Rotational Diffusion Model of 2-Dimensional Magnetic Alignment

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Abstract. The dynamic process of the 2-dimensional magnetic alignment was analyzed by the rotational diffusion model which took account of the influence of thermal disturbance. Solving the rotational diffusion equation yielded the distribution function and the order parameter. The influences of diffusion were discussed by comparison with the magnetic alignment without diffusion.

1. Introduction
Applying high magnetic fields causes magnetically weak particles to align, then creates new materials functions.[1] However, usually the magnetic alignment (MA) is disturbed by the Brownian motion of the particles because the magnetic free energy is comparable to the thermal energy in ordinary magnetic fields. Consequently, the MA of such particles show the Boltzmann distribution in direction at equilibrium.[2] This indicates that the influence of Brownian motions must be considered in the dynamic process of the MA also. So far the alignment process was analyzed by the mechanical model which took no account of thermal disturbance. [3,4] Recently, this model is improved by including random torque representing thermal disturbance.[5]

Meanwhile, the influence of Brownian motion on MA is systematically formulated as the rotational diffusion equation (RDE).[6] In this paper, we apply this formulation to the 2-dimensional magnetic alignment (2d-MA) where the rotation of particles is restricted in a plane. The present paper intends to study the essential influences of rotational diffusion on MA, but not to explain a specific experimental result.

2. Formulation
2.1. Rotational diffusion equation
The particle is supposed to have uniaxial anisotropy of magnetic susceptibility \( \chi_1=\chi_2\neq\chi_3 \) with an ellipsoid of revolution in shape where the principle axes of magnetic susceptibility \( \chi_1, \chi_2 \) and \( \chi_3 \) coincide with the \( x_1, x_2 \) and \( x_3 \) axes of ellipsoid, respectively. The anisotropic susceptibility is denoted by \( \Delta \chi=\chi_3-\chi_1 \). The unit vectors \( \mathbf{u}, \mathbf{v} \) and \( \mathbf{w} \) are taken along the \( x_3, x_1 \) and \( x_2 \) axes while \( \mathbf{u} \) is used as the director of a particle. The cylindrical coordinates of \( \mathbf{u} \) are \((r=1, \theta, z=0)\) with respect to the space-fixed axes of \( X_1, X_2 \) and \( X_3 \) as shown in Fig. 1.
It is presumed that the system consists of \( N_0 \) particles which have no interaction with each other. A static magnetic field \( B \) is applied to the particles along the \( X_1 \) axis and their rotational plane is restricted in the \( X_1X_2 \) plane. Thus, the potential energy \( U \) of the particle depends on the angle \( \theta \).

\[
U = -\frac{1}{2\mu_0} VB^2 X_1 - \frac{1}{2\mu_0} VB^2 \Delta \chi \cos^2 \theta = U_0 - \beta \cos^2 \theta ,
\]

where \( V \) is the volume of a particle, \( U_0 \) is the \( \theta \)-independent term and \( \beta \) is the magnetic energy related to the magneto-anisotropy.

\[
\beta = \frac{1}{2\mu_0} VB^2 \Delta \chi .
\]

It is well-known that the particle tends to align in parallel or perpendicular to the magnetic field according to the positive or negative \( \Delta \chi \).

![Figure 1. Cylindrical coordinates for the 2-dimensional magnetic alignment of uniaxial particles.](image)

The RDE has been proposed previously.[6]

\[
\frac{\partial C}{\partial t} = -\mathbf{R} \cdot \mathbf{J}_R = D_R \mathbf{R}^2 C - \mu_R \mathbf{R} \cdot (CN) ,
\]

where \( C \) is the concentration of particles, \( t \) is the time, \( \mathbf{R} \) is the rotational operator, \( \mathbf{J}_R \) is the rotational flux and \( \mathbf{N} \) is the magnetic torque acting upon a particle. \( D_R \) and \( \mu_R \) are the rotational diffusion constant and the rotational mobility, respectively, which are related by the Einstein relation \( D_R = k_B T \mu_R \). Eq. (3) can be derived as an analogy to the electric field-induced rotation of polymers.[7] The torque is obtained from the potential energy by \( \mathbf{N} = -\mathbf{R} U \). In the present problem, the rotational operator is expressed by \( \mathbf{R} = w \hat{n} \hat{n} \cdot \hat{n} \). We replace the concentration \( C \) by the distribution function \( P \) by \( P = C/N_0 \) which is a function of angle \( \theta \) and time \( t \). Thus, the rotational flux and the RDE for the 2d-MA are written in the cylindrical coordinates as follows.

