PHYSICAL REVIEW E **71**, 051407 (2005)

Anisotropic diffusion in nematic liquid crystals and in ferrofluids

Patrick Ilg*
Institut für Theoretische Physik, Technische Universität Berlin, Hardenbergstrasse 36, D-10623 Berlin, Germany

(Received 19 January 2005; revised manuscript received 15 February 2005; published 31 May 2005)

A unified, mean-field kinetic theory approach to the anisotropic translational diffusion observed in liquid crystals and in ferrofluids is proposed. In the dilute regime, unified expressions for the parallel as well as for the perpendicular diffusion coefficient in terms of orientational order parameters are found that apply for liquid crystals as well as for ferrofluids. This result explains the common origin of the anisotropic diffusion found in liquid crystals and in ferrofluids. Differences between the two liquids appear in the semi-dilute regime, where the diffusion coefficients depend on the specific interaction potentials. Explicit expressions for the diffusion coefficients are worked out also in this regime within a mean-field approximation. Comparisons with previous theoretical and experimental results are performed, showing satisfactory agreement to the present results.

DOI: 10.1103/PhysRevE.71.051407 PACS number(s): 82.70.Dd, 83.80.Xz, 05.20.Dd, 75.50.Mm

I. INTRODUCTION

Anisotropic translational diffusion has been observed in experimental investigations [1–3] and computer simulations [4,5] of nematic liquid crystals as well as in experiments on ferrofluids [6]. In liquid crystals, the anisotropy is described by the direction of the nematic order $\boldsymbol{n}$, while in ferrofluids $\boldsymbol{n}$ denotes the direction of the magnetic field. It is observed in both systems that the translational diffusion parallel to the director is enhanced while the diffusion in the perpendicular direction is decreased when compared to the isotropic values. While so far different models have been proposed in order to explain these phenomena separately, we here propose a unified approach based on a kinetic model. This approach has the additional benefit that the kinetic models considered here are also used in order to describe other dynamical and rheological properties. The predictions of the present approach are compared with those of the modified affine transformation model for liquid crystals [5] and with the mean-field model proposed in Ref. [6] to describe the anisotropic diffusion in ferrofluids found in their experiments.

Both kinds of fluids show additional anisotropic macroscopic properties like anisotropic viscosities, for example [7]. A phenomenological so-called modified affine transformation model has been proposed in order to account for anisotropic viscosities in liquid crystals [8] This model is also able to describe computer simulation results of anisotropic diffusion [5]. Results of recent experimental studies on the anisotropic diffusion in a thermotropic liquid crystal are well described by this model [1,2]. An earlier approach based on the hydrodynamic theory of isotropic liquids [9] was found to be incompatible with the experimental results in [1,2]. In the case of ferrofluids, the methods used to derive diffusion coefficients [6,10,11] have not directly been applied in order to predict also viscous behavior and vice versa.

A complementary approach to the dynamics of anisotropic fluids by kinetic models has proven to be very useful in order to provide a better microscopic understanding of the macroscopic phenomena. In the case of nematic liquid crystals, the kinetic model proposed independently by Hess [12] and Doi [13] is still widely used in order to describe various dynamical and rheological properties of liquid crystals. In ferrofluids, a similar role is played by the kinetic model proposed by Martsenyuk *et al.* [14] and later on generalized by Zubarev *et al.* [15]. In the present work, we propose a kinetic theory approach building upon these models in order to explain and describe the common origin and the differences of the anisotropic diffusion in liquid crystals and in ferrofluids.

This paper is organized as follows. In Sec. II the unified kinetic model for the dynamics of liquid crystals and ferrofluid is introduced. For dilute systems of nonmagnetic particles, expressions for the diffusion coefficients are derived in Sec. III. The corresponding diffusion coefficients are worked out for nematic liquid crystals in Sec. IV and for ferrofluids in Sec. V. It is shown that in certain limiting cases, the diffusion coefficients reduce to those derived in Sec. III. Finally, some conclusions are offered in Sec. VI.

II. KINETIC MODEL

The microstructure of both nematic liquid crystals and ferrofluids can be described by a vector $\boldsymbol{u}$ on the three-dimensional unit sphere. In the case of nematic liquid crystals, $\boldsymbol{u}$ denotes the orientation of the liquid crystal molecules, while in the case of ferrofluids $\boldsymbol{u}$ denotes the orientation of the magnetic moment of the ferromagnetic colloidal particles. In ferrofluids, two main relaxation mechanisms for the magnetic moment are known. In the Brownian relaxation, the particle as a whole rotates, while in the Néel relaxation the magnetic moment rotates within the particle. Following Ref. [14], we here employ the so-called rigid dipole approximation, i.e., we assume that the anisotropy constant of the magnetic material is sufficiently large that the moment remains parallel to the easy axes of the ferromagnetic particle. In this case, the unit vector $\boldsymbol{u}$ characterizes also the orientation of the ferromagnetic particle. In order to investigate translational diffusion, spatial inhomogeneities have to be included into the description. Let $f(\boldsymbol{r},\boldsymbol{u};t)$ denote the probability distribution function of finding a molecule (liquid

---

*Email address: ilg@physik.tu-berlin.de

crystal or ferromagnetic colloid) with orientation $\boldsymbol{u}$ at position $\boldsymbol{r}$ at time t. The orientational distribution function $\psi(\boldsymbol{u};t)$ is recovered as $\psi(\boldsymbol{u};t)=\int d^3r f(\boldsymbol{r},\boldsymbol{u};t)$, while the number density is computed by $\rho(\boldsymbol{r};t)=\int d^2u f(\boldsymbol{r},\boldsymbol{u};t)$. The normalization is chosen as $\int d^3r d^2u f(\boldsymbol{r},\boldsymbol{u};t)=N$, with N being the number of particles. In the sequel, we assume particles of ellipsoidal shape. Then, the translational and rotational dynamics under the influence of Brownian motion and the potential $U=V/k_{\mathrm{B}}T$ can be described by the following Fokker-Planck equation [16–18]

$$\frac{\partial}{\partial t}f=(L_{\mathrm{tr}}+L_{\mathrm{rot}})f, \tag{1}$$

where the translational and rotational parts read

$$L_{\mathrm{tr}}f=\boldsymbol{\nabla}\cdot\{D_{\parallel}^{0}\boldsymbol{uu}+D_{\perp}^{0}(\mathbf{1}-\boldsymbol{uu})\}\cdot(\boldsymbol{\nabla}f+f\boldsymbol{\nabla}U) \tag{2}$$

and

$$L_{\mathrm{rot}}f=D_r\boldsymbol{\mathcal{L}}\cdot(\boldsymbol{\mathcal{L}}f+f\boldsymbol{\mathcal{L}}U), \tag{3}$$

