal{K}^{qq}_{overlap} + \mathcal{K}^{qq})\phi + \cdots, \tag{2.2a,b}$$

where the superscript $qq = tt$ for translational degrees of freedom and $qq = rr$ for rotational degrees of freedom. $\mathcal{K}^{qq}_{overlap}$ is the contribution from virtual overlaps (cf. Cichocki *et al.* 2002), which can be evaluated analytically for both the translational degrees of freedom $\mathcal{K}^{tt}_{overlap} = -5$ and rotational degrees of freedom $\mathcal{K}^{rr}_{overlap} = -1$ (cf. Cichocki & Felderhof 1989). In case of the effective viscosity of a suspension of hard spherical particles immersed in a fluid of viscosity η, the first-order correction $5\eta\phi/2$ derived by Einstein (1956) is obtained by neglecting any interparticle interactions, and hence it is necessary to consider the next-order correction, which is quadratic in the volume fraction ϕ,

$$\eta_{eff} = \eta[1 + \tfrac{5}{2}\phi + (\tfrac{5}{2} + \mathcal{G})\phi^2 + \cdots], \tag{2.3}$$

where the virtually overlapping part is equal to $5\phi^2/2$ (cf. Cichocki *et al.* 2003). In that way we calculate the corrections to all the aforementioned coefficients, i.e. $\mathcal{D}^{tt}$, $\mathcal{D}^{rr}$, $\mathcal{K}^{tt}$, $\mathcal{K}^{rr}$ and $\mathcal{G}$ which include interactions between pairs of suspended particles.

In the case of magnetic particles with magnetic moments $\boldsymbol{m}_i$, all of the same length $\|\boldsymbol{m}_i\| = m$, where i is the particle number, the magnetic interactions between them must be included via the probabilistic factor

$$\mathrm{e}^{-E_{ij}/k_B T}, \tag{2.4}$$

where

$$E_{ij} = \frac{\mu_0 m^2}{4\pi a^3 r_{ij}^3}[\hat{\boldsymbol{m}}_i \cdot \hat{\boldsymbol{m}}_j - 3(\hat{\boldsymbol{m}}_i \cdot \hat{\boldsymbol{r}})(\hat{\boldsymbol{m}}_j \cdot \hat{\boldsymbol{r}})] \tag{2.5}$$

is the potential energy of magnetic interaction between a pair of particles, μ_0 is the magnetic permeability of vacuum, k_B the Boltzmann constant, T the temperature and $\hat{\boldsymbol{m}}_i = \boldsymbol{m}_i/m_i$. Moreover, we denote the position vector of particle i by $\boldsymbol{R}_i$, the interparticle position vector, i.e. the vector of position of particle i with respect to particle j, by $\boldsymbol{R}_{ij} = \boldsymbol{R}_i - \boldsymbol{R}_j$, its length by R_{ij}, and with this notation $r_{ij} = R_{ij}/a$ and $\hat{\boldsymbol{r}}_{ij} = \boldsymbol{R}_{ij}/R_{ij}$. Assuming the z axis to be along the non-dimensional interparticle position vector $\boldsymbol{r}_{ij}$ and the dipole moment $\boldsymbol{m}_i$ in the xz plane, the first-order virial corrections to the self- and collective diffusion coefficients and second-order virial correction to the effective viscosity can be expressed by the following formulae (for detailed derivations see Cichocki *et al.* 1999, 2002, 2003)

$$\mathcal{D}^{qq} = \frac{\zeta^{qq}}{8\pi}\int_0^{\pi} \mathrm{d}\theta' \int_0^{\pi} \mathrm{d}\theta \int_0^{2\pi} \mathrm{d}\varphi \int_2^{\infty} \mathrm{d}r \sin\theta' \sin\theta r^2 d^{qq}(r) \times \exp\left[-\frac{\epsilon}{r^3}(\sin\theta' \sin\theta \cos\varphi - 2\cos\theta' \cos\theta)\right], \tag{2.6}$$

$$\mathcal{K}^{qq} = \frac{\zeta^{qq}}{8\pi}\int_0^{\pi} \mathrm{d}\theta' \int_0^{\pi} \mathrm{d}\theta \int_0^{2\pi} \mathrm{d}\varphi \int_2^{\infty} \mathrm{d}r \sin\theta' \sin\theta r^2 \times \left\{k^{qq}(r) \exp\left[-\frac{\epsilon}{r^3}(\sin\theta' \sin\theta \cos\varphi - 2\cos\theta' \cos\theta)\right] - \kappa^{qq}(r)\right\}, \tag{2.7}$$

$$\mathcal{G} = \frac{3\zeta^{dd}}{16\pi}\int_0^{\pi} \mathrm{d}\theta' \int_0^{\pi} \mathrm{d}\theta \int_0^{2\pi} \mathrm{d}\varphi \int_2^{\infty} \mathrm{d}r \sin\theta' \sin\theta r^2 \times \left\{g(r) \exp\left[-\frac{\epsilon}{r^3}(\sin\theta' \sin\theta \cos\varphi - 2\cos\theta' \cos\theta)\right] - \gamma^{dd}(r)\right\}, \tag{2.8}$$

$$\epsilon = \frac{\mu_0 m^2}{4\pi k_B T a^3}, \tag{2.9}$$

where again $qq = tt$ or $qq = rr$ for translational and rotational coefficients, respectively. In the above, $\zeta^{tt} = 6\pi\eta a$, $\zeta^{rr} = 8\pi\eta a^3$ and $\zeta^{dd} = 20\pi\eta a^3/3$ are the single-sphere translational, rotational and hydrodynamic dipole friction coefficients, θ' is the angle between the magnetic dipole moment $\boldsymbol{m}_i$ and the z axis (cf. figure 3*a*), and r is a variable corresponding to the interparticle distance no longer dependent on the particular particle numbers i and j after taking the volume average in the virial expressions. The hydrodynamic mobility functions $d^{qq}(r)$, $k^{qq}(r)$ and $g(r)$ are obtained in the following way. The necessary mobility matrix components can be expressed as follows:

$$\boldsymbol{\mu}_{11}^{qq}(\boldsymbol{r}) = \frac{1}{\zeta^{qq}}[\boldsymbol{I} + M_{11\parallel}^{qq}(r)\hat{\boldsymbol{r}}\hat{\boldsymbol{r}} + M_{11\perp}^{qq}(r)(\boldsymbol{I} - \hat{\boldsymbol{r}}\hat{\boldsymbol{r}})], \tag{2.10a}$$

$$M_{11\parallel}^{qq}(r) = \sum_{n=1}^{\infty}\frac{a_{\parallel n}^{qq}}{r^n}, \quad M_{11\perp}^{qq}(r) = \sum_{n=1}^{\infty}\frac{a_{\perp n}^{qq}}{r^n}, \tag{2.10b,c}$$

$$\boldsymbol{\mu}_{12}^{qq}(\boldsymbol{r}) = \frac{1}{\zeta^{qq}}[\boldsymbol{M}_{RP}^{qq}(\boldsymbol{r}) + M_{12\parallel}^{qq}(r)\hat{\boldsymbol{r}}\hat{\boldsymbol{r}} + M_{12\perp}^{qq}(r)(\boldsymbol{I} - \hat{\boldsymbol{r}}\hat{\boldsymbol{r}})], \tag{2.11a}$$

$$M_{12\parallel}^{qq}(r) = \sum_{n=4}^{\infty}\frac{b_{\parallel n}^{qq}}{r^n}, \quad M_{12\perp}^{qq}(r) = \sum_{n=4}^{\infty}\frac{b_{\perp n}^{qq}}{r^n}, \tag{2.11b,c}$$

$$\boldsymbol{\mu}_{11}^{dd}(\boldsymbol{r}) = \frac{1}{\zeta^{dd}}[\boldsymbol{I} + M_{11\,0}^{dd}(r)\boldsymbol{t}_0(\hat{\boldsymbol{r}}) + M_{11\,1}^{dd}(r)\boldsymbol{t}_1(\hat{\boldsymbol{r}}) + M_{11\,2}^{dd}(r)\boldsymbol{t}_2(\hat{\boldsymbol{r}})], \tag{2.12a}$$

$$\boldsymbol{\mu}_{12}^{dd}(\boldsymbol{r}) = \frac{1}{\zeta^{dd}}[\boldsymbol{M}_{RP}^{dd}(\boldsymbol{r}) + M_{12\,0}^{dd}(r)\boldsymbol{t}_0(\hat{\boldsymbol{r}}) + M_{12\,1}^{dd}(r)\boldsymbol{t}_1(\hat{\boldsymbol{r}}) + M_{12\,2}^{dd}(r)\boldsymbol{t}_2(\hat{\boldsymbol{r}})], \tag{2.12b}$$

$$M_{11\,0}^{dd}(r) = \sum_{n=6}^{\infty}\frac{c_{11\,0n}^{dd}}{r^n}, \quad M_{11\,1}^{dd}(r) = \sum_{n=6}^{\infty}\frac{c_{11\,1n}^{dd}}{r^n}, \quad M_{11\,2}^{dd}(r) = \sum_{n=6}^{\infty}\frac{c_{11\,2n}^{dd}}{r^n}, \tag{2.12c-e}$$

$$M_{12\,0}^{dd}(r) = \sum_{n=6}^{\infty}\frac{c_{12\,0n}^{dd}}{r^n}, \quad M_{12\,1}^{dd}(r) = \sum_{n=6}^{\infty}\frac{c_{12\,1n}^{dd}}{r^n}, \quad M_{12\,2}^{dd}(r) = \sum_{n=6}^{\infty}\frac{c_{12\,2n}^{dd}}{r^n}, \tag{2.12f-h}$$

where the subscript *RP* for the mobility tensor denotes the Rotne–Prager–Yamakawa level of approximation for hydrodynamic interactions (cf. Rotne & Prager 1969;