\[
\mathbf{J}_R = w N_0 \left( -D_x \frac{\partial P}{\partial \theta} - \beta \mu_x P \sin 2\theta \right) ,
\]

\[
\frac{\partial P}{\partial t} = D_x \frac{\partial^2 P}{\partial \theta^2} + \mu_R \beta \frac{\partial}{\partial \theta} (P \sin 2\theta) .
\]
Alternatively, Eq. (5) can be derived from the viewpoint that point particles move on the arc of a semi-circle with unit radius by the translational diffusion. We take the translational coordinate $s$ along the arc to write the translational diffusion equation.

$$\frac{\partial P}{\partial t} = D_s \frac{\partial^2 P}{\partial s^2} + \mu_s \frac{\partial}{\partial s} (P F).$$  \hfill (6)

This is the same as Eq. (5) because the force acting on a point particle is $F = \beta \sin^2 s$ with $s = \theta$.

The distribution function must be normalized by

$$\int_{-\pi/2}^{\pi/2} P d\theta = 1.$$ \hfill (7)

In addition, it is assumed that the particles are randomly distributed in the initial state at $t=0$.

$$P(\theta,0) = \frac{1}{\pi}.$$ \hfill (8)

We introduce the reduced time $\tau$ and the field parameter $\alpha$ which represents the ratio of magnetic energy to thermal energy.

$$\tau = D_s t = k_s T \mu_s t,$$ \hfill (9)

$$\alpha = \frac{\beta \mu_s}{D_s} = \frac{\beta}{k_s T}.$$ \hfill (10)

Then, the RDE is written by the dimensionless form.

$$\frac{\partial P}{\partial \tau} = \frac{\partial}{\partial \theta} \left( P \sin 2\theta \right).$$ \hfill (11)

Meanwhile, the order parameter $<m>$ is defined for the 2d-MA.

$$<m> = \int_{-\pi/2}^{\pi/2} (2 \cos^2 \theta - 1) P d\theta.$$ \hfill (12)

The value of this quantity is 0 for a random distribution, 1 for the perfect parallel alignment along the field and $-1$ for the perfect perpendicular one.

2.2. Equilibrium state

The equilibrium state is attained when the time tends to infinity, then the time derivative of the distribution function $\partial P/\partial \tau$ disappears in Eq. (5). Simultaneously, the rotational flux of Eq. (4) becomes zero.

$$\frac{dP}{d\theta} + \alpha P \sin 2\theta = 0.$$ \hfill (13)

Solving this gives the equilibrium distribution $P_{eq}$:

$$P_{eq}(\theta) = \frac{\exp(\alpha \cos^2 \theta)}{\int_{-\pi/2}^{\pi/2} \exp(\alpha \cos^2 \theta) d\theta}.$$ \hfill (14)

This is a Boltzmann distribution. Substituting Eq. (14) into Eq. (12) gives the order parameter at equilibrium $<m>_{eq}$.

2.3. Diffusion-free system
It is instructive to investigate the hypothetical system without rotational diffusion. This situation replaces Eq. (11) to
\[
\frac{\partial P}{\partial \tau} = \alpha \frac{\partial}{\partial \theta} (P \sin 2\theta).
\] (15)
This has the analytical solution with the aid of Eqs. (7) and (8).
\[
P = \frac{e^{\alpha \tau} \sec^2 \theta}{\pi (1 + e^{\alpha \tau} \tan \theta)}, \quad (\alpha \tau = \beta \mu \eta),
\] (16)
Furthermore, this leads to the analytical expression of the order parameter.
\[
\langle m \rangle = \tanh (\alpha \tau).
\] (17)
When the time tends to infinity, \(\langle m \rangle\) becomes 1 for \(\alpha > 0\) and \(-1\) for \(\alpha < 0\). In the initial region of time with small \(\tau\), this is approximated by
\[
\langle m \rangle \approx \alpha \tau.
\] (18)