where $\boldsymbol{\nabla}=\partial/\partial\boldsymbol{r}$, $\boldsymbol{\mathcal{L}}=\boldsymbol{u}\times\partial/\partial\boldsymbol{u}$ denotes the rotational operator, D_r is the rotational diffusion coefficient, and $D_{\parallel}^{0}$, $D_{\perp}^{0}$ are the bare translational diffusion coefficients parallel and perpendicular to the axes of the molecule, respectively. For prolate ellipsoidal particles, one has $D_{\parallel}^{0}>D_{\perp}^{0}$, while the opposite sign holds for oblate ellipsoidal particles. For moderate axis ratios, recent molecular dynamics simulation results indicate that $D_{\parallel}$ and $D_{\perp}$ are proportional to the axis ratio Q [19]. For rodlike particles with large axes ratios $Q=a/b\gg 1$, where a and b denote the semi-axes, the diffusion coefficients $D_{\parallel}^{0}$ and $D_{\perp}^{0}$ are well approximated by $D_{\parallel}^{0}=2D_{\perp}^{0}=k_{\mathrm{B}}T\ln(a/b)/2\pi\eta_{\mathrm{s}}a$, where η_{s} is the viscosity of the solvent and k_{B} and T denote Boltzmann's constant and temperature, respectively [7]. For moderate axes ratios Q, useful expressions have been derived in Ref. [20] within the affine transformation model which predicts $D_{\perp}^{0}=Q^{-2/3}D_{\mathrm{sp}}$ and $D_{\parallel}^{0}=Q^{4/3}D_{\mathrm{sp}}$, where D_{sp} is the corresponding diffusion coefficient of spherical particles.

In ferrofluids, depending on details of the preparation conditions, the ferromagnetic particles are either more or less spherical (*i*) or show pronounced asphericity (*ii*), e.g., due to permanent clustering. In case (*ii*), the present approach is directly applicable. To deal with the first case, we follow Refs. [15,21] and define effective ellipsoidal particles representing temporary or permanent aggregates due to dipolar interactions. In the latter case, we assume that the axis ratio of the effective ellipsoidal particles varies only weakly with the magnetic field. Then, the axis ratio is determined solely by the value of the dipolar interaction. Cases (*i*) and (*ii*) will be considered in more detail in Secs. V B and V A, respectively.

The equilibrium distribution $f_0(\boldsymbol{r},\boldsymbol{u})=\exp(-U)/Z$, $Z=\int d^3r d^2u \exp(-U)$, is the stationary solution to the kinetic equation (1). In the sequel, we assume that the interaction potential is of the form

$$U(\boldsymbol{r},\boldsymbol{u})=-\boldsymbol{u}\cdot\boldsymbol{h}+U_1\boldsymbol{uu}:\overbracket{\boldsymbol{a}}, \tag{4}$$

where $\boldsymbol{h}=\mu\boldsymbol{H}/k_{\mathrm{B}}T$ is the reduced magnetic field, μ is the magnetic moment of the particle, and $\boldsymbol{a}=\int d^2u f(\boldsymbol{r},\boldsymbol{u};t)\overbracket{\boldsymbol{uu}}$ denotes the second rank orientational order parameter tensor with $\overbracket{\boldsymbol{a}}\equiv(\boldsymbol{a}+\boldsymbol{a}^{\mathrm{T}})/2-(1/3)\mathbf{1}$ the symmetric traceless part of the matrix $\boldsymbol{a}$ and $\mathbf{1}$ the unit matrix. For nematic liquid crystals, the magnetic moment μ is usually very small, so that the first term in Eq. (4) may be neglected. For ferrofluids, on the contrary, due to the large magnetic moment of the ferromagnetic particles, the magnetic field energy represented by the first term in Eq. (4) is significant. Usually, the contribution of the second term in Eq. (4) can be neglected compared with the first term, at least for moderate concentrations. To lowest order, magnetic dipolar interactions can be incorporated in a mean-field description by replacing the magnetic field $\boldsymbol{H}$ by an effective field [22]. Thus, the second term in Eq. (4) may be neglected in the case of ferrofluids. Equations (1)–(3) have been proposed to describe the dynamics of nematic liquid crystals. Here, we propose that these equations with the corresponding choice for the potential U are also valid for ferrofluids within the rigid dipole approximation. For a spatially homogeneous situation, L_{tr} does not contribute to the dynamics. In this case, Eqs. (1)–(3) reduce to the kinetic model of nematic liquid crystals proposed in Refs. [12,13] and to the kinetic model of ferrofluids introduced in Ref. [14,15], respectively. Here, spatially homogeneous external magnetic fields are considered. Due to Maxwell's relation $\boldsymbol{\nabla}\cdot\boldsymbol{H}+\boldsymbol{\nabla}\cdot\boldsymbol{M}=0$, the internal magnetic field $\boldsymbol{H}$ becomes spatially inhomogeneous only if the magnetization, due to concentration gradients, becomes spatially inhomogeneous.

For the case of nematic liquid crystals, Eqs. (1)–(4) with $h=0$ have been used in Ref. [23] in order to study the influence of the anisotropic diffusion coefficients on the Franck elasticity coefficients.

It should be stressed that the kinetic model defined by Eqs. (1)–(3) neglects hydrodynamic interactions between the particles. While hydrodynamic interactions are known to affect dynamical properties, the model defined above has been of great help to study various dynamical properties of nematic liquid crystals and ferrofluids [3,7]. Therefore, the present work can be useful to study the influence of orientational order and interaction potentials on the diffusion, leaving the effect of hydrodynamic interactions for future studies.

Integrating the kinetic equation (1) over all orientations $\boldsymbol{u}$ we arrive at the continuity equation

$$\frac{\partial}{\partial t}\rho(\boldsymbol{r};t)=\int d^2u(L_{\mathrm{tr}}+L_{\mathrm{rot}})f(\boldsymbol{r},\boldsymbol{u};t)=-\boldsymbol{\nabla}\cdot\boldsymbol{j}(\boldsymbol{r};t), \tag{5}$$

with the diffusion flow

$$\boldsymbol{j}(\boldsymbol{r},t)=-\int d^2u\{D_{\parallel}^{0}\boldsymbol{uu}+D_{\perp}^{0}(\mathbf{1}-\boldsymbol{uu})\}\cdot[\boldsymbol{\nabla}f(\boldsymbol{r},\boldsymbol{u};t) + f(\boldsymbol{r},\boldsymbol{u};t)\boldsymbol{\nabla}U(\boldsymbol{r},\boldsymbol{u})]. \tag{6}$$

Note, that although the rotational operator L_{rot} does not contribute to the diffusion flow, the coupling of translational and rotational motion appears in $\boldsymbol{j}$ via the diffusion tensor (for $D_{\parallel}^{0} \neq D_{\perp}^{0}$) and the interaction potential.

If we define the distribution function $\varphi(\boldsymbol{r},\boldsymbol{u};t)$ by $f(\boldsymbol{r},\boldsymbol{u};t)=\rho(\boldsymbol{r};t)\varphi(\boldsymbol{r},\mathbf{u};t)$, the normalization $\int d^2 u f = \rho$ requires $\int d^2 u \varphi \equiv \langle 1 \rangle = 1$. Then, the diffusion flow (6) can be written as

$$\boldsymbol{j}(\boldsymbol{r};t) = \boldsymbol{j}^0(\boldsymbol{r};t) + \boldsymbol{j}^S(\boldsymbol{r};t) + \boldsymbol{j}^U(\boldsymbol{r};t) \tag{7}$$

with

$$\boldsymbol{j}^0 = -D_{\perp}^0 \, \boldsymbol{\nabla} \rho - (D_{\parallel}^0 - D_{\perp}^0)\langle \boldsymbol{uu} \rangle \cdot \boldsymbol{\nabla} \rho, \tag{8}$$

the contribution from spatially inhomogeneous ordering

$$\boldsymbol{j}^S = -(D_{\parallel}^0 - D_{\perp}^0)\rho \boldsymbol{\nabla} \cdot \langle \boldsymbol{uu} \rangle, \tag{9}$$

and the contribution from the interaction potential

$$\boldsymbol{j}^U = -D_{\perp}^0 \rho \langle \boldsymbol{\nabla} U \rangle - (D_{\parallel}^0 - D_{\perp}^0)\rho \langle \boldsymbol{uu} \cdot \boldsymbol{\nabla} U \rangle. \tag{10}$$

Evaluation of the diffusion coefficients via the diffusion flow thus requires the calculation of the moments appearing in Eqs. (8)–(10).