Yamakawa 1970; Wajnryb *et al.* 2013), corresponding to the effect that a flow modified by the presence of one particle has on the velocities of another particle; thus, terms are included up to order $(1/r)^3$ in the mobility components $\boldsymbol{M}^{tt}_{RP}(\boldsymbol{r})$ and $\boldsymbol{M}^{rr}_{RP}(\boldsymbol{r})$,

$$\boldsymbol{M}^{qq}_{RP}(\boldsymbol{r}) = \sum_{n=1}^{3} \frac{b^{qq}_{\parallel n}}{r^n}\hat{\boldsymbol{r}}\hat{\boldsymbol{r}} + \sum_{n=1}^{3} \frac{b^{qq}_{\perp n}}{r^n}(\boldsymbol{I} - \hat{\boldsymbol{r}}\hat{\boldsymbol{r}}), \tag{2.13}$$

and up to order $(1/r)^5$ in the dipolar component $\boldsymbol{M}^{dd}_{RP}(\boldsymbol{r})$,

$$\boldsymbol{M}^{dd}_{RP}(\boldsymbol{r}) = \sum_{n=3}^{5} \frac{c^{dd}_{12\,0n}}{r^n}\boldsymbol{t}_0(\hat{\boldsymbol{r}}) + \sum_{n=3}^{5} \frac{c^{dd}_{12\,1n}}{r^n}\boldsymbol{t}_1(\hat{\boldsymbol{r}}) + \sum_{n=3}^{5} \frac{c^{dd}_{12\,2n}}{r^n}\boldsymbol{t}_2(\hat{\boldsymbol{r}}); \tag{2.14}$$

where the fourth-rank tensors $\boldsymbol{t}_0(\hat{\boldsymbol{r}})$, $\boldsymbol{t}_1(\hat{\boldsymbol{r}})$ and $\boldsymbol{t}_2(\hat{\boldsymbol{r}})$ are given in appendix A (cf. also Cichocki, Felderhof & Schmitz 1988; Kim & Karrila 1991). Coefficients $a^{qq}_{\parallel n}$, $a^{qq}_{\perp n}$, $b^{qq}_{\parallel n}$, $b^{qq}_{\perp n}$ and $c^{dd}_{11\,0n}$, $c^{dd}_{11\,1n}$, $c^{dd}_{11\,2n}$, $c^{dd}_{12\,0n}$, $c^{dd}_{12\,1n}$, $c^{dd}_{12\,2n}$ are calculated from $n=0$ up to $n=1000$ via the multipole expansion method using the HYDROMULTIPOLE code (cf. Cichocki *et al.* 1994; Zurita-Gotor, Blawzdziewicz & Wajnryb 2007). The final formulae for the functions $d^{qq}(r)$, $k^{qq}(r)$ and $g(r)$, according to Cichocki *et al.* (1999, 2003) and Cichocki *et al.* (2002), are obtained by subtracting the Rotne–Prager–Yamakawa level terms and contracting the tensor indices in the following way:

$$d^{qq}(r) = \mathrm{Tr}\left[\boldsymbol{\mu}^{qq}_{11}(\boldsymbol{r}) - \frac{1}{\zeta^{qq}}\boldsymbol{I}\right] = \frac{1}{\zeta^{qq}}[M^{qq}_{11\parallel}(r) + 2M^{qq}_{11\perp}(r)], \tag{2.15}$$

$$\begin{aligned} k^{qq}(r) &= \mathrm{Tr}\left[\boldsymbol{\mu}^{qq}_{11}(\boldsymbol{r}) - \frac{1}{\zeta^{qq}}\boldsymbol{I} + \boldsymbol{\mu}^{qq}_{12}(\boldsymbol{r})\right] \\ &= \frac{1}{\zeta^{qq}}\left[M^{qq}_{11\parallel}(r) + M^{qq}_{12\parallel}(r) + \sum_{n=1}^{3}\frac{b^{qq}_{\parallel n}}{r^n} + 2\left(M^{qq}_{11\perp}(r) + M^{qq}_{12\perp}(r) + \sum_{n=1}^{3}\frac{b^{qq}_{\perp n}}{r^n}\right)\right], \end{aligned} \tag{2.16}$$

$$\kappa^{tt}(r) = \frac{1}{\zeta^{tt}}\mathrm{Tr}\,\boldsymbol{M}^{tt}_{RP}(\boldsymbol{r}) = \sum_{n=1}^{3}\frac{b^{tt}_{\parallel n} + 2b^{tt}_{\perp n}}{r^n}, \quad \kappa^{rr}(r) = \frac{1}{\zeta^{rr}}\mathrm{Tr}\,\boldsymbol{M}^{rr}_{RP}(\boldsymbol{r}) = \sum_{n=1}^{3}\frac{b^{rr}_{\parallel n} + 2b^{rr}_{\perp n}}{r^n} = 0, \tag{2.17a,b}$$

$$\begin{aligned} g(r) &= \left[\boldsymbol{\mu}^{dd}_{11}(\boldsymbol{r}) - \frac{1}{\zeta^{dd}}\boldsymbol{I} + \boldsymbol{\mu}^{dd}_{12}(\boldsymbol{r})\right]_{\alpha\beta\beta\alpha} \\ &= \frac{1}{\zeta^{dd}}\left[M^{dd}_{11\,0}(r) + M^{dd}_{12\,0}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,0n}}{r^n} + 2\left(M^{dd}_{11\,1}(r) + M^{dd}_{12\,1}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,1n}}{r^n}\right)\right. \\ &\quad \left. + 2\left(M^{dd}_{11\,2}(r) + M^{dd}_{12\,2}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,2n}}{r^n}\right)\right], \end{aligned} \tag{2.18}$$

$$\gamma^{dd}(r) = \frac{1}{\zeta^{dd}}\mathrm{Tr}\,\boldsymbol{M}^{dd}_{RP}(\boldsymbol{r}) = \sum_{n=3}^{5}\frac{c^{dd}_{12\,0n} + 2c^{dd}_{12\,1n} + 2c^{dd}_{12\,2n}}{r^n} = 0, \tag{2.19}$$

where $\mathrm{Tr}\,\boldsymbol{A} = A_{\alpha\alpha}$. Note that $\kappa^{rr}(r) = \gamma^{dd}(r) = 0$, and only $\kappa^{tt}(r) \neq 0$.

2.2. *Monotonicity of coefficients and their numerical calculation*

Because the mobility functions satisfy

$$d^{qq}(r) < 0, \quad k^{tt}(r) > 0, \quad k^{rr}(r) < 0, \quad g(r) > 0, \quad \text{for all } r > 2 \text{ and } qq = tt \text{ or } rr, \tag{2.20a–d}$$

it is a simple task to demonstrate that the following inequalities for the diffusion coefficients and the viscosity must be satisfied

$$\frac{\mathrm{d}D^{qq}}{\mathrm{d}\epsilon} < 0, \quad \frac{\mathrm{d}^2 D^{qq}}{\mathrm{d}\epsilon^2} < 0, \tag{2.21a,b}$$

$$\frac{\mathrm{d}K^{rr}}{\mathrm{d}\epsilon} < 0, \quad \frac{\mathrm{d}^2 K^{rr}}{\mathrm{d}\epsilon^2} < 0, \tag{2.21c,d}$$

$$\frac{\mathrm{d}K^{tt}}{\mathrm{d}\epsilon} > 0, \quad \frac{\mathrm{d}^2 K^{tt}}{\mathrm{d}\epsilon^2} > 0, \tag{2.21e,f}$$

$$\frac{\mathrm{d}\eta_{eff}}{\mathrm{d}\epsilon} > 0, \quad \frac{\mathrm{d}^2 \eta_{eff}}{\mathrm{d}\epsilon^2} > 0, \tag{2.21g,h}$$

and for the virial corrections we have

$$\mathcal{D}^{qq} < 0, \quad \mathcal{K}^{rr} < 0, \quad \mathcal{G} > 0. \tag{2.22a–c}$$

The proof of the above relations is done by simply taking the derivatives in (2.6)–(2.8) with respect to ϵ and considering the dominant contribution in the integrand determined by the positive sign under the exponential function, i.e. $(\sin\theta' \sin\theta \cos\varphi - 2\cos\theta' \cos\theta) < 0$. Therefore, the virial corrections to the self-diffusion coefficients $\mathcal{D}^{tt}$ and $\mathcal{D}^{rr}$ and the rotational collective diffusion coefficient $\mathcal{K}^{rr}$ are negative and decrease at an increasing rate with increasing strength of the magnetic interactions ϵ between particles. The virial corrections to the collective diffusion coefficient $\mathcal{K}^{tt}$ and the effective viscosity $\mathcal{G}$ increase at an increasing rate with ϵ; $\mathcal{G}$ is positive for all values of ϵ, whereas $\mathcal{K}^{tt}$ is negative for $\epsilon = 0$ and monotonically increases to reach positive values for ϵ greater than some critical value ϵ_{crit} defined by $\mathcal{K}^{tt}(\epsilon_{crit}) = 0$. Furthermore, by making use of the MAPLE software we have numerically computed the integrals in (2.6)–(2.8), and the dependence on ϵ of the virial corrections to all the diffusion and effective viscosity coefficients is depicted on figure 1. The dashed lines represent the asymptotic dependence of each of the plotted coefficients in the limit of weak magnetic interactions $\epsilon \ll 1$, studied in detail in § 3. The explicit asymptotic dependencies are provided in appendix B. Note that the asymptotic formulae (B 4a–e) provide a good approximation of the coefficients $\mathcal{D}^{tt}$, $\mathcal{D}^{rr}$, $\mathcal{K}^{rr}$ and $\mathcal{G}$ even for values of ϵ up to approximately 10. However, in the case of the translational collective diffusion coefficient $\mathcal{K}^{tt}$ the asymptotic approximation for $\epsilon \ll 1$ is satisfactory only for very small values of ϵ. This is because of the term

$$\begin{aligned} &\frac{\zeta^{tt}}{8\pi} \sin\theta' \sin\theta r^2 \kappa^{tt}(r) \left\{ \exp\left[-\frac{\epsilon}{r^3} (\sin\theta' \sin\theta \cos\varphi - 2\cos\theta' \cos\theta) \right] - 1 \right\} \\ &\quad = \frac{\zeta^{tt}}{8\pi} \sin\theta' \sin\theta 3r \left\{ \exp\left[-\frac{\epsilon}{r^3} (\sin\theta' \sin\theta \cos\varphi - 2\cos\theta' \cos\theta) \right] - 1 \right\} \end{aligned} \tag{2.23}$$

in the integrand of expression (2.7), the Taylor expansion of which is convergent for all values of $r > 2$ only for small values of ϵ. Moreover, it is the above term (2.23)

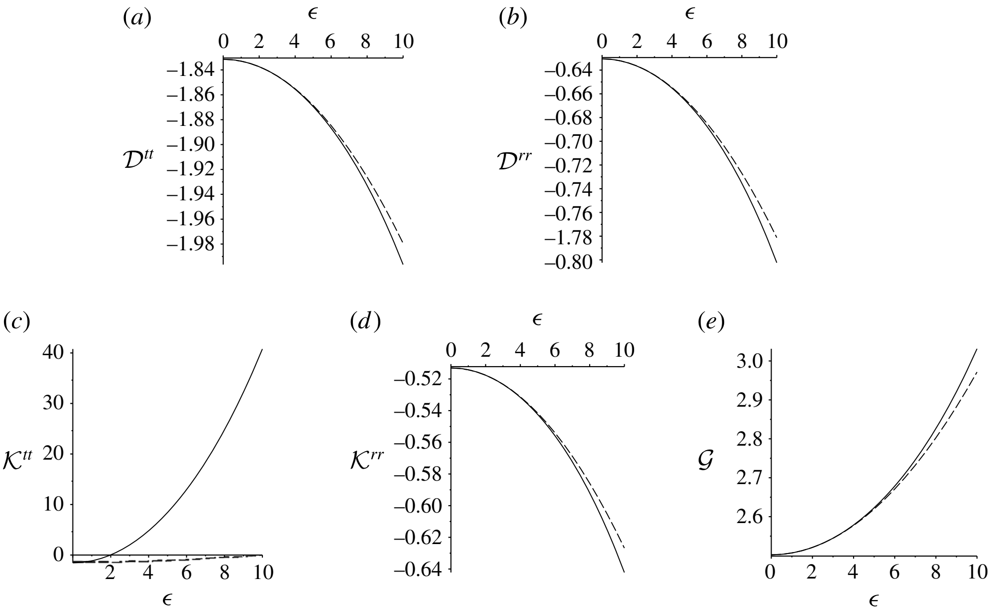

FIGURE 1. The dependencies of the first-order virial corrections to the self-diffusion coefficients $\mathcal{D}^{tt}$, $\mathcal{D}^{rr}$, the collective diffusion coefficients $\mathcal{K}^{tt}$, $\mathcal{K}^{rr}$ and the second-order virial corrections to the effective viscosity $\mathcal{G}$ on the energy of magnetic interactions between particles measured by the parameter $\epsilon = \mu_0 m^2/4\pi k_B T a^3$. Solid lines represent the numerically evaluated integrals in (2.6)–(2.8) and dashed lines represent the asymptotic dependencies for $\epsilon \ll 1$ provided in appendix B.

which, for moderate and large values of ϵ, makes the coefficient $\mathcal{K}^{tt}$ significantly larger than the other diffusion and viscosity coefficients. In fact, at $\epsilon \approx 10$ the virial correction to the coefficient of translational collective diffusion is of different sign and approximately 25 times greater in absolute value than its non-magnetic limit, whereas the absolute values of the virial corrections to $\mathcal{D}^{tt}$, $\mathcal{D}^{rr}$, $\mathcal{K}^{rr}$ and $\mathcal{G}$ are increased by approximately 10–30 %.