It is considered that this diffusion-free system corresponds to the mechanical model without thermal disturbance. [3,4] As a matter of fact, essentially the same relations as Eqs. (16) and (17) have been derived from the mechanical model for the 2-dimensional magnetic alignment by Kimura et al.[8] Comparing Eq. (17) to the corresponding equation Eq. (15A) in ref. 8 gives the relation \(\mu = (L_1 \eta)^{-1}\) where \(L_1\) is a hydrodynamic coefficient of the particle and \(\eta\) is the viscosity of the ambient medium in the mechanical model.

3. Calculated results and discussion
First, we deal with the MA with rotational diffusion. The distribution function was numerically calculated from Eq. (11) by the difference method for the partial differential equation with Visual C++. The solutions were stably obtained by using the steps of \(\Delta \tau = 0.0001\) and \(\Delta \theta = 1.0\) deg with significant figures of three. Figure 2 illustrates the time-variation of the distribution function for \(\alpha = 10\) where the curves are drawn only in the positive \(\theta\) region because of the even function. The figure clearly indicates the time-developing alignment of particles: The distribution function is flat at \(\tau = 0\), then gradually becomes narrower with time and tends to the curve at equilibrium. It must be emphasized that the distribution function at \(\tau = 0.4\) or more agreed completely with the equilibrium distribution calculated from Eq. (14). This warrants the validity of the computing code used.

![Figure 2. Computed curve of the distribution function \(P\) at different times for the 2-dimensional magnetic alignment with rotational diffusion (\(\alpha = 10\)).](image-url)
The time-dependence of order parameter \( <m> \) is calculated according to Eq. (12) from the computed distribution functions as shown in Fig. 3. Hence, the saturated value of the order parameter agreed with that calculated from Eqs. (12) and (14) for every field parameter. The magnetic alignment attains the equilibrium state faster with larger field parameters \( \alpha \). In addition, it also proceeds faster with larger values of the rotational diffusion constant \( D_R \) because the real time is \( t=\tau/D_R \). It is recognized that the order parameter is abruptly increased in the region between \( \alpha=1 \) and 10, meaning that the MA needs the magnetic energy which overcomes the thermal energy.

![Figure 3.](image1)

**Figure 3.** Time-dependence of the order parameter for the 2-dimensional magnetic alignment with rotational diffusion.

Next, as for the MA without rotational diffusion, the distribution function is calculated by the formula of Eq. (16). The distribution function rapidly gets narrower with passing time, which tends to a delta function \( P=\delta(\theta) \) at infinite time for every value of \( \alpha \). Then, the order parameter was calculated by the formula of Eq. (17) as shown in Fig. 4. The order parameter necessarily takes unity at infinite time for any value of the field parameter \( \alpha \).

![Figure 4.](image2)

**Figure 4.** Time-dependence of the order parameter in the 2-dimensional magnetic alignment without rotational diffusion.

In the MA without rotational diffusion, the order parameter varies linearly with time at the initial stage, obeying Eq. (18) as exemplified in Fig. 5. In the MA with rotational diffusion, diffusion is negligible at the initial stage when the particle concentration is nearly homogeneous. Accordingly, the
order parameter is approximately linear against time in the MA with rotational diffusion also. As time passes, however, diffusion occurs noticeably because the distribution has a gradient. Then, the magnetic alignment is suppressed by diffusion.

![Graph showing linear and diffused order parameters against time.](image)

**Figure 5.** Comparison of the order parameter for the 2-dimensional magnetic alignment with/without rotational diffusion ($\alpha=3$).

As the field parameter $\alpha$ is increased, the diffusion term in the RDE of Eq. (5) or (11) becomes much smaller than the drift one. As a consequence, the MA with rotational diffusion behaves like that without rotational diffusion. Finally, all of the above results are concerned with the positive $\alpha$. When $\alpha$ is negative, the results are the same as above except that the order parameter is negative with the same absolute value.

In conclusion, we have made clear the influence of thermal disturbance on the dynamic process of the 2-dimensional magnetic alignment by obtaining the distribution function and the order parameter for the systems with/without rotational diffusion.

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