The equilibrium distribution φ_0 is uniaxially symmetric with respect to the director $\boldsymbol{n}$. Uniaxial distribution functions can be represented by

$$\varphi_{\text{uni}}(\boldsymbol{r},\boldsymbol{u}\cdot\boldsymbol{n};t) = \sum_{j=0}^{\infty} (2j+1) S_j(\boldsymbol{r};t) P_j(\boldsymbol{u}\cdot\boldsymbol{n}), \tag{11}$$

where $P_j(x)$ denotes the Legendre polynomial of order j and S_j are the orientational order parameters, $-1/2 \leqslant S_j \leqslant 1$. For liquid crystals, only even terms appear in Eq. (11) due to their head-tail symmetry. For ferrofluids, this symmetry is broken by the magnetic moments of the particles. Consequently, all terms appear in Eq. (11). The implications of uniaxial symmetry on equilibrium and nonequilibrium properties of liquid crystals and ferrofluids have been considered, e.g., in Refs. [21,24,25].

III. DILUTE SUSPENSIONS OF ANISOTROPIC COLLOIDAL PARTICLES

For dilute systems, the interaction contribution to the mean field potential U, Eq. (4), can be neglected. Furthermore, if we consider nonmagnetic particles, the contribution of the magnetic field to the potential U also vanishes. Therefore, no contribution of the interaction potential to the diffusion flow appears, $\boldsymbol{j}^U = \boldsymbol{0}$. For a spatially homogeneous ordering with uniaxial symmetry, i.e., $S_j(\boldsymbol{r})=S_j$ and $\boldsymbol{n}(\boldsymbol{r})=\boldsymbol{n}$ are independent of $\boldsymbol{r}$, also $\boldsymbol{j}^S$ vanishes. Equation (8) can be evaluated to give

$$\boldsymbol{j} = \boldsymbol{j}^0 = -D_{\perp}(\boldsymbol{1} - \boldsymbol{nn}) \cdot \boldsymbol{\nabla}\rho - D_{\parallel} \boldsymbol{nn} \cdot \boldsymbol{\nabla}\rho, \tag{12}$$

where the effective diffusion coefficients are given by

$$D_{\perp} = D_{\perp}^0 + \frac{1 - S_2}{3}(D_{\parallel}^0 - D_{\perp}^0) \tag{13}$$

and

$$D_{\parallel} = D_{\parallel}^0 - \tfrac{2}{3}(1 - S_2)(D_{\parallel}^0 - D_{\perp}^0). \tag{14}$$

Equations (13) and (14) are our main result in this section. As will be shown later, these expressions hold not only for dilute nematic liquid crystals but also for very dilute ferrofluids, if both are subjected to Brownian motion in a spatially homogeneous potential. In the case of perfect orientational order we have $S_2=1$. In this case, we find from Eqs. (13) and (14) $D_{\perp}=D_{\perp}^0$, $D_{\parallel}=D_{\parallel}^0$, i.e., the effective diffusion coefficients reduce to their bare values. The average diffusion coefficient is defined by

$$\overline{D} \equiv \tfrac{1}{3}(D_{\parallel} + 2D_{\perp}). \tag{15}$$

In the present case, the average diffusion coefficient

$$\overline{D} = \tfrac{1}{3}(D_{\parallel}^0 + 2D_{\perp}^0) = D^0 \tag{16}$$

is found to be constant, independent of the degree of orientational order. In the presence of orientational ordering, the difference of the expressions (13) and (14) reflects the anisotropy of the translational diffusion,

$$D_{\perp} - D^0 = -\tfrac{1}{3}(D_{\parallel}^0 - D_{\perp}^0)S_2 \tag{17}$$

and

$$D_{\parallel} - D^0 = \tfrac{2}{3}(D_{\parallel}^0 - D_{\perp}^0)S_2. \tag{18}$$

In qualitative agreement with experimental results, the diffusion perpendicular to the director $\boldsymbol{n}$ is decreased while it is enhanced parallel to $\boldsymbol{n}$ when compared to the isotropic value, the anisotropy being more pronounced the stronger is the orientational ordering.

Following Ref. [5], we define the ratio

$$R \equiv \frac{D_{\parallel} - D_{\perp}}{D_{\parallel} + 2D_{\perp}} \tag{19}$$

in order to characterize the anisotropy of diffusion. The value for the perfectly ordered liquid is $R^0=(D_{\parallel}^0 - D_{\perp}^0)/(D_{\parallel}^0 + 2D_{\perp}^0)$. In the present case we find

$$R = R^0 S_2. \tag{20}$$

A linear dependence of R on S_2 is confirmed by computer simulations of hard ellipsoids [5]. The result (16) for the average diffusion coefficient is in disagreement with experiments [1] and simulation results [5]. This discrepancy is due to interactions between the particles as will be demonstrated in the next section.

IV. DIFFUSION IN NEMATIC LIQUID CRYSTALS

The kinetic model of rotational dynamics of nonmagnetic, elongated particles, Eqs. (1) and (3), was proposed independently by Hess [12] and Doi [13]. These equations provide a very useful model system for nematic liquid crystals that has been studied extensively [3,7]. The mean-field potential (4) with $\boldsymbol{h}=\boldsymbol{0}$ is frequently chosen as the Maier-Saupe potential, where the strength of the interaction U_1 is proportional to the local density ρ [3,7]. Excluded volume interactions of rod-like particles are more accurately described by the Onsager potential [7]. Further manipulations with the Onsager potential are hindered by its complicated structure. However, systematic approximations have been suggested in [26] which lead to more tractable potentials. The lowest order approximation to the Onsager potential is of the form (4) with

$$U_1 = \Phi(1 - (3/2)\boldsymbol{a}:\boldsymbol{a})^{-1/2} \tag{21}$$

depending on the reduced excluded volume Φ and the orientational order parameter $\boldsymbol{a}$. In the uniaxial regime, where Eq. (11) applies, $\boldsymbol{a}=S_2\overline{\boldsymbol{nn}}$ and the Onsager potential becomes $U_1=\Phi(1-S_2^2)^{-1/2}$. For rodlike particles, the reduced excluded volume is $\Phi=2b\ell^2\rho$, where $2b$ and ℓ are the diameter and length of the particles. The following results hold for arbitrary potentials of the form $U_1(\rho,\boldsymbol{a})$. Inserting Eq. (4) into (6) and performing the integration over the orientations, the diffusion flow contains second- and fourth-order moments. Assuming again a spatially homogeneous uniaxial nematic order, the diffusion flow $\boldsymbol{j}=\boldsymbol{j}^0+\boldsymbol{j}^U$ is of the form (12) with the effective diffusion coefficients