In the non-magnetic limit, $\epsilon = 0$, the short-time transport coefficients take the form (cf. (2.2*a*,*b*), (2.3) and (B 4*a*–*e*))

$$D^{tt} \approx D_0(1\text{–}1.8315\phi), \tag{2.24a}$$

$$D^{rr} \approx D_0(1\text{–}0.6305\phi), \tag{2.24b}$$

$$K^{tt} \approx 1\text{–}6.5464\phi, \tag{2.24c}$$

$$K^{rr} \approx 1\text{–}1.5131\phi, \tag{2.24d}$$

$$\eta_{eff} \approx \eta(1 + 2.5\phi + 5.0021\phi^2), \tag{2.24e}$$

which agrees very well with previous calculations of these coefficients first done by Batchelor & Green (1972) and Batchelor (1976) and then with higher precision due to inclusion of hydrodynamic interactions at higher orders by Cichocki & Felderhof (1988), Jones (1988), Wajnryb & Dahler (1997), Cichocki *et al.* (1999), Cichocki *et al.* (2002) and Cichocki *et al.* (2003).

It is also worth pointing out that, since the absolute values of the coefficients $\mathcal{D}^{qq}$, $\mathcal{K}^{qq}$ and $\mathcal{G}$ increase quickly with ϵ, the validity of the asymptotic expansion in the volume fraction ϕ may break down for large values of ϵ; in other words for $\epsilon \gg 1$ the two-particle corrections are no longer much smaller than unity.

3. The diffusion coefficients and viscosity in an external magnetic field

When the external magnetic field, denoted by $\boldsymbol{B}$, is switched on one must also include the energy of interaction of the magnetic particles with the field, $\boldsymbol{m}_i \cdot \boldsymbol{B}$, thus the energies of particle interactions with the magnetic field generated by other particles and the external magnetic field take the form (cf. (2.5))

$$\frac{E_{ij}}{k_B T} = \frac{\epsilon}{r_{ij}^3}[\hat{\boldsymbol{m}}_i \cdot \hat{\boldsymbol{m}}_j - 3(\hat{\boldsymbol{m}}_i \cdot \hat{\boldsymbol{r}}_{ij})(\hat{\boldsymbol{m}}_j \cdot \hat{\boldsymbol{r}}_{ij})], \tag{3.1}$$

$$\frac{E_{Bij}}{k_B T} = \mathscr{E}(\hat{\boldsymbol{m}}_i + \hat{\boldsymbol{m}}_j) \cdot \hat{\boldsymbol{B}}, \quad \mathscr{E} = \frac{mB}{k_B T}. \tag{3.2}$$

In such a case the diffusion coefficients and the effective viscosity are anisotropic. The self- and collective diffusion coefficients along the field $\boldsymbol{B}$ are different from the coefficients in the directions perpendicular to the external field; the former will be denoted by subscript $\|_{\boldsymbol{B}}$ and the latter by subscript $\perp_{\boldsymbol{B}}$. However, the effective viscosity in the presence of an external magnetic field has three different components associated with three different types of strain in the flow of the suspension with respect to the direction of the field $\boldsymbol{B}$. These are the axially symmetric strain along the field $\boldsymbol{B}$, which will be denoted by subscript $\|_{\boldsymbol{B}}$, strain at an angle $\pi/4$ with respect to the field $\boldsymbol{B}$, which will be denoted by subscript $\angle_{\boldsymbol{B}}$ and strain in the plane perpendicular to the field $\boldsymbol{B}$, which will be denoted by subscript $\perp_{\boldsymbol{B}}$. Assuming a coordinate system such that the z axis is aligned with the external field $\boldsymbol{B}$ and a flow at infinity in which the magnetic particles are immersed in linear form in the coordinates

$$\boldsymbol{v}_\infty = \boldsymbol{A}_\infty \cdot \boldsymbol{x}, \tag{3.3}$$

where $\boldsymbol{A}_\infty$ is a constant matrix, the three different types of strain flow are obtained by double contraction of the axial tensors $\boldsymbol{t}_0(\hat{\boldsymbol{e}})$, $\boldsymbol{t}_1(\hat{\boldsymbol{e}})$ and $\boldsymbol{t}_2(\hat{\boldsymbol{e}})$, where $\hat{\boldsymbol{e}}$ denotes the unit vector along the z axis (cf. appendix A and (2.12a), (2.12b)), with the symmetric part of the velocity gradient $\boldsymbol{A}^s_\infty$, i.e.

$$[\boldsymbol{v}_{\|_{\boldsymbol{B}}}]_\alpha = [\boldsymbol{t}_0(\hat{\boldsymbol{e}})]_{\alpha\beta\gamma\delta} A^s_{\infty\gamma\delta} x_\beta \quad \Rightarrow \quad \boldsymbol{v}_{\|_{\boldsymbol{B}}} = A^s_{\infty 11} \begin{bmatrix} -\frac{1}{2} & 0 & 0 \\ 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 1 \end{bmatrix} \begin{bmatrix} x \\ y \\ z \end{bmatrix}, \tag{3.4}$$

$$[\boldsymbol{v}_{\angle_{\boldsymbol{B}}}]_\alpha = [\boldsymbol{t}_1(\hat{\boldsymbol{e}})]_{\alpha\beta\gamma\delta} A^s_{\infty\gamma\delta} x_\beta \quad \Rightarrow \quad \boldsymbol{v}_{\angle_{\boldsymbol{B}}} = \begin{bmatrix} 0 & 0 & A^s_{\infty 13} \\ 0 & 0 & A^s_{\infty 23} \\ A^s_{\infty 13} & A^s_{\infty 23} & 0 \end{bmatrix} \begin{bmatrix} x \\ y \\ z \end{bmatrix}, \tag{3.5}$$

$$\begin{aligned} [\boldsymbol{v}_{\perp_{\boldsymbol{B}}}]_\alpha &= [\boldsymbol{t}_2(\hat{\boldsymbol{e}})]_{\alpha\beta\gamma\delta} A^s_{\infty\gamma\delta} x_\beta \\ &\Rightarrow \quad \boldsymbol{v}_{\perp_{\boldsymbol{B}}} = \begin{bmatrix} \frac{1}{2}(A^s_{\infty 11} - A^s_{\infty 22}) & A^s_{\infty 12} & 0 \\ A^s_{\infty 12} & -\frac{1}{2}(A^s_{\infty 11} - A^s_{\infty 22}) & 0 \\ 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} x \\ y \\ z \end{bmatrix}. \end{aligned} \tag{3.6}$$

The above types of flow are depicted in figure 2.

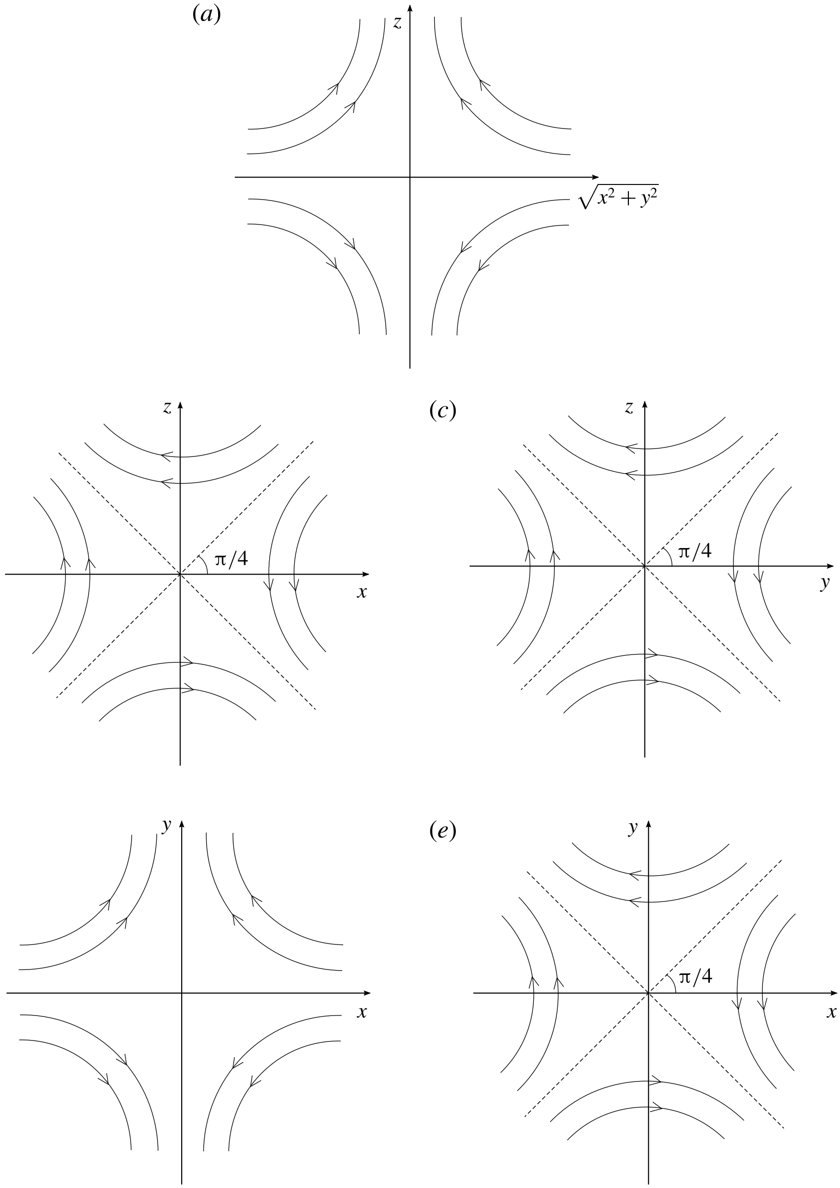

FIGURE 2. Three different types of strain flows $\boldsymbol{v}_{\parallel \boldsymbol{B}}$ (a), $\boldsymbol{v}_{\angle \boldsymbol{B}}$ (b,c) and $\boldsymbol{v}_{\perp \boldsymbol{B}}$ (d,e) associated with three different viscosity components. The arrows correspond to the choice of $A^s_{\infty 11} > 0$ for (a), $A^s_{\infty 13} < 0$ for (b), $A^s_{\infty 23} < 0$ for (c), $(A^s_{\infty 11} - A^s_{\infty 22})/2 < 0$ for (d) and $A^s_{\infty 12} < 0$ for (e).

At this stage it is important to point out that the term linear in concentration in the expression for the effective viscosity $\eta_{eff} = \eta[1 + 5/2\,\phi + (5/2 + \mathcal{G})\phi^2 + \cdots]$ comes from the single-particle hydrodynamic contribution to the stress on the particle. The particle rotation in the shear flow under the action of the external magnetic field is described by the mobility functions, which appear in the integrands of the presented formulae, and in a spherical geometry they do not depend on the external field. Since the particles are spherical and their hydrodynamic mobilities are independent of the orientation of the magnetic dipole moment, averaging over all possible orientations (with the normalized distribution function $(\mathscr{E}/\sinh\mathscr{E})^2\exp(-\boldsymbol{m}\cdot\boldsymbol{B}/k_BT)$) leads to the same result for the order-ϕ term as in the non-magnetic case, i.e. 5/2.

3.1. *Self- and collective diffusion coefficients*

3.1.1. *Parallel to* $\boldsymbol{B}$

When an external magnetic field $\boldsymbol{B}=B\hat{\boldsymbol{e}}$ is present, the parallel components of the virial corrections to the diffusion coefficients are given by the following sixfold integrals:

$$\mathcal{D}^{qq}_{\|\boldsymbol{B}} = \frac{3\zeta^{qq}}{32\pi^2}\left(\frac{\mathscr{E}}{\sinh\mathscr{E}}\right)^2\int_0^{\pi}\mathrm{d}\theta''\int_0^{\pi}\mathrm{d}\theta'\int_0^{2\pi}\mathrm{d}\varphi'\int_0^{\pi}\mathrm{d}\theta\int_0^{2\pi}\mathrm{d}\varphi\int_2^{\infty}\mathrm{d}r\,\sin\theta'' \times\sin\theta'\sin\theta\, r^2 d^{qq}_{\|\boldsymbol{B}}(r,\theta'')\exp\left[-\frac{E+E_B}{k_BT}\right], \tag{3.7}$$

$$\mathcal{K}^{qq}_{\|\boldsymbol{B}} = \frac{3\zeta^{qq}}{32\pi^2}\left(\frac{\mathscr{E}}{\sinh\mathscr{E}}\right)^2\int_0^{\pi}\mathrm{d}\theta''\int_0^{\pi}\mathrm{d}\theta'\int_0^{2\pi}\mathrm{d}\varphi'\int_0^{\pi}\mathrm{d}\theta\int_0^{2\pi}\mathrm{d}\varphi\int_2^{\infty}\mathrm{d}r\,\sin\theta'' \times\sin\theta'\sin\theta\, r^2\left\{k^{qq}_{\|\boldsymbol{B}}(r,\theta'')\exp\left[-\frac{E}{k_BT}\right]-\kappa^{qq}_{\|\boldsymbol{B}}(r,\theta'')\right\}\exp\left[-\frac{E_B}{k_BT}\right], \tag{3.8}$$

with

$$\frac{E}{k_BT} = \frac{\epsilon}{r^3}[\sin\theta'\sin\theta\cos\varphi'\cos\varphi(1-3\sin^2\theta'')+\sin\theta'\sin\theta\sin\varphi'\sin\varphi + \cos\theta'\cos\theta(1-3\cos^2\theta'')-3\sin\theta''\cos\theta''(\sin\theta\cos\theta'\cos\varphi + \cos\theta\sin\theta'\cos\varphi')], \tag{3.9}$$

$$\frac{E_B}{k_BT}=\mathscr{E}(\cos\theta'+\cos\theta) \tag{3.10}$$

where $(\mathscr{E}/\sinh\mathscr{E})^2$ in front of the integrals is a normalization factor associated with the interactions of particles with the external magnetic field and the hydrodynamic functions $d^{qq}_{\|\boldsymbol{B}}(r,\theta'')$ and $k^{qq}_{\|\boldsymbol{B}}(r,\theta'')$ are obtained by double contraction of the mobility matrix components (2.10) and (2.11) with the tensor $\hat{\boldsymbol{e}}\hat{\boldsymbol{e}}$ in the following way:

$$d^{qq}_{\|\boldsymbol{B}}(r,\theta'')=\left[\boldsymbol{\mu}^{qq}_{11}(\boldsymbol{r})-\frac{1}{\zeta^{qq}}\boldsymbol{I}\right]_{\alpha\beta}e_\beta e_\alpha=\frac{1}{\zeta^{qq}}\left[M^{qq}_{11\perp}(r)+(M^{qq}_{11\|}(r)-M^{qq}_{11\perp}(r))\frac{z^2}{r^2}\right], \tag{3.11}$$

$$k^{qq}_{\|\boldsymbol{B}}(r,\theta'')=\left[\boldsymbol{\mu}^{qq}_{11}(\boldsymbol{r})-\frac{1}{\zeta^{qq}}\boldsymbol{I}+\boldsymbol{\mu}^{qq}_{12}(\boldsymbol{r})\right]_{\alpha\beta}e_\beta e_\alpha = \frac{1}{\zeta^{qq}}\left[M^{qq}_{11\perp}(r)+M^{qq}_{12\perp}(r)+\sum_{n=1}^{3}\frac{b^{qq}_{\perp n}}{r^n} + \left(M^{qq}_{11\|}(r)+M^{qq}_{12\|}(r)-M^{qq}_{11\perp}(r)-M^{qq}_{12\perp}(r)+\sum_{n=1}^{3}\frac{b^{qq}_{\|n}-b^{qq}_{\perp n}}{r^n}\right)\frac{z^2}{r^2}\right], \tag{3.12}$$

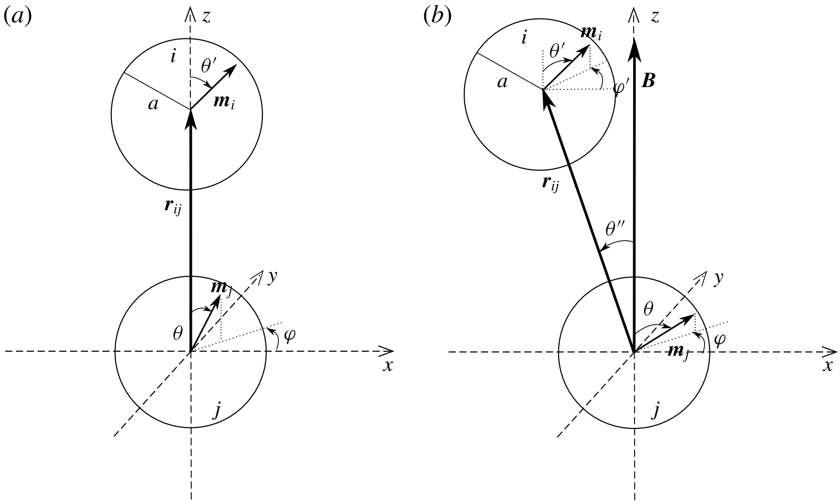

FIGURE 3. A schematic picture of the particles i, j and their magnetic dipole moments; the notation used for integration in the formulae for the virial corrections to the diffusion coefficients and the effective viscosity is also depicted.

$$\kappa_{\parallel \boldsymbol{B}}^{qq}(r,\theta'')=\frac{1}{\zeta^{qq}}[\boldsymbol{M}_{RP}^{qq}(\boldsymbol{r})]_{\alpha\beta}e_\beta e_\alpha=\frac{1}{\zeta^{qq}}\left[\sum_{n=1}^{3}\frac{b_{\perp n}^{qq}}{r^n}+\frac{z^2}{r^2}\sum_{n=1}^{3}\frac{b_{\parallel n}^{qq}-b_{\perp n}^{qq}}{r^n}\right], \tag{3.13}$$

$$\frac{z^2}{r^2}=\cos^2\theta''. \tag{3.14}$$

Furthermore θ, φ are the spherical angles associated with the magnetic dipole vector $\boldsymbol{m}_j$; θ', φ' are the spherical angles associated with $\boldsymbol{m}_i$; and θ'' is the angle between the magnetic field induction $\boldsymbol{B}$ and $\boldsymbol{r}_{ij}$ in the xz plane (see figure 3b).

3.1.2. *Perpendicular to* $\boldsymbol{B}$

With the notation introduced above the virial corrections to perpendicular diffusion coefficients are defined as follows

$$\mathcal{D}_{\perp \boldsymbol{B}}^{qq}=\frac{3\zeta^{qq}}{32\pi^2}\left(\frac{\mathscr{E}}{\sinh\mathscr{E}}\right)^2\int_0^{\pi}\mathrm{d}\theta''\int_0^{\pi}\mathrm{d}\theta'\int_0^{2\pi}\mathrm{d}\varphi'\int_0^{\pi}\mathrm{d}\theta\int_0^{2\pi}\mathrm{d}\varphi\int_2^{\infty}\mathrm{d}r\sin\theta'' \times\sin\theta'\sin\theta\, r^2 d_{\perp \boldsymbol{B}}^{qq}(r,\theta'')\exp\left[-\frac{E+E_B}{k_BT}\right], \tag{3.15}$$

$$\mathcal{K}_{\perp \boldsymbol{B}}^{qq}=\frac{3\zeta^{qq}}{32\pi^2}\left(\frac{\mathscr{E}}{\sinh\mathscr{E}}\right)^2\int_0^{\pi}\mathrm{d}\theta''\int_0^{\pi}\mathrm{d}\theta'\int_0^{2\pi}\mathrm{d}\varphi'\int_0^{\pi}\mathrm{d}\theta\int_0^{2\pi}\mathrm{d}\varphi\int_2^{\infty}\mathrm{d}r\sin\theta'' \times\sin\theta'\sin\theta\, r^2\left\{k_{\perp \boldsymbol{B}}^{qq}(r,\theta'')\exp\left[-\frac{E}{k_BT}\right]-\kappa_{\perp \boldsymbol{B}}^{qq}(r,\theta'')\right\}\exp\left[-\frac{E_B}{k_BT}\right], \tag{3.16}$$

where E/k_BT and E_B/k_BT are given in (3.9) and (3.10) and the hydrodynamic functions $d_{\perp \boldsymbol{B}}^{qq}(r,\theta'')$ and $k_{\perp \boldsymbol{B}}^{qq}(r,\theta'')$ are obtained by double contraction of the mobility matrix components (2.10) and (2.11) with the tensor $\boldsymbol{I}-\hat{\boldsymbol{e}}\hat{\boldsymbol{e}}$ in the following way:

$$\begin{aligned} d_{\perp_B}^{qq}(r,\theta'') &= \frac{1}{2}\left[\boldsymbol{\mu}_{11}^{qq}(\boldsymbol{r}) - \frac{1}{\zeta^{qq}}\boldsymbol{I}\right]_{\alpha\beta}(\delta_{\beta\alpha} - e_\beta e_\alpha) \\ &= \frac{1}{\zeta^{qq}}\left[M_{11\perp}^{qq}(r) + \frac{1}{2}(M_{11\parallel}^{qq}(r) - M_{11\perp}^{qq}(r))\sin^2\theta''\right], \end{aligned} \tag{3.17}$$

$$\begin{aligned} k_{\perp_B}^{qq}(r,\theta'') &= \frac{1}{2}\left[\boldsymbol{\mu}_{11}^{qq}(\boldsymbol{r}) - \frac{1}{\zeta^{qq}}\boldsymbol{I} + \boldsymbol{\mu}_{12}^{qq}(\boldsymbol{r})\right]_{\alpha\beta}(\delta_{\beta\alpha} - e_\beta e_\alpha) \\ &= \frac{1}{\zeta^{qq}}\left[M_{11\perp}^{qq}(r) + M_{12\perp}^{qq}(r) + \sum_{n=1}^{3}\frac{b_{\perp n}^{qq}}{r^n}\right. \\ &\quad \left. + \frac{1}{2}\left(M_{11\parallel}^{qq}(r) + M_{12\parallel}^{qq}(r) - M_{11\perp}^{qq}(r) - M_{12\perp}^{qq}(r) + \sum_{n=1}^{3}\frac{b_{\parallel n}^{qq} - b_{\perp n}^{qq}}{r^n}\right)\sin^2\theta''\right], \end{aligned} \tag{3.18}$$

$$\kappa_{\perp_B}^{qq}(r,\theta'') = \frac{1}{2\zeta^{qq}}[\boldsymbol{M}_{RP}^{qq}(\boldsymbol{r})]_{\alpha\beta}(\delta_{\beta\alpha} - e_\beta e_\alpha) = \frac{1}{\zeta^{qq}}\left[\sum_{n=1}^{3}\frac{b_{\perp n}^{qq}}{r^n} + \frac{1}{2}\sin^2\theta''\sum_{n=1}^{3}\frac{b_{\parallel n}^{qq} - b_{\perp n}^{qq}}{r^n}\right]. \tag{3.19}$$