$$D_\perp = D^0_\perp(1+\tilde{U}_1S_2^2) + (D^0_\parallel - D^0_\perp)\left(\frac{1-S_2}{3} + \tilde{U}_1S_2\Sigma_\perp\right) \tag{22}$$

and

$$D_\parallel = D^0_\perp(1+\tilde{U}_1S_2^2) + (D^0_\parallel - D^0_\perp)\left(\frac{1+2S_2}{3} + \tilde{U}_1S_2\Sigma_\parallel\right), \tag{23}$$

where $\tilde{U}_1=\frac{2}{3}\rho(\partial U_1/\partial\rho)$, $\Sigma_\perp=(-7+25S_2-18S_4)/105$, and $\Sigma_\parallel=(14+55S_2+36S_4)/105$. Some details of the derivation are provided in Appendix A. For the Maier-Saupe potential and the potential (21), the strength of the potential U_1 is proportional to the density ρ. Thus, it follows that $\tilde{U}_1=\frac{2}{3}U_1$, which depends on the density and, in the case of the potential (21), also on the alignment tensor $\boldsymbol{a}$.

In the limit of vanishing interactions, $U_1\to 0$, Eqs. (22) and (23) coincide with Eqs. (13) and (14). From Eqs. (22) and (23), the average diffusion constant $\overline{D}$ is computed as

$$\overline{D} = D^0(1+\tilde{U}_1S_2^2). \tag{24}$$

From Eq. (24) we observe that, due to interactions, the average diffusion coefficient is enhanced with increasing nematic order. This result is in qualitative agreement with experimental results [1] and computer simulations [4].

A quantitative comparison of Eq. (24) to computer simulations of hard ellipsoidal particles [5] can be done. For highly elongated ellipsoids with axis ratio $Q\gtrsim 5$, the excluded volume interaction is well described by the Onsager potential. Figure 1 shows Eq. (24) with the mean field potential (21) together with the results of molecular dynamics simulations of elongated hard ellipsoidal particles [5]. For a constant value $\Phi=2$ of the reduced excluded volume, we observe good agreement with the simulation results for the largest axis ratio $Q=10$.

In Ref. [5], the modified affine transformation model was proposed to explain the simulation results. Within this model, the average diffusion coefficient is predicted to be given by $\overline{D}=D^0(1+2BS_2)^{-1/3}(1-BS_2)^{-2/3}$, where $B=(Q^2-1)/(Q^2+2)$. Although the functional form is different, the two expressions lead to similar results for highly elongated ellipsoids, as demonstrated in Fig. 1.

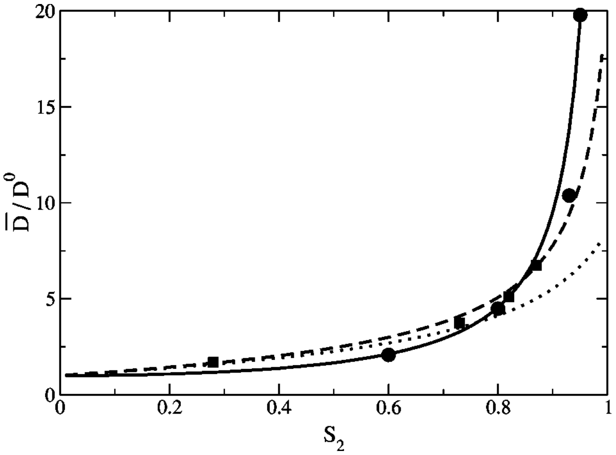

FIG. 1. The average diffusion coefficient $\overline{D}$ of hard ellipsoids as a function of the orientational order parameter S_2. Circles and squares are data from Ref. [5], corresponding to axis ratios $Q=10$ and $Q=5$, respectively. The dashed lines are the predictions of the affine transformation model; the solid line is the result (24) with $\Phi=2$.

From Eqs. (22) and (23), the ratio R, defined in Eq. (19) is given by

$$R = R^0\frac{1+\tilde{U}_1(\Sigma_\parallel-\Sigma_\perp)}{1+\tilde{U}_1S_2^2}S_2. \tag{25}$$

In the dilute limit, $\tilde{U}_1\to 0$, the result (19) is recovered from Eq. (25). For a quantitative comparison to experiments or simulation results, one has to supply the missing information on S_4. Ehrentraut and Hess have estimated $S_4\approx S_2[1-(1-S_2)^\nu]$, with $\nu=0.6$ on the basis of an ansatz for the orientational distribution function [8]. Figure 2 shows R as a function of S_2 with this ansatz for S_4 and a comparison to the modified affine transformation model and simulation results of [5]. Again, satisfactory agreement of the present results with computer simulations is found for the largest axis ratio $Q=10$. The modified affine transformation model [5] predicts a linear dependence of R on the orientational order parameter, $R=BS_2$, whereas the present result Eq. (25) shows a more complicated dependence. Interestingly, R given by Eq. (25) varies almost linearly with S_2 for low and high orientational ordering (see Fig. 2). Deviations from the linear behavior occur for intermediate ordering, which are in accordance with the simulation results. Thus, the present results are similar to the predictions of the modified affine transformation model. The variations of $\overline{D}$ and R on the orientational order parameter predicted by the modified affine transformation model have recently been confirmed in experimental results on a thermotropic liquid crystal [1]. Within the decoupling approximation, the order parameter S_2 has been calcu-

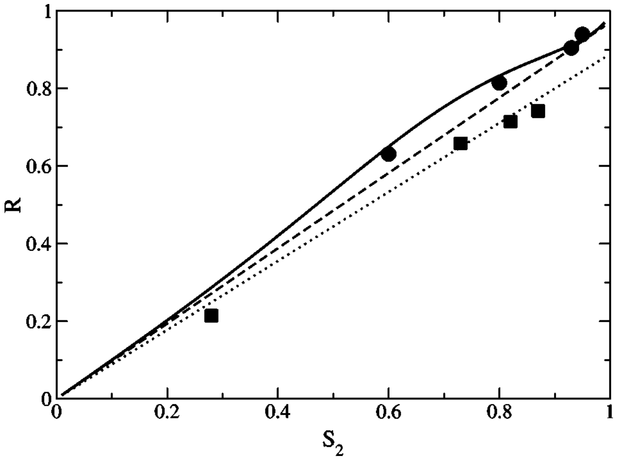

FIG. 2. The normalized anisotropy of diffusion R of hard ellipsoids as a function of the orientational order parameter S_2. Circles and squares are data from Ref. [5], corresponding to axis ratios $Q=10$ and $Q=5$, respectively. The dashed lines are the predictions of the affine transformation model; the solid line is the result (25) with $\Phi=2$.

lated as a function of the reduced excluded volume Φ for the Maier-Saupe and the potential (21) in [26].