In the limit of vanishing magnetic field the potential energy of magnetic interactions for a particle reduces to (2.5) and, since $\int_0^\pi \sin\theta''\cos^2\theta''\,\mathrm{d}\theta'' = 2/3$, $\int_0^\pi \sin^3\theta''\,\mathrm{d}\theta = 4/3$ and $\int_0^\pi \sin\theta''\,\mathrm{d}\theta = 2$, both the parallel (3.7), (3.8) and perpendicular (3.15), (3.16) coefficients become equal to the non-magnetic coefficients (2.6)–(2.8).

3.2. *Effective viscosity – three components*

The three components of the effective viscosity associated with different directions of strain with respect to the applied field are given by

$$\begin{aligned} \mathcal{G}_{\parallel_B} &= \frac{15\zeta^{dd}}{64\pi^2}\left(\frac{\mathscr{E}}{\sinh\mathscr{E}}\right)^2\int_0^\pi \mathrm{d}\theta''\int_0^\pi \mathrm{d}\theta'\int_0^{2\pi}\mathrm{d}\varphi'\int_0^\pi \mathrm{d}\theta\int_0^{2\pi}\mathrm{d}\varphi\int_2^\infty \mathrm{d}r\,\sin\theta'' \\ &\quad \times \sin\theta'\sin\theta\, r^2\left\{g_{\parallel_B}(r,\theta'')\exp\left[-\frac{E}{k_BT}\right] - \gamma_{\parallel_B}^{dd}(r,\theta'')\right\}\exp\left[-\frac{E_B}{k_BT}\right], \end{aligned} \tag{3.20}$$

$$\begin{aligned} \mathcal{G}_{\angle_B} &= \frac{15\zeta^{dd}}{64\pi^2}\left(\frac{\mathscr{E}}{\sinh\mathscr{E}}\right)^2\int_0^\pi \mathrm{d}\theta''\int_0^\pi \mathrm{d}\theta'\int_0^{2\pi}\mathrm{d}\varphi'\int_0^\pi \mathrm{d}\theta\int_0^{2\pi}\mathrm{d}\varphi\int_2^\infty \mathrm{d}r\,\sin\theta'' \\ &\quad \times \sin\theta'\sin\theta\, r^2\left\{g_{\angle_B}(r,\theta'')\exp\left[-\frac{E}{k_BT}\right] - \gamma_{\angle_B}^{dd}(r,\theta'')\right\}\exp\left[-\frac{E_B}{k_BT}\right], \end{aligned} \tag{3.21}$$

$$\begin{aligned} \mathcal{G}_{\perp_B} &= \frac{15\zeta^{dd}}{64\pi^2}\left(\frac{\mathscr{E}}{\sinh\mathscr{E}}\right)^2\int_0^\pi \mathrm{d}\theta''\int_0^\pi \mathrm{d}\theta'\int_0^{2\pi}\mathrm{d}\varphi'\int_0^\pi \mathrm{d}\theta\int_0^{2\pi}\mathrm{d}\varphi\int_2^\infty \mathrm{d}r\,\sin\theta'' \\ &\quad \times \sin\theta'\sin\theta\, r^2\left\{g_{\perp_B}(r,\theta'')\exp\left[-\frac{E}{k_BT}\right] - \gamma_{\perp_B}^{dd}(r,\theta'')\right\}\exp\left[-\frac{E_B}{k_BT}\right], \end{aligned} \tag{3.22}$$

where E/k_BT and E_B/k_BT are given in (3.9) and (3.10) and the hydrodynamic functions $g_\parallel(r,\theta'')$, $g_\angle(r,\theta'')$ and $g_\perp(r,\theta'')$ are obtained by double contraction of the dipolar mobility matrix components (2.12*a*) and (2.12*b*) with the tensors $\hat{\boldsymbol{t}}_0(\hat{\boldsymbol{e}})$, $\boldsymbol{t}_1(\hat{\boldsymbol{e}})$ and $\boldsymbol{t}_2(\hat{\boldsymbol{e}})$ (cf. appendix A) respectively in the following way:

$$\begin{aligned} g_{\parallel_{\boldsymbol{B}}}(r,\theta'') &= \left[\boldsymbol{\mu}^{dd}_{11}(\boldsymbol{r}) - \frac{1}{\zeta^{dd}}\boldsymbol{I} + \boldsymbol{\mu}^{dd}_{12}(\boldsymbol{r})\right]_{\alpha\beta\gamma\delta}[\boldsymbol{t}_0(\hat{\boldsymbol{e}})]_{\gamma\delta\beta\alpha} \\ &= \frac{1}{\zeta^{dd}}\left[\left(M^{dd}_{11\,0}(r) + M^{dd}_{12\,0}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,0n}}{r^n}\right)\left(\frac{9}{4}\cos^4\theta'' - \frac{3}{2}\cos^2\theta'' + \frac{1}{4}\right)\right. \\ &\quad + 3\left(M^{dd}_{11\,1}(r) + M^{dd}_{12\,1}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,1n}}{r^n}\right)(-\cos^4\theta'' + \cos^2\theta'') \\ &\quad \left. + 3\left(M^{dd}_{11\,2}(r) + M^{dd}_{12\,2}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,2n}}{r^n}\right)\left(\frac{1}{4}\cos^4\theta'' - \frac{1}{2}\cos^2\theta'' + \frac{1}{4}\right)\right], \end{aligned} \tag{3.23}$$

$$\begin{aligned} \gamma^{dd}_{\parallel_{\boldsymbol{B}}}(r,\theta'') &= \frac{1}{\zeta^{dd}}[\boldsymbol{M}^{dd}_{RP}(\boldsymbol{r})]_{\alpha\beta\gamma\delta}[\boldsymbol{t}_0(\hat{\boldsymbol{e}})]_{\gamma\delta\beta\alpha} \\ &= \frac{1}{\zeta^{dd}}\left[\left(\frac{9}{4}\cos^4\theta'' - \frac{3}{2}\cos^2\theta'' + \frac{1}{4}\right)\sum_{n=3}^{5}\frac{c^{dd}_{12\,0n}}{r^n}\right. \\ &\quad + 3(-\cos^4\theta'' + \cos^2\theta'')\sum_{n=3}^{5}\frac{c^{dd}_{12\,1n}}{r^n} \\ &\quad \left. + 3\left(\frac{1}{4}\cos^4\theta'' - \frac{1}{2}\cos^2\theta'' + \frac{1}{4}\right)\sum_{n=3}^{5}\frac{c^{dd}_{12\,2n}}{r^n}\right], \end{aligned} \tag{3.24}$$

$$\begin{aligned} g_{\angle_{\boldsymbol{B}}}(r,\theta'') &= \frac{1}{2}\left[\boldsymbol{\mu}^{dd}_{11}(\boldsymbol{r}) - \frac{1}{\zeta^{dd}}\boldsymbol{I} + \boldsymbol{\mu}^{dd}_{12}(\boldsymbol{r})\right]_{\alpha\beta\gamma\delta}[\boldsymbol{t}_1(\hat{\boldsymbol{e}})]_{\gamma\delta\beta\alpha} \\ &= \frac{1}{\zeta^{dd}}\left[\frac{3}{2}\left(M^{dd}_{11\,0}(r) + M^{dd}_{12\,0}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,0n}}{r^n}\right)(-\cos^4\theta'' + \cos^2\theta'')\right. \\ &\quad + \frac{1}{2}\left(M^{dd}_{11\,1}(r) + M^{dd}_{12\,1}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,1n}}{r^n}\right)(4\cos^4\theta'' - 3\cos^2\theta'' + 1) \\ &\quad \left. + \frac{1}{2}\left(M^{dd}_{11\,2}(r) + M^{dd}_{12\,2}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,2n}}{r^n}\right)(-\cos^4\theta'' + 1)\right], \end{aligned} \tag{3.25}$$

$$\begin{aligned} \gamma^{dd}_{\angle_{\boldsymbol{B}}}(r,\theta'') &= \frac{1}{\zeta^{dd}}[\boldsymbol{M}^{dd}_{RP}(\boldsymbol{r})]_{\alpha\beta\gamma\delta}[\boldsymbol{t}_1(\hat{\boldsymbol{e}})]_{\gamma\delta\beta\alpha} \\ &= \frac{1}{\zeta^{dd}}\left[\frac{3}{2}(-\cos^4\theta'' + \cos^2\theta'')\sum_{n=3}^{5}\frac{c^{dd}_{12\,0n}}{r^n} + \frac{1}{2}(4\cos^4\theta'' - 3\cos^2\theta'' + 1)\right. \\ &\quad \left. \times\sum_{n=3}^{5}\frac{c^{dd}_{12\,1n}}{r^n} + \frac{1}{2}(-\cos^4\theta'' + 1)\sum_{n=3}^{5}\frac{c^{dd}_{12\,2n}}{r^n}\right], \end{aligned} \tag{3.26}$$

$$
\begin{aligned}
g_{\perp_{\boldsymbol{B}}}(r,\theta'') &= \left[\boldsymbol{\mu}^{dd}_{11}(\boldsymbol{r}) - \frac{1}{\zeta^{dd}}\boldsymbol{I} + \boldsymbol{\mu}^{dd}_{12}(\boldsymbol{r})\right]_{\alpha\beta\gamma\delta}[\boldsymbol{t}_2(\hat{\boldsymbol{e}})]_{\gamma\delta\beta\alpha}\\
&= \frac{1}{\zeta^{dd}}\left[\frac{3}{8}\left(M^{dd}_{11\,0}(r) + M^{dd}_{12\,0}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,0n}}{r^n}\right)(\cos^4\theta'' - 2\cos^2\theta'' + 1)\right.\\
&\quad + \frac{1}{2}\left(M^{dd}_{11\,1}(r) + M^{dd}_{12\,1}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,1n}}{r^n}\right)(-\cos^4\theta'' + 1)\\
&\quad \left. + \frac{1}{8}\left(M^{dd}_{11\,2}(r) + M^{dd}_{12\,2}(r) + \sum_{n=3}^{5}\frac{c^{dd}_{12\,2n}}{r^n}\right)(\cos^4\theta'' + 6\cos^2\theta'' + 1)\right],
\end{aligned}
\tag{3.27}
$$

$$
\begin{aligned}
\gamma^{dd}_{\perp_{\boldsymbol{B}}}(r,\theta'') &= \frac{1}{\zeta^{dd}}[\boldsymbol{M}^{dd}_{RP}(\boldsymbol{r})]_{\alpha\beta\gamma\delta}[\boldsymbol{t}_2(\hat{\boldsymbol{e}})]_{\gamma\delta\beta\alpha}\\
&= \frac{1}{\zeta^{dd}}\left[\frac{3}{8}(\cos^4\theta'' - 2\cos^2\theta'' + 1)\sum_{n=3}^{5}\frac{c^{dd}_{12\,0n}}{r^n} + \frac{1}{2}(-\cos^4\theta'' + 1)\sum_{n=3}^{5}\frac{c^{dd}_{12\,1n}}{r^n}\right.\\
&\quad \left. + \frac{1}{8}(\cos^4\theta'' + 6\cos^2\theta'' + 1)\sum_{n=3}^{5}\frac{c^{dd}_{12\,2n}}{r^n}\right].
\end{aligned}
\tag{3.28}
$$

When the magnetic field vanishes and the potential energy of magnetic interactions for a particle reduces to (2.5), the integrals over θ, θ' and φ, φ' become elementary and, since $\int_0^{\pi}\sin\theta''\cos^4\theta''\,\mathrm{d}\theta'' = 2/5$, $\int_0^{\pi}\sin\theta''\cos^2\theta''\,\mathrm{d}\theta'' = 2/3$, $\int_0^{\pi}\sin^3\theta''\,\mathrm{d}\theta'' = 4/3$ and $\int_0^{\pi}\sin\theta''\,\mathrm{d}\theta'' = 2$, all three effective viscosity coefficients (3.20)–(3.22) become equal to the non-magnetic coefficient (2.8).

3.3. *Limit of weak interparticle interactions for ferrofluids (up to $O(\epsilon^4)$)*

To make analytical progress we will study now the limit of weak magnetic interparticle interactions, the strength of which is measured by

$$
\epsilon = \frac{\mu_0 m^2}{4\pi k_B T a^3} \ll 1,
\tag{3.29}
$$

a limit mostly relevant to magnetic suspensions of rather small magnetic nanoparticles. In principle, the strength of the interactions of particles with the magnetic field can be arbitrary; however, naturally we would assume that they dominate the interparticle interactions (but we emphasize that this assumption is not strictly necessary), i.e.

$$
\mathscr{E} = \frac{mB}{k_B T} \gg \epsilon \quad \Rightarrow \quad B \gg \frac{\mu_0 m}{4\pi a^3}.
\tag{3.30}
$$

Under these assumptions one may expand the exponential term

$$
\begin{aligned}
&\exp\left\{-\frac{\epsilon}{r^3}[\sin\theta'\sin\theta\cos\varphi'\cos\varphi(1 - 3\sin^2\theta'') + \sin\theta'\sin\theta\sin\varphi'\sin\varphi\right.\\
&\quad + \cos\theta'\cos\theta(1 - 3\cos^2\theta'') - 3\sin\theta''\cos\theta''(\sin\theta\cos\theta'\cos\varphi\\
&\quad \left. + \cos\theta\sin\theta'\cos\varphi')]\right\}
\end{aligned}
\tag{3.31}
$$

in powers of ϵ up to ϵ^2 and compute explicitly the integrals in (3.7), (3.8), (3.15), (3.16) and (3.20)–(3.22). Simple symmetry arguments allow one to show the odd-power corrections to be zero. Therefore, the virial corrections to the four self-diffusion coefficients take the asymptotic form

$$\mathcal{D}^{qq}_{\parallel_{\boldsymbol{B}}} = \mathcal{D}^{qq}_0 + \epsilon^2 \mathcal{D}^{qq}_{\parallel_{\boldsymbol{B}}2}(\mathscr{E}) + O(\epsilon^4), \tag{3.32}$$

$$\mathcal{D}^{qq}_{\perp_{\boldsymbol{B}}} = \mathcal{D}^{qq}_0 + \epsilon^2 \mathcal{D}^{qq}_{\perp_{\boldsymbol{B}}2}(\mathscr{E}) + O(\epsilon^4), \tag{3.33}$$

$$\mathcal{D}^{qq}_0 = \sum_{n=1}^{\infty} \frac{1}{2^{n-3}(n-3)}[a^{qq}_{\parallel n} + 2a^{qq}_{\perp n}], \tag{3.34}$$

$$\begin{aligned}\mathcal{D}^{qq}_{\parallel_{\boldsymbol{B}}2}(\mathscr{E}) &= \frac{1}{35} f(\mathscr{E})^2 \sum_{n=1}^{\infty} \frac{1}{2^{n+3}(n+3)}(29a^{qq}_{\parallel n} + 118a^{qq}_{\perp n}) \\ &\quad + \frac{2}{35}[1-2f(\mathscr{E})]^2 \sum_{n=1}^{\infty} \frac{1}{2^{n+3}(n+3)}(11a^{qq}_{\parallel n} + 10a^{qq}_{\perp n}) \\ &\quad + \frac{18}{35} f(\mathscr{E})[1-2f(\mathscr{E})] \sum_{n=1}^{\infty} \frac{1}{2^{n+3}(n+3)}(3a^{qq}_{\parallel n} + 4a^{qq}_{\perp n}),\end{aligned} \tag{3.35}$$

$$\begin{aligned}\mathcal{D}^{qq}_{\perp_{\boldsymbol{B}}2}(\mathscr{E}) &= \frac{1}{35} f(\mathscr{E})^2 \sum_{n=1}^{\infty} \frac{1}{2^{n+3}(n+3)}(59a^{qq}_{\parallel n} + 88a^{qq}_{\perp n}) \\ &\quad + \frac{2}{35}[1-2f(\mathscr{E})]^2 \sum_{n=1}^{\infty} \frac{1}{2^{n+3}(n+3)}[5a^{qq}_{\parallel n} + 16a^{qq}_{\perp n}] \\ &\quad + \frac{18}{35} f(\mathscr{E})[1-2f(\mathscr{E})] \sum_{n=1}^{\infty} \frac{1}{2^{n+3}(n+3)}(2a^{qq}_{\parallel n} + 5a^{qq}_{\perp n}),\end{aligned} \tag{3.36}$$

the virial corrections to the four collective diffusion coefficients in the asymptotic regime $\epsilon \ll 1$ are

$$\mathcal{K}^{qq}_{\parallel_{\boldsymbol{B}}} = \mathcal{K}^{qq}_0 + \epsilon^2 \mathcal{K}^{qq}_{\parallel_{\boldsymbol{B}}2}(\mathscr{E}) + O(\epsilon^4), \tag{3.37}$$

$$\mathcal{K}^{qq}_{\perp_{\boldsymbol{B}}} = \mathcal{K}^{qq}_0 + \epsilon^2 \mathcal{K}^{qq}_{\perp_{\boldsymbol{B}}2}(\mathscr{E}) + O(\epsilon^4), \tag{3.38}$$

$$\mathcal{K}^{qq}_0 = \sum_{n=4}^{\infty} \frac{1}{2^{n-3}(n-3)}[a^{qq}_{\parallel n} + b^{qq}_{\parallel n} + 2(a^{qq}_{\perp n} + b^{qq}_{\perp n})], \tag{3.39}$$

$$\begin{aligned}\mathcal{K}^{qq}_{\parallel_{\boldsymbol{B}}2}(\mathscr{E}) &= \frac{1}{35} f(\mathscr{E})^2 \sum_{n=1}^{\infty} \frac{1}{2^{n+3}(n+3)}[29(a^{qq}_{\parallel n} + b^{qq}_{\parallel n}) + 118(a^{qq}_{\perp n} + b^{qq}_{\perp n})] \\ &\quad + \frac{2}{35}[1-2f(\mathscr{E})]^2 \sum_{n=1}^{\infty} \frac{1}{2^{n+3}(n+3)}[11(a^{qq}_{\parallel n} + b^{qq}_{\parallel n}) + 10(a^{qq}_{\perp n} + b^{qq}_{\perp n})] \\ &\quad + \frac{18}{35} f(\mathscr{E})[1-2f(\mathscr{E})] \sum_{n=1}^{\infty} \frac{1}{2^{n+3}(n+3)}[3(a^{qq}_{\parallel n} + b^{qq}_{\parallel n}) + 4(a^{qq}_{\perp n} + b^{qq}_{\perp n})],\end{aligned} \tag{3.40}$$

$$\mathcal{K}^{qq}_{\perp_{\boldsymbol{B}}2}(\mathscr{E}) = \frac{1}{35}f(\mathscr{E})^2\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}[59(a^{qq}_{\parallel n}+b^{qq}_{\parallel n})+88(a^{qq}_{\perp n}+b^{qq}_{\perp n})]$$
$$+\frac{2}{35}[1-2f(\mathscr{E})]^2\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}[5(a^{qq}_{\parallel n}+b^{qq}_{\parallel n})+16(a^{qq}_{\perp n}+b^{qq}_{\perp n})]$$
$$+\frac{18}{35}f(\mathscr{E})[1-2f(\mathscr{E})]\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}[2(a^{qq}_{\parallel n}+b^{qq}_{\parallel n})+5(a^{qq}_{\perp n}+b^{qq}_{\perp n})], \quad (3.41)$$

and finally the virial corrections to the three effective viscosities for $\epsilon \ll 1$ take the form

$$\mathcal{G}_{\parallel_{\boldsymbol{B}}} = \mathcal{G}_0 + \epsilon^2\mathcal{G}_{\parallel_{\boldsymbol{B}}2}(\mathscr{E}) + O(\epsilon^4), \quad (3.42)$$
$$\mathcal{G}_{\angle_{\boldsymbol{B}}} = \mathcal{G}_0 + \epsilon^2\mathcal{G}_{\angle_{\boldsymbol{B}}2}(\mathscr{E}) + O(\epsilon^4), \quad (3.43)$$
$$\mathcal{G}_{\perp_{\boldsymbol{B}}} = \mathcal{G}_0 + \epsilon^2\mathcal{G}_{\perp_{\boldsymbol{B}}2}(\mathscr{E}) + O(\epsilon^4), \quad (3.44)$$

$$\mathcal{G}_0 = \frac{3}{2}\sum_{n=6}^{\infty}\frac{1}{2^{n-3}(n-3)}[c^{dd}_{11\,0n}+c^{dd}_{12\,0n}+2(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})+2(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})], \quad (3.45)$$

$$\mathcal{G}_{\parallel_{\boldsymbol{B}}2}(\mathscr{E}) = \frac{3}{14}f(\mathscr{E})^2\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}[9(c^{dd}_{11\,0n}+c^{dd}_{12\,0n})+14(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})$$
$$+26(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})]+\frac{3}{7}[1-2f(\mathscr{E})]^2\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}[3(c^{dd}_{11\,0n}+c^{dd}_{12\,0n})$$
$$+2(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})+2(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})]+\frac{3}{7}f(\mathscr{E})[1-2f(\mathscr{E})]\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}$$
$$\times[3(c^{dd}_{11\,0n}+c^{dd}_{12\,0n})+12(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})+6(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})], \quad (3.46)$$