V. DIFFUSION IN MODERATELY CONCENTRATED FERROFLUIDS

In ferrofluids, the magnetic colloidal particles interact with an internal magnetic field $\boldsymbol{H}$. The dimensionless interaction energy is given by the first term on the right-hand side of Eq. (4), where $h=\mu|\boldsymbol{H}|/k_BT$ denotes the Langevin parameter. For spatially homogeneous systems, the kinetic model (1)–(3) with the potential $U=-h\boldsymbol{u}\cdot\boldsymbol{n}$ has been proposed in Ref. [14] to describe the dynamics of dilute ferrofluids. Here and in the sequel, $\boldsymbol{n}=\boldsymbol{H}/|\boldsymbol{H}|$ denotes the direction of the internal magnetic field.

Dipolar interactions can been incorporated into the potential U within a mean-field approach by replacing the magnetic field by an effective field $h\rightarrow\xi_e$. Different mean-field models have been proposed in the literature [22]. A common feature of these mean-field models is that the lowest order correction to the magnetic field is proportional to the Langevin susceptibility $\chi_L=8\lambda\phi$, where $\phi=\rho v$ is the volume fraction and v is the volume of the ferromagnetic particles. The dipolar interactions parameter is defined as $\lambda=\mu^2/(4\pi\mu_0k_BTd^3)$, where d denotes the diameter of the magnetic particles.

For these systems, the equilibrium magnetization is given by $\boldsymbol{M}=M_{\text{sat}}\langle\boldsymbol{u}\rangle=M_{\text{sat}}S_1\boldsymbol{n}$, where $M_{\text{sat}}=\rho\mu$ is the saturation magnetization. In the local equilibrium approximation, the orientational order parameter is given explicitly as $S_1=L_1(\xi_e)$ with $L_1(x)=\coth(x)-x^{-1}$ the Langevin function.

Contrary to the case of liquid crystals, steric interactions in ferrofluids are not accounted for explicitly in this section. In the spirit of [14,15], we here focus on the regime where these interactions can either be neglected or be accounted for by modified diffusion coefficient D^0. Within an approximate treatment presented in [11], hydrodynamic and steric interactions can indeed be accounted for by a modified diffusion coefficient D^0. In either case, no explicit contribution to the potential U, given by Eq. (4), appears.

As discussed in Sec. II it is helpful to distinguish between (*i*) ferrofluids composed of rather spherical particles and (*ii*) those showing pronounced asphericity of the individual magnetic colloidal particles.

A. Weakly interacting, anisotropic ferromagnetic particles

Consider first case (*ii*), a dilute suspension of anisotropic, ferromagnetic particles with weak dipolar interactions. In this case, $D^0_\parallel$ and $D^0_\perp$ are the bare diffusion coefficients of the individual anisotropic particles parallel and perpendicular to the magnetic moment. Remember that we assume magnetically hard particles, where the magnetic moment remains fixed within the particle.

Under these assumptions, the diffusion flow (6) can be evaluated from Eqs. (8)–(10). Details of the derivation are provided in Appendix B. The result for the effective diffusion coefficients is

$$D_\perp=\overline{D}-\tfrac{1}{3}(D^0_\parallel-D^0_\perp)L_2(\xi_e)c(\xi_e), \tag{26}$$

$$D_\parallel=\overline{D}+\tfrac{2}{3}(D^0_\parallel-D^0_\perp)L_2(\xi_e)c(\xi_e), \tag{27}$$

where we have defined $c(\xi_e)=1-\rho(\partial\xi_e/\partial\rho)L_1(\xi_e)$. Note that the orientational order parameter S_2 can be evaluated explicitly in the local equilibrium approximation, $S_2=L_2(\xi_e)$ with $L_2(x)=1-3L_1(x)/x$. The average diffusion coefficient $\overline{D}=(D_\parallel+2D_\perp)/3$ in Eqs. (26) and (27) is given by

$$\overline{D}=D^0c(\xi_e). \tag{28}$$

From Eqs. (26) and (27), the anisotropy of diffusion R, defined in Eq. (19), is found to be given by $R=R^0L_2(\xi_e)$, where $R^0=(D^0_\parallel-D^0_\perp)/(D^0_\parallel+2D^0_\perp)$. Since the function $L_2(x)$ is bounded, $0\leqslant L_2\leqslant 1$, the anisotropy R is smaller than the bare value R^0. Note that by definition $D_\parallel-\overline{D}=-2(D_\perp-\overline{D})$ holds. It is interesting to note that the diffusion coefficients (26) and (27) differ from the corresponding expressions for a dilute suspension of anisotropic, nonmagnetic particles (17) and (18) by a multiplicative factor $c(\xi_e)$.

Since $L_2(0)=0$ one finds from Eqs. (26) and (27) that the diffusion is isotropic in the absence of a magnetic field, $D_\perp(h=0)=D_\parallel(h=0)=D^0$. For very strong magnetic fields, L_1, $L_2\rightarrow 1$ and we find from Eqs. (26) and (27) that the infinite field limit of the diffusion coefficients is given by $D_\perp(\infty)=D^0_\perp c(\infty)$ and $D_\parallel(\infty)=D^0_\parallel c(\infty)$, with the coefficient $c(\infty)=1-\rho(\partial\xi_e/\partial\rho)$.

Further analysis requires explicit knowledge of the effective field ξ_e and thus depends on the special mean-field model considered. In the dilute limit and for weak dipolar interactions, the Langevin susceptibility $\chi_L=8\lambda\phi$ is small. In this case, the effective field reduces to the magnetic field $\xi_e\approx h$. Note that due to Maxwell's equation $\boldsymbol{\nabla}\cdot\boldsymbol{H}+\boldsymbol{\nabla}\cdot\boldsymbol{M}=0$, the magnetic field is also concentration dependent. In this case we find $c(h)=1+3\chi_LL_1(h)/(1+3\chi_LL_1'(h))$. Since $c>1$

we observe that the average diffusion coefficient is enhanced compared to the bare value, $\overline{D} > D^0$. The analogous result was obtained in Sec. IV for the case of nematic liquid crystals. In the infinite field limit, $h \rightarrow \infty$, we find $c(\infty) = 1 + 3\chi_L > 1$ and therefore both the diffusion parallel and perpendicular to the applied field are enhanced compared to the bare values.

In the very dilute limit and for weak dipolar interactions where the Langevin parameter becomes very small, $\chi_L \ll 1$, the function $c(h)$ is well approximated by $c(h) \approx 1$. In this case, Eqs. (26) and (27) reduce to Eqs. (13) and (14) with the orientational order parameter $S_2 = L_2(h)$. The latter equations have been derived for a dilute suspension of nonmagnetic particles. In this regime, where interactions between the clusters are unimportant, the magnetic field merely provides the orientational order. Otherwise, the diffusion properties of the magnetic and nonmagnetic suspensions are identical.

B. Dilute suspension of clusters

In case (*i*), where the individual magnetic particles can be considered spherical, the present approach can still be used, if we follow the approach of the chain model proposed in Ref. [15]. Within this model, it is assumed that dipolar interactions lead to temporary or permanent clustering of the ferromagnetic particles, such that the dynamics can be described by a dilute suspension of effective magnetic particles of ellipsoidal shape. Under these assumptions, the present model applies not only in case (*ii*) but also in case (*i*), where the individual magnetic particles have an anisotropic shape. Thus, Eqs. (26) and (27) remain valid also in this case. A similar approach was taken in [21], where nonequilibrium dynamical properties of magnetic suspensions of ellipsoidal particles were studied.

Note, however, that the parameters of the model now correspond to the effective particles representing the clusters, not the individual spherical particles. The diffusion coefficients $D_\parallel^0 = D_\parallel^0(n)$ and $D_\perp^0 = D_\perp^0(n)$ now denote the bare diffusion coefficients of the effective, *n*-particle cluster parallel and perpendicular to its axis of symmetry. Within the chain model [15], the clusters are assumed to be chainlike with an ellipsoidal shape. Thus, for an *n*-particle cluster, the axis ratio Q of the ellipsoid is $Q = n$. If the affine transformation model [20] is employed, the dependencies $D_\parallel^0(n) = n^{-2/3} D_{\text{sp}}$ and $D_\perp^0(n) = n^{4/3} D_{\text{sp}}$ are predicted, where D_{sp} denotes the bare, single particle diffusion coefficient of an individual, spherical particle. Further, the effective magnetic moment of an *n*-particle cluster is *n* times bigger than that of an individual particle. Therefore, the Langevin parameter of the cluster is $h_n = n h_1$, where h_1 is the value for the single particle. Similarly, λ_n and ϕ_n denote the effective dipolar interaction parameter and the effective volume fraction of the *n*-particle clusters, respectively, and $\chi_L(n) = 8\lambda_n \phi_n$ denotes the effective Langevin parameter.

It should be emphasized that cluster formation in general leads to a broad distribution of cluster sizes [15]. Let g_n denote the number of *n*-particle clusters per unit volume of the system. An explicit expression for g_n was derived in [15] by minimizing an appropriate free energy function. The cluster size distribution is found to depend strongly on the magnetic field and the dipolar interaction strength.

Within the chain model [15], the clusters are assumed to be noninteracting. Therefore, Eqs. (26) and (27) apply for clusters of a fixed size *n*, where the effective field ξ_e is replaced by h_n. Let $[\bullet]_n$ denote the average over the cluster size distributions. Therefore, in a suspension of clusters of different sizes, the average diffusion coefficient $[\overline{D}]_n = [D^0(n) c(h_n)]_n$ would be measured. The average diffusion coefficients perpendicular and parallel to the magnetic field are given by $[D_\perp]_n = [\overline{D}]_n - (1/3)\Delta$ and $[D_\parallel]_n = [\overline{D}]_n + (2/3)\Delta$, where $\Delta = [(D_\parallel^0 - D_\perp^0) L_2(h_n) c(h_n)]_n$ denotes the difference of the parallel and perpendicular diffusion coefficients. Except for special cases, the averages over the cluster size distribution cannot be performed analytically.

C. Comparison to previous theoretical results

The anisotropic translational diffusion coefficients of dilute ferrofluids have been calculated within the mean spherical model by Morozov in [11]. The influence of steric and dipolar interactions on the average diffusion coefficient was found to be given by $\overline{D} = D_{\text{sp}}[1 + 1.45\phi - (4/3)\pi\lambda\chi_L]$. Thus, $\overline{D}$ is found to be independent of the magnetic field strength. In [11], the diffusion coefficients perpendicular and parallel to the magnetic field are given by $D_\perp = \overline{D} - (1/3) a L_1^2(h)$ and $D_\parallel = \overline{D} + (2/3) a L_1^2(h)$, respectively, with $a = 13.38\pi\chi_L D_{\text{sp}}$.

Contrary to the results of [11], here the average diffusion coefficient $\overline{D}$ is found to depend on the magnetic field [see Eq. (28)]. Only in the very dilute regime, where the Langevin susceptibility becomes very small, this field dependence becomes negligible. If we therefore compare the results of [11] to those of the present model in the very dilute regime, we find that the expressions for $D_\perp$ and $D_\parallel$ in [11] are of the form (17) and (18) if $D_\parallel^0 - D_\perp^0 = a$ denotes the difference $D_\parallel - D_\perp$ for infinite field and $L_2(x)$ is approximated by $L_1^2(x)$. The latter two functions are indeed very similar.

D. Comparison to experimental results

Finally, we compare the predictions made above to the experimental results obtained in [6]. For the ferrofluid used in these experiments, a value of $\chi_L \approx 1.4$ is reported in [6]. According to the chain model [15], the effect of dipolar interactions is taken into account by considering ideal solutions of effective, chainlike particles of ellipsoidal shape. In the present case, due to the moderate value of χ_L, moderate axis ratios of the chainlike clusters are expected. Thus, the cluster size distribution is expected to be rather narrow, such that the averages over the size distribution of the diffusion coefficients can be approximated by their values for the average cluster size. Using the results (17) and (18) obtained for dilute suspensions, the anisotropic diffusion coefficients are predicted to scale as $D_{\parallel,\perp} - D^0 \propto S_2$ with $S_2 = L_2(h)$, while the average diffusion coefficient $\overline{D}$ should be constant, independent of the magnetic field [see Eq. (16)].

Figure 3 shows the normalized average diffusion coefficient $\overline{D}/D^0$ as a function of the Langevin parameter *h*.

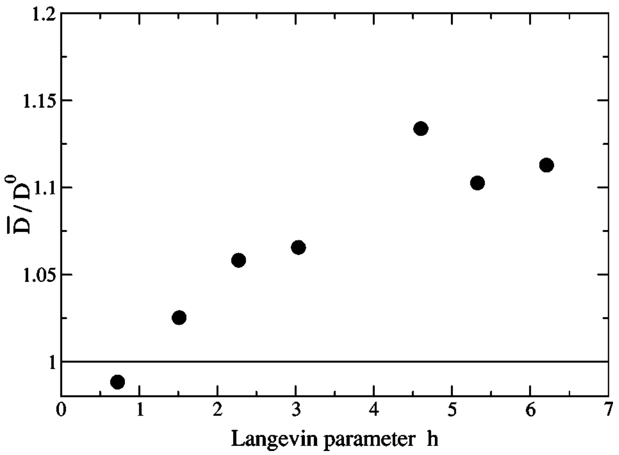

FIG. 3. The normalized average diffusion coefficient $\overline{D}/D^0$ is shown as a function of the Langevin parameter h. Data are taken from Ref. [6]. The solid line is the prediction for noninteracting, chainlike particles.

Within 10%, the average diffusion coefficient remains constant. Upon closer inspection, a slight increase of $\overline{D}$ with h is observed. This increase can be explained by dipolar interactions between the chainlike particles. In that case, $\overline{D}$ is given by Eq. (28), where c has to be evaluated with the effective parameters of the chainlike particles.

Figure 4 shows the reduced diffusion coefficients $(D_\parallel - D^0)/D^0$ and $(D_\perp - D^0)/D^0$ as a function of the Langevin parameter h. Figure 4 shows that the experimental data follow nicely the predicted behavior $D_{\parallel,\perp} - D^0 \propto L_2$. Small corrections to this behavior due to interactions of the clusters as indicated by the results shown in Fig. 3 can be included via Eqs. (26) and (27) using the effective parameters for the cluster particles. Note that a similar agreement of the experimental results to model predictions has been obtained in [6] by adjusting an additional fit parameter.