$$\mathcal{G}_{\angle_{\boldsymbol{B}}2}(\mathscr{E}) = \frac{3}{14}f(\mathscr{E})^2\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}[7(c^{dd}_{11\,0n}+c^{dd}_{12\,0n})+20(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})$$
$$+22(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})]+\frac{3}{7}[1-2f(\mathscr{E})]^2\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}[c^{dd}_{11\,0n}+c^{dd}_{12\,0n}$$
$$+4(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})+2(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})]+\frac{3}{7}f(\mathscr{E})[1-2f(\mathscr{E})]\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}$$
$$\times[6(c^{dd}_{11\,0n}+c^{dd}_{12\,0n})+7(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})+8(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})], \quad (3.47)$$

$$\mathcal{G}_{\perp_{\boldsymbol{B}}2}(\mathscr{E}) = \frac{3}{14}f(\mathscr{E})^2\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}[13(c^{dd}_{11\,0n}+c^{dd}_{12\,0n})+22(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})$$
$$+14(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})]+\frac{3}{7}[1-2f(\mathscr{E})]^2\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}[c^{dd}_{11\,0n}+c^{dd}_{12\,0n}$$
$$+2(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})+4(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})]+\frac{3}{7}f(\mathscr{E})[1-2f(\mathscr{E})]\sum_{n=1}^{\infty}\frac{1}{2^{n+3}(n+3)}$$
$$\times[3(c^{dd}_{11\,0n}+c^{dd}_{12\,0n})+8(c^{dd}_{11\,1n}+c^{dd}_{12\,1n})+10(c^{dd}_{11\,2n}+c^{dd}_{12\,2n})], \quad (3.48)$$

where

$$f(\mathscr{E}) = \frac{\mathscr{E}\coth\mathscr{E} - 1}{\mathscr{E}^2}; \tag{3.49}$$

alternatively $f(\mathscr{E}) = L(\mathscr{E})/\mathscr{E}$ can be expressed by the Langevin function $L(\mathscr{E}) = (\mathscr{E}\coth\mathscr{E} - 1)/\mathscr{E}$. Note that the anisotropy associated with the presence of an external magnetic field does not appear at the leading order ϵ^0, but is evident at the order ϵ^2. The computation of the sums in the above equations leads to the final formulae for the virial corrections to the diffusion coefficients and the effective viscosity (cf. (2.2*a*,*b*), (2.3))

$$\mathcal{D}^{tt}_{\parallel_B} = -1.8315 - \epsilon^2[2.4742\times10^{-3} - 3.5237\times10^{-3}f(\mathscr{E}) + 1.6124\times10^{-3}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50a}$$

$$\mathcal{D}^{tt}_{\perp_B} = -1.8315 - \epsilon^2[1.4248\times10^{-3} - 9.0001\times10^{-4}f(\mathscr{E}) + 3.1866\times10^{-3}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50b}$$

$$\mathcal{D}^{rr}_{\parallel_B} = -0.6305 - \epsilon^2[1.3753\times10^{-3} - 7.2477\times10^{-4}f(\mathscr{E}) + 3.3641\times10^{-3}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50c}$$

$$\mathcal{D}^{rr}_{\perp_B} = -0.6305 - \epsilon^2[2.0259\times10^{-3} - 2.3512\times10^{-3}f(\mathscr{E}) + 2.3883\times10^{-3}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50d}$$

$$\mathcal{K}^{tt}_{\parallel_B} = -1.5464 + \epsilon^2[1.9169\times10^{-2} - 2.1331\times10^{-2}f(\mathscr{E}) + 2.4430\times10^{-2}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50e}$$

$$\mathcal{K}^{tt}_{\perp_B} = -1.5464 + \epsilon^2[1.7007\times10^{-2} - 1.5926\times10^{-2}f(\mathscr{E}) + 2.7673\times10^{-2}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50f}$$

$$\mathcal{K}^{rr}_{\parallel_B} = -0.5131 - \epsilon^2[1.4346\times10^{-4} + 1.6875\times10^{-3}f(\mathscr{E}) + 3.8772\times10^{-3}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50g}$$

$$\mathcal{K}^{rr}_{\perp_B} = -0.5131 - \epsilon^2[1.9745\times10^{-3} - 2.8900\times10^{-3}f(\mathscr{E}) + 1.1307\times10^{-3}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50h}$$

$$\mathcal{G}_{\parallel_B} = 2.5021 + \epsilon^2[1.6371\times10^{-2} - 5.7779\times10^{-2}f(\mathscr{E}) + 6.8250\times10^{-2}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50i}$$

$$\mathcal{G}_{\angle_B} = 2.5021 + \epsilon^2[2.5682\times10^{-3} + 1.8898\times10^{-2}f(\mathscr{E}) - 3.7556\times10^{-2}f(\mathscr{E})^2] + O(\epsilon^4), \tag{3.50j}$$

$$\mathcal{G}_{\perp_B} = 2.5021 + \epsilon^2[3.3301\times10^{-3} - 4.0920\times10^{-3}f(\mathscr{E}) + 2.4556\times10^{-2}f(\mathscr{E})^2] + O(\epsilon^4). \tag{3.50k}$$

The above transport coefficients are plotted in figure 4. To give an idea of the influence of magnetic field and magnetic interactions on the short-time transport coefficients, one could estimate the relative change in the coefficients at some significant values of ϵ and $\mathscr{E}$. We have seen, in the previous section, that the asymptotic formulae (B 4*a*–*e*) describing the effect of magnetic interactions between particles in the absence of an external magnetic field give a good approximation of the precise numerical results even up to $\epsilon = 10$. With the external magnetic field switched on, the ϵ^2 corrections to the transport coefficients are all analytic functions of $\mathscr{E}$ and, for fixed ϵ, of the same order or even smaller than the ϵ^2 corrections in (B 4*a*–*e*). Therefore, the asymptotic formulae (3.50*a*–*k*) are expected to provide a

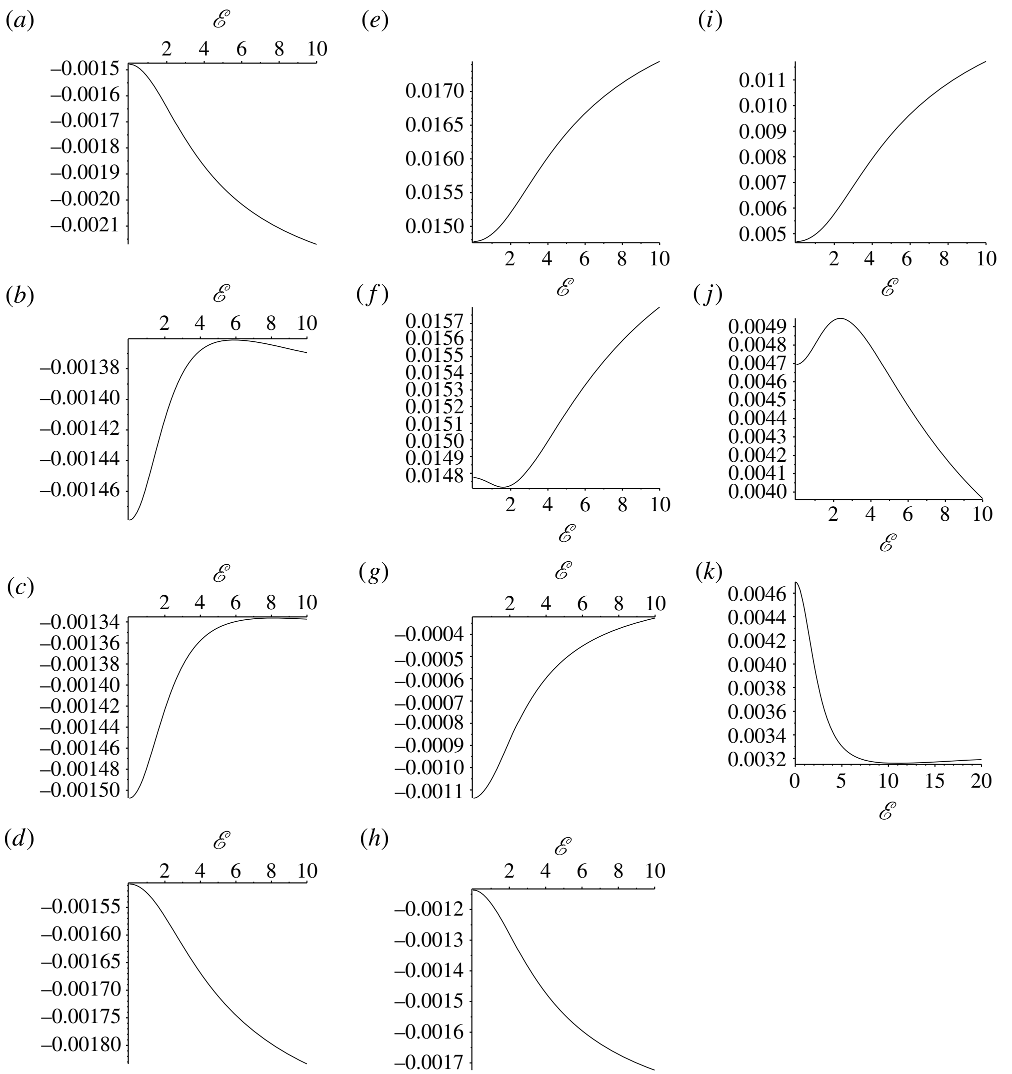

FIGURE 4. The dependence of the transport coefficients on the magnitude of the external magnetic field $\mathscr{E}$. (*a–d*) The corrections to self-diffusion $\mathcal{D}^{tt}_{\parallel_B 2}$, $\mathcal{D}^{tt}_{\perp_B 2}$ with maximum at $\mathscr{E} \approx 5.88$, $\mathcal{D}^{rr}_{\parallel_B 2}$ with maximum at $\mathscr{E} \approx 8.14$ and $\mathcal{D}^{rr}_{\perp_B 2}$. (*e–h*) The corrections to collective diffusion $\mathcal{K}^{tt}_{\parallel_B 2}$, $\mathcal{K}^{tt}_{\perp_B 2}$ with minimum at $\mathscr{E} \approx 1.61$, $\mathcal{K}^{rr}_{\parallel_B 2}$ and $\mathcal{K}^{rr}_{\perp_B 2}$. (*i–k*) The corrections to effective viscosity $\mathcal{G}_{\parallel_B 2}$, $\mathcal{G}_{\angle_B 2}$ with maximum at $\mathscr{E} \approx 2.36$ and $\mathcal{G}_{\perp_B 2}$ with minimum at $\mathscr{E} \approx 10.90$.

reasonable approximation for the actual values of the transport coefficients in a similar range of values of ϵ. It can be seen from figure 4, and the above formulae (3.50*a*–*k*), that at $\epsilon = 1$ the relative change in all the analysed short-time transport coefficients except for $\mathcal{K}^{tt}_{\parallel_B}$ and $\mathcal{K}^{tt}_{\perp_B}$ is rather small – approximately 0.1–2.5 ‰. However, as argued above, we can also provide a rough estimate of the transport coefficients at $\epsilon = 10$, when their relative change is between a few % and approximately 23 %, achieved for $\mathcal{G}_{\parallel_B}$. Note that from figure 4 it is evident that for higher values of $\mathscr{E}$ the relative change would be stronger. Similar to the previous section in the case of the translational collective diffusion coefficients $\mathcal{K}^{tt}_{\parallel_B}$ and $\mathcal{K}^{tt}_{\perp_B}$, the relative influence of the magnetic field and magnetic interactions should be much more significant and the asymptotic formulae (3.50*e*) and (3.50*f*) are expected to provide a reasonable approximation only for very small values of ϵ.