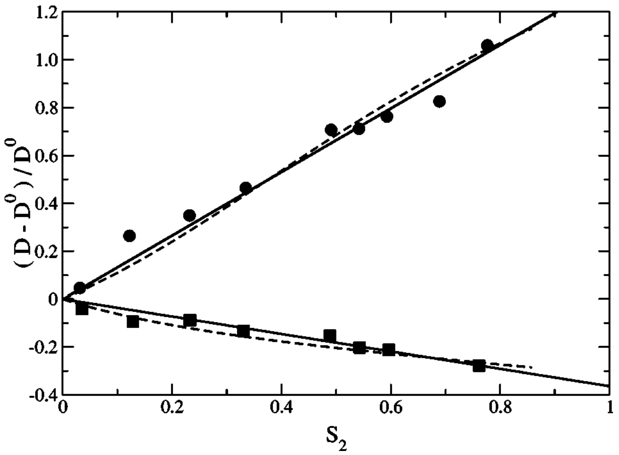

FIG. 4. The reduced diffusion coefficients $(D_\perp - D^0)/D^0$ (squares) and $(D_\parallel - D^0)/D^0$ (circles) as a function of the order parameter S_2 defined in the text. Data are taken from Ref. [6]. The dashed lines correspond to the fit presented in Ref. [6], while the solid lines are the result of a linear relation proposed in this work, assuming noninteracting, chainlike particles.

VI. CONCLUSIONS

A unified approach to the anisotropic diffusion in liquid crystals and in ferrofluids is proposed. Based on a mean-field kinetic equation, explicit expressions for the diffusion coefficients parallel and perpendicular to the director and magnetic field have been derived.

In the dilute limit, common expressions for the diffusion coefficients are found for both, liquid crystals and ferrofluids, showing the common origin of the phenomenon in both liquids. In both kinds of fluids, the anisotropy of the particle or cluster shape provides the basic mechanism for the anisotropic diffusion. For higher concentrations, when the specific interparticle interactions become important, different dependences of the diffusion coefficients on the orientational order parameters are found. This difference is directly related to the difference of the excluded volume and dipolar interactions, which dominate the dynamics in liquid crystals and in ferrofluids, respectively.

The present results for liquid crystals are similar to the predictions of the modified affine transformation model [5], while the results for ferrofluids are similar to those of Bacri [6] and Morozov [11]. Further comparisons with computer simulations and experimental results show satisfactory agreement. Except for elucidating the common origin of the anisotropic diffusion in both kinds of fluids, the main benefit of the present approach is that the same kinetic models that are used to predict viscous properties and the orientational dynamics can now also be used to predict diffusion properties. It would be very interesting to see whether the available kinetic models can describe all the dynamical properties of these fluids with the same set of material parameters.

ACKNOWLEDGMENTS

It is a pleasure to thank Siegfried Hess for valuable discussions. Helpful comments of two referees are also gratefully acknowledged. This work was supported in part by DFG-SPP 1104 “Kolloidale magnetische Systeme,” Grant No. HE1104/03.

APPENDIX A: DERIVATION OF DIFFUSION COEFFICIENTS FOR NEMATIC LIQUID CRYSTALS

In this appendix, the derivation of the anisotropic diffusion coefficients (22) and (23) is outlined.

The contribution of the diffusion flow $\boldsymbol{j}^U$ due to the mean-field interaction potential U is given by Eq. (10). For nematic liquid crystals, we consider potentials of the form $U = U_1(\rho)\boldsymbol{uu}:\langle\overline{\boldsymbol{uu}}\rangle$, where the strength of the interaction depends on the local concentration. We assume a spatially homogeneous uniaxial ordering, where the distribution function is of the form given in Eq. (11) with $S_j(\boldsymbol{r}) = S_j$.

The diffusion flow $\boldsymbol{j}^U$ contains two moments that have to be evaluated. First, $\langle\boldsymbol{\nabla}U\rangle = (\langle\boldsymbol{uu}\rangle:\langle\overline{\boldsymbol{uu}}\rangle)\boldsymbol{\nabla}U_1$, where $\boldsymbol{\nabla}U_1 = (\partial U_1/\partial\rho)\boldsymbol{\nabla}\rho$. With Eq. (11), the second moment is evaluated to give $\langle\boldsymbol{uu}\rangle = S_2\boldsymbol{nn} + [(1-S_2)/3]\mathbf{1}$. Therefore, one finds $\langle\boldsymbol{\nabla}U\rangle = \frac{2}{3}S_2^2\boldsymbol{\nabla}U_1$.

In order to evaluate also the term $\langle \boldsymbol{uu}\cdot\boldsymbol{\nabla} U\rangle$, the fourth moment is needed,

$$\langle u_\alpha u_\beta u_\gamma u_\delta\rangle = S_4 n_\alpha n_\beta n_\gamma n_\delta + \frac{S_2 - S_4}{7} n_{(\alpha}\, n_\beta \delta_{\ \gamma\delta)} + C_4 \delta_{(\alpha\beta}\ \delta_{\ \gamma\delta)}, \tag{A1}$$

where $C_4=(7-10S_2+3S_4)/105$ and $A_{(\alpha\beta\gamma\delta)}$ denotes the sum over the symmetrization of the expression [24]. With the help of Eq. (A1), the term $\langle \boldsymbol{uu}\cdot\boldsymbol{\nabla} U\rangle$ can be evaluated to give

$$\langle \boldsymbol{uu}\cdot\boldsymbol{\nabla} U\rangle = (\widetilde{U}_1/\rho)\Sigma_\perp(\mathbf{1}-\boldsymbol{nn})\cdot\boldsymbol{\nabla}\rho + (\widetilde{U}_1/\rho)\Sigma_\parallel \boldsymbol{nn}\cdot\boldsymbol{\nabla}\rho \tag{A2}$$

where $\widetilde{U}_1$ and $\Sigma_{\parallel,\perp}$ are defined after Eq. (22). With these results, the diffusion flow $\boldsymbol{j}=\boldsymbol{j}^0+\boldsymbol{j}^U$ can be expressed as $\boldsymbol{j}=-D_\perp(\mathbf{1}-\boldsymbol{nn})\cdot\rho+D_\parallel \boldsymbol{nn}\cdot\boldsymbol{\nabla}\rho$, where the diffusion coefficients $D_\perp$, $D_\parallel$ are given by Eqs. (22) and (23).

APPENDIX B: DERIVATION OF DIFFUSION COEFFICIENTS FOR FERROFLUIDS

Here, we briefly outline the derivation of the anisotropic diffusion coefficients (27) and (26).

The diffusion flow $\boldsymbol{j}^S$ is given by Eq. (9). In the local equilibrium approximation, the second moment $\langle u_\alpha u_\beta\rangle$ is given by

$$\langle u_\alpha u_\beta\rangle = L_2(\xi_e)\hat{H}_\alpha\hat{H}_\beta + \frac{1-L_2(\xi_e)}{3}\delta_{\alpha\beta}, \tag{B1}$$

where ξ_e is the local effective field. Using Eq. (B1), the diffusion flow $\boldsymbol{j}^S$ is given by

$$\boldsymbol{j}^S = -(D_\parallel^0 - D_\perp^0)\rho\frac{\partial\xi_e}{\partial\rho}\left(L_2'\hat{\mathbf{H}}\hat{\mathbf{H}}\cdot\boldsymbol{\nabla}\rho - \frac{1}{3}L_2'\,\boldsymbol{\nabla}\,\rho\right) \tag{B2}$$

where $L_2'\equiv \mathrm{d}L_2(\xi_e)/\mathrm{d}\xi_e$ denotes the total derivative. For the ease of notation, the argument ξ_e of the functions L_j is suppressed here. From Eq. (B2), the diffusion flow $\boldsymbol{j}^S$ can be written in the form $\boldsymbol{j}^S=-D_\perp^S(\mathbf{1}-\hat{\boldsymbol{H}}\hat{\boldsymbol{H}})\cdot\boldsymbol{\nabla}\rho-D_\parallel^S\boldsymbol{\nabla}\rho$, where the diffusion coefficients can be read off,

$$D_\perp^S = -\frac{1}{3}(D_\parallel^0 - D_\perp^0)\rho\frac{\partial\xi_e}{\partial\rho}L_2', \tag{B3}$$

$$D_\parallel^S = \frac{2}{3}(D_\parallel^0 - D_\perp^0)\rho\frac{\partial\xi_e}{\partial\rho}L_2'. \tag{B4}$$

Next, we consider the potential contribution of the diffusion flow $\boldsymbol{j}^U$, defined in Eq. (10). Within the local equilibrium approximation, the interaction potential is given by $U=-\xi_e\boldsymbol{u}\cdot\hat{\boldsymbol{H}}$. The gradient of the potential is therefore given by $\boldsymbol{\nabla}U=-(\boldsymbol{u}\cdot\hat{\boldsymbol{H}})\partial\xi_e/\partial\rho\boldsymbol{\nabla}\rho$. For the diffusion flow $\boldsymbol{j}^U$, the moments $\langle\boldsymbol{\nabla}U\rangle$ and $\langle \boldsymbol{uu}\cdot\boldsymbol{\nabla} U\rangle$ have to be evaluated. In the local equilibrium approximation one finds $\langle\boldsymbol{\nabla}U\rangle=-L_1(\partial\xi_e/\partial\rho)\boldsymbol{\nabla}\rho$. With the help of the third moment

$$\langle u_\alpha u_\beta u_\gamma\rangle = L_3\hat{H}_\alpha\hat{H}_\beta\hat{H}_\gamma + \frac{L_1-L_3}{5}(\hat{H}_\alpha\delta_{\beta\gamma}+\hat{H}_\beta\delta_{\alpha\gamma}+\hat{H}_\gamma\delta_{\alpha\beta}), \tag{B5}$$

where $L_3(\xi_e)=L_1(\xi_e)-5L_2(\xi_e)/\xi_e$; also, the moment

$$\langle u_\alpha u_\beta\boldsymbol{\nabla}_\beta U\rangle = -\langle u_\alpha u_\beta u_\gamma\rangle\hat{H}_\gamma\frac{\partial\xi_e}{\partial\rho}\boldsymbol{\nabla}_\beta\rho \tag{B6}$$

can be evaluated. Inserting these relations into the diffusion flow $\boldsymbol{j}^U$, Eq. (10), it becomes $\boldsymbol{j}^U=-D_\perp^U(\mathbf{1}-\hat{\boldsymbol{H}}\hat{\boldsymbol{H}})\cdot\boldsymbol{\nabla}_\rho-D_\parallel^U\boldsymbol{\nabla}_\rho$, where

$$D_\perp^U = -\rho\frac{\partial\xi_e}{\partial\rho}\left[D_\perp^0L_1+(D_\parallel^0-D_\perp^0)\frac{L_1-L_3}{5}\right], \tag{B7}$$

$$D_\parallel^U = -\rho\frac{\partial\xi_e}{\partial\rho}\left[D_\perp^0L_1+(D_\parallel^0-D_\perp^0)\frac{3L_1+2L_3}{5}\right]. \tag{B8}$$

Summing the different contributions to the total diffusion flow $\boldsymbol{j}$, Eq. (7), the total diffusion coefficients (27) and (26) are obtained from Eqs. (17), (18), (B3), (B4), (B7), and (B8). The resulting expression has been simplified by use of the mathematical identity $L_2'(x)-[2L_1(x)+3L_3(x)]/5=-L_1(x)L_2(x)$.

---

[1] S. V. Dvinskikh and I. Furó, J. Chem. Phys. **115**, 1946 (2001).

[2] S. V. Dvinskikh, I. Furó, H. Zimmermann, and A. Maliniak, Phys. Rev. E **65**, 061701 (2002).

[3] H. Kelker and R. Hatz, *Handbook of Liquid Crystals* (Verlag Chemie, Weinheim, 1980).

[4] M. P. Allen, Phys. Rev. Lett. **65**, 2881 (1990).

[5] S. Hess, D. Frenkel, and M. P. Allen, Mol. Phys. **74**, 765 (1991).

[6] J.-C. Bacri, A. Cebers, A. Bourdon, G. Demouchy, B. M. Heegaard, and R. Perzinski, Phys. Rev. Lett. **74**, 5032 (1995).

[7] R. G. Larson, *The Structure and Rheology of Complex Fluids* (Oxford University Press, New York, 1999).

[8] H. Ehrentraut and S. Hess, Phys. Rev. E **51**, 2203 (1995).

[9] W. Franklin, Phys. Rev. A **11**, 2156 (1975).

[10] E. Blums, J. Magn. Magn. Mater. **149**, 111 (1995).

[11] K. I. Morozov, J. Magn. Magn. Mater. **122**, 98 (1993).

[12] S. Hess, Z. Naturforsch. A **31A**, 1034 (1976).

[13] M. Doi, J. Polym. Sci., Polym. Phys. Ed. **19**, 229 (1981).

[14] M. A. Martsenyuk, Y. L. Raikher, and M. I. Shliomis, Sov. Phys. JETP **38**, 413 (1974).

[15] A. Y. Zubarev and L. Y. Iskakova, JETP **80**, 857 (1995).

[16] R. Gans, Ann. Phys. **86**, 628 (1928).

[17] S. Prager, J. Chem. Phys. **23**, 2404 (1955).

[18] S. Hess and R. Müller, Opt. Commun. **2**, 172 (1974).

[19] R. Vasanthi, S. Bhattacharyya, and B. Bagchi, J. Chem. Phys. **116**, 1092 (2002).

[20] D. Baalss and S. Hess, Phys. Rev. Lett. **57**, 86 (1986).

[21] P. Ilg and M. Kröger, Phys. Rev. E **66**, 021501 (2002), erra-

tum: Phys. Rev. E **67**, 049901 (2003).
[22] B. Huke and M. Lücke, Rep. Prog. Phys. **67**, 1731 (2004).
[23] M. Kröger and H. S. Sellers, in *Complex Fluids*, edited by L. Garrido (Springer, New York, 1992), Vol. 415 of Lecture Notes in Physics, pp. 295–301.
[24] M. Kröger and H. S. Sellers, J. Chem. Phys. **103**, 807 (1995).
[25] P. Ilg, M. Kröger, and S. Hess, J. Chem. Phys. **116**, 9078 (2002).
[26] P. Ilg, I. V. Karlin, and H. C. Öttinger, Phys. Rev. E **60**, 5783 (1999).