Furthermore, figure 4 clearly demonstrates non-monotonic behaviour of a number of short-time transport coefficients. In the case of the long-time effective viscosity, the mechanism described in a number of papers (e.g. the review by Odenbach 2004), based on the balance between the magnetic torque on a particle and mechanic torque exerted by a shearing flow, clearly leads to an increase of the effective long-time viscosity. However, at short times, or alternatively if the external field is alternating at high frequency, it is no longer obvious whether the effect of the magnetic field is to increase or decrease the viscosity, since the times are not long enough for the magnetic moments of the particles to align with the external field, and thus the orientations of the particles' magnetic moments are random. The final stresses and torques on the particles depend not only on their interactions with the external field but also strongly on the long-range and nonlinear hydrodynamic interactions, and it is those effects that contribute together to the observed non-monotonic behaviour.

Note that, in the limit of vanishing magnetic field,

$$f(\mathscr{E}) = \frac{\mathscr{E}\coth\mathscr{E} - 1}{\mathscr{E}^2} \xrightarrow{\mathscr{E}\to 0} \frac{1}{3}, \tag{3.51}$$

and the analytic expressions (3.32)–(3.48), and likewise the final numerical formulae (3.50*a*–*k*), reduce to the Taylor expansions for $\epsilon \ll 1$ of the coefficients obtained in § 3, $\mathcal{D}^{qq}_{\parallel_B} = \mathcal{D}^{qq}_{\perp_B} = \mathcal{D}^{qq}$, $\mathcal{K}^{qq}_{\parallel_B} = \mathcal{K}^{qq}_{\perp_B} = \mathcal{K}^{qq}$ and $\mathcal{G}_{\parallel_B} = \mathcal{G}_{\angle_B} = \mathcal{G}_{\perp_B} = \mathcal{G}$, which are provided in appendix B. On the other hand, in the limit of vanishing magnetic interactions, $\epsilon \to 0$, we recover the purely non-magnetic values $\mathcal{D}^{qq}_{\parallel_B} = \mathcal{D}^{qq}_{\perp_B} = \mathcal{D}^{qq}_0$, $\mathcal{K}^{qq}_{\parallel_B} = \mathcal{K}^{qq}_{\perp_B} = \mathcal{K}^{qq}_0$ and $\mathcal{G}_{\parallel_B} = \mathcal{G}_{\angle_B} = \mathcal{G}_{\perp_B} = \mathcal{G}_0$ (cf. Cichocki *et al.* 1999, 2002, 2003).

4. Conclusions

By using basic principles of statistical mechanics, explicit formulae have been derived for the virial corrections to the short-time self-diffusion and collective diffusion coefficients and effective viscosity for magnetic suspensions under the influence of magnetic interactions between particles and their interactions with the external magnetic field. In the virial expansion, the interactions between pairs of particles have been included and higher-order terms connected with multiparticle interactions have been neglected. The magnetic interactions between the particles and interactions of particles with the external magnetic field, with energies of interactions denoted by E and E_B respectively, are included via the introduction of the probabilistic factors $\exp(E/k_BT)$ and $\exp(E_B/k_BT)$. The first-order virial corrections to the self- and collective diffusion coefficients and second-order virial correction to the effective viscosity have been numerically evaluated for the case when no external magnetic field is present, and their dependence on the magnetic energy of interparticle interactions, ϵ, has been established; it was shown that all the diffusion coefficients and the viscosity are monotonic in ϵ, and their rate of variation increases with ϵ.

When the external magnetic field is switched on, the diffusion coefficients and the effective viscosity become anisotropic. Under the assumption of weak interparticle interactions, $\epsilon \ll 1$, all the components of the short-time translational and rotational self-diffusion $\mathcal{D}^{tt}_{\parallel_B}$, $\mathcal{D}^{tt}_{\perp_B}$, $\mathcal{D}^{rr}_{\parallel_B}$, $\mathcal{D}^{rr}_{\perp_B}$, the translational and rotational collective diffusion $\mathcal{K}^{tt}_{\parallel_B}$, $\mathcal{K}^{tt}_{\perp_B}$, $\mathcal{K}^{rr}_{\parallel_B}$, $\mathcal{K}^{rr}_{\perp_B}$ and effective viscosity $\mathcal{G}_{\parallel_B}$, $\mathcal{G}_{\angle_B}$, $\mathcal{G}_{\perp_B}$ were calculated up to $O(\epsilon^4)$. Their dependence on the energy of particle interaction with the external magnetic field $\mathscr{E}$ has been explicitly obtained and is provided in (3.50*a*–*k*).

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Appendix A. The fourth-order axial tensors

The tensors $\boldsymbol{t}_0(\hat{\boldsymbol{r}})$, $\boldsymbol{t}_1(\hat{\boldsymbol{r}})$ and $\boldsymbol{t}_2(\hat{\boldsymbol{r}})$ after Kim & Karrila (1991) can be expressed in the following form

$$t_{0\alpha\beta\gamma\delta}(\hat{\boldsymbol{r}}) = \tfrac{3}{2}(\hat{r}_\alpha\hat{r}_\beta - \tfrac{1}{3}\delta_{\alpha\beta})(\hat{r}_\gamma\hat{r}_\delta - \tfrac{1}{3}\delta_{\gamma\delta}), \tag{A 1}$$

$$t_{1\alpha\beta\gamma\delta}(\hat{\boldsymbol{r}}) = \tfrac{1}{2}(\delta_{\beta\delta}\hat{r}_\alpha\hat{r}_\gamma + \delta_{\alpha\delta}\hat{r}_\beta\hat{r}_\gamma + \delta_{\beta\gamma}\hat{r}_\alpha\hat{r}_\delta + \delta_{\alpha\gamma}\hat{r}_\beta\hat{r}_\delta - 4\hat{r}_\alpha\hat{r}_\beta\hat{r}_\gamma\hat{r}_\delta), \tag{A 2}$$

$$\begin{aligned} t_{2\alpha\beta\gamma\delta}(\hat{\boldsymbol{r}}) \;=\; & \tfrac{1}{2}\,(\delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\beta\gamma}\delta_{\alpha\delta} - \delta_{\alpha\beta}\delta_{\gamma\delta} + \delta_{\gamma\delta}\hat{r}_\alpha\hat{r}_\beta + \delta_{\alpha\beta}\hat{r}_\gamma\hat{r}_\delta, \\ & - \delta_{\beta\delta}\hat{r}_\alpha\hat{r}_\gamma - \delta_{\alpha\delta}\hat{r}_\beta\hat{r}_\gamma - \delta_{\beta\gamma}\hat{r}_\alpha\hat{r}_\delta - \delta_{\alpha\gamma}\hat{r}_\beta\hat{r}_\delta + \hat{r}_\alpha\hat{r}_\beta\hat{r}_\gamma\hat{r}_\delta). \end{aligned} \tag{A 3}$$

For any vectorial argument, denoted here by $\hat{\boldsymbol{r}}$, they satisfy the following identities

$$t_{0\alpha\beta\beta\alpha}(\hat{\boldsymbol{r}}) = 1, \quad t_{1\alpha\beta\beta\alpha}(\hat{\boldsymbol{r}}) = 2, \quad t_{2\alpha\beta\beta\alpha}(\hat{\boldsymbol{r}}) = 2, \tag{A 4a–c}$$

$$t_{0\alpha\beta\gamma\delta}(\hat{\boldsymbol{r}})t_{0\gamma\delta\zeta\eta}(\hat{\boldsymbol{r}}) = t_{0\alpha\beta\zeta\eta}(\hat{\boldsymbol{r}}), \quad t_{1\alpha\beta\gamma\delta}(\hat{\boldsymbol{r}})t_{1\gamma\delta\zeta\eta}(\hat{\boldsymbol{r}}) = t_{1\alpha\beta\zeta\eta}(\hat{\boldsymbol{r}}), \quad t_{2\alpha\beta\gamma\delta}(\hat{\boldsymbol{r}})t_{2\gamma\delta\zeta\eta}(\hat{\boldsymbol{r}}) = t_{2\alpha\beta\zeta\eta}(\hat{\boldsymbol{r}}), \tag{A 4d–f}$$

$$t_{0\alpha\beta\gamma\delta}(\hat{\boldsymbol{r}})t_{1\gamma\delta\zeta\eta}(\hat{\boldsymbol{r}}) = 0, \quad t_{0\alpha\beta\gamma\delta}(\hat{\boldsymbol{r}})t_{2\gamma\delta\zeta\eta}(\hat{\boldsymbol{r}}) = 0, \quad t_{1\alpha\beta\gamma\delta}(\hat{\boldsymbol{r}})t_{2\gamma\delta\zeta\eta}(\hat{\boldsymbol{r}}) = 0. \tag{A 4g–i}$$

Appendix B. The virial corrections to diffusion and viscosity coefficients in the absence of an external magnetic field under weak interparticle interactions

The virial corrections to the diffusion and viscosity coefficients in the absence of an external magnetic field ($\mathscr{E} \to 0$) and in the limit $\epsilon \ll 1$ take the form

$$\mathcal{D}^{qq} = \sum_{n=1}^{\infty}\left[\frac{1}{2^{n-3}(n-3)} + \frac{\epsilon^2}{3}\frac{1}{2^{n+3}(n+3)}\right](a^{qq}_{\parallel n} + 2a^{qq}_{\perp n}) + O(\epsilon^4), \tag{B 1}$$

$$\mathcal{K}^{qq} = \sum_{n=4}^{\infty}\left[\frac{1}{2^{n-3}(n-3)} + \frac{\epsilon^2}{3}\frac{1}{2^{n+3}(n+3)}\right][a^{qq}_{\parallel n} + b^{qq}_{\parallel n} + 2(a^{qq}_{\perp n} + b^{qq}_{\perp n})] + O(\epsilon^4), \tag{B 2}$$

$$\begin{aligned} \mathcal{G} \;=\; & \frac{3}{2}\sum_{n=6}^{\infty}\left[\frac{1}{2^{n-3}(n-3)} + \frac{\epsilon^2}{3}\frac{1}{2^{n+3}(n+3)}\right] \\ & \times [c^{dd}_{11\,0n} + c^{dd}_{12\,0n} + 2(c^{dd}_{11\,1n} + c^{dd}_{12\,1n}) + 2(c^{dd}_{11\,2n} + c^{dd}_{12\,2n})] + O(\epsilon^4), \end{aligned} \tag{B 3}$$

and thus their numerical values can be easily obtained as

$$\mathcal{D}^{tt} = -1.8315 - \epsilon^2 1.4788 \times 10^{-3} + O(\epsilon^4), \tag{B 4a}$$

$$\mathcal{D}^{rr} = -0.6305 - \epsilon^2 1.5075 \times 10^{-3} + O(\epsilon^4), \tag{B 4b}$$

$$\mathcal{K}^{tt} = -1.5464 + \epsilon^2 1.4773 \times 10^{-2} + O(\epsilon^4), \tag{B 4c}$$

$$\mathcal{K}^{rr} = -0.5131 - \epsilon^2 1.1368 \times 10^{-3} + O(\epsilon^4), \tag{B 4d}$$

$$\mathcal{G} = 2.5021 + \epsilon^2 4.6946 \times 10^{-3} + O(\epsilon^4). \tag{B 4e}$$

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