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Multiparametric rotational drift spectroscopy

M. A. Rückert[a,*] · P. Vogel[a] · T. Kampf[a,b] · V. C. Behr[a]

[a]Department of Experimental Physics 5 (Biophysics), University of Würzburg, 97074, Würzburg, Germany.
[b]Department of Diagnostic and Interventional Neuroradiology, University Hospital Würzburg, 97080, Würzburg, Germany
[*]Corresponding author, email: Martin.Rueckert@physik.uni-wuerzburg.de

Abstract

Rotational Drift Spectroscopy (RDS) is a novel spectroscopic method for magnetic nanoparticles. It is based on measuring the rotational drift of magnetic nanoparticle ensembles in a rotating magnetic field, which is below the magnetic field strength necessary for rotating the magnetic nanoparticles synchronously. The resulting asynchronous rotational drift strongly depends on the properties of the magnetic nanoparticles and their environment. This provides a promising basis for spectroscopic measurements with high sensitivity as well as high specificity, e.g., detecting specific molecules in a liquid via functionalized magnetic nanoparticles. In this work, multiparametric Rotational Drift Spectroscopy (mRDS) is presented, which makes use of the nonlinear dependency of the RDS signal of, e.g., the sequence amplitude and the viscosity of the suspending liquid of the magnetic particle sample. This allows access to a variety of parameters of magnetic nanoparticle suspensions.

I. Introduction

Magnetic particles show rotational drift in rotating magnetic fields, which are too weak to rotate them synchronously. The resulting drift frequency of the magnetic particles strongly depends on the rotational friction or the amplitude and frequency of the rotating magnetic field. This phenomenon was used in [1] for constructing a single bacteria detector by optically measuring the rotating frequency of a single functionalized magnetic microparticle. Rotating Drift Spectroscopy [2, 3] aims at inductively measuring the rotational drift of magnetic nanoparticle ensembles in liquid suspension. This requires orienting all magnetic moments in one direction before the measurements starts, otherwise all magnetic moments are oriented randomly, resulting in zero net magnetization with respect of any asynchronous frequency components. In [2] this was achieved by applying a short magnetic pulse 3 ms before the start of the rotating magnetic field, with a pulse duration of 25 µs and a pulse strength of 200 mT. This 3 ms dead time didn't allow measuring typical suspensions with short relaxation times. Solutions to this are presented in [4, 5]. The presented work evaluates the possibilities of multiparametric RDS with respect to the strength of the rotating magnetic field and the viscosity. Due to the corresponding nonlinear dependence of the rotational drift frequency in addition to the critical field strength, where asynchronous rotation transitions into synchronous rotation, this method has the potential of being highly sensitive with respect of small differences in particle properties controlled via functionalization.

II. Material and methods

In the following, multiparametric RDS refers to series of RDS measurements for different parameter settings. The corresponding parameters considered in this work are the sequence amplitude and the sample viscosity. The setup described in [2] is shown schematically in Fig. 1 and used for generating the rotating magnetic field with

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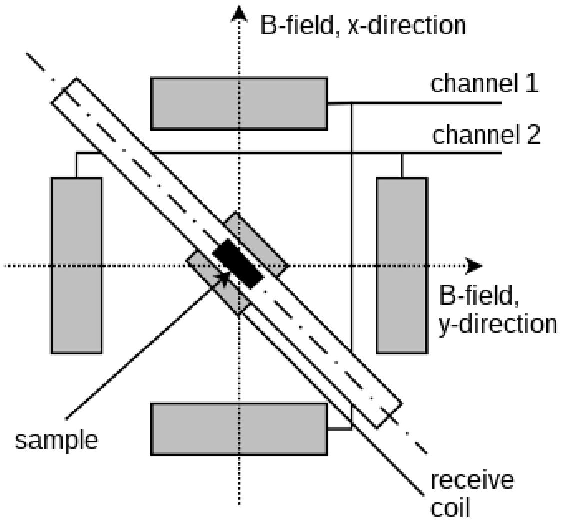

Figure 1: Schema of the used setup. Two orthogonal coil pairs generate the rotating magnetic field up to 60 mT at 50 kHz. The samples were 5 mm glass tubes filled with 40 µl suspension.

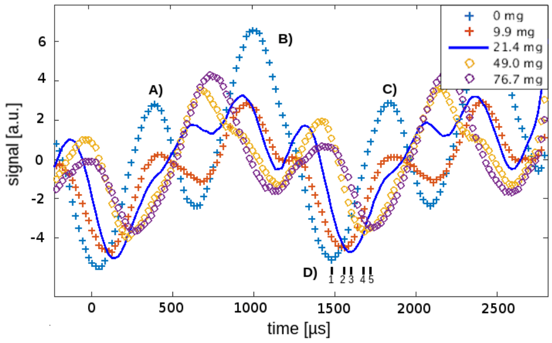

Figure 2: Magnetic nanoparticles with 30 nm core diameter (SHP-30) in liquid suspension for different amounts of sugar.

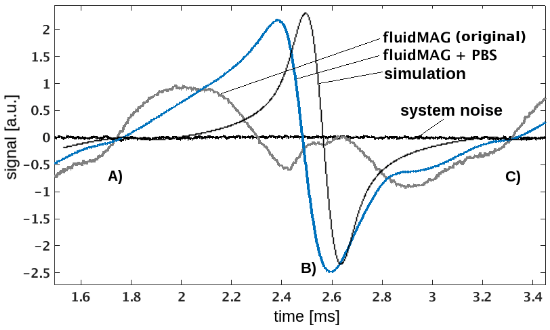

Figure 3: The signal shape of fluidMAG-UC/C changes if PBS is added. The simulated graph corresponds to a particle system with a relaxation time of zero. Particle conc: 25 mg/ml.

a frequency of 50 kHz and a magnetic field strength up to 60 mT. The frequency applied to the x-channel and y-channel are slightly different, in order to generate a rotating field that periodically changes its rotating direction. In [2] this resulted in a signal echo train. In the presented work this frequency is to be understood as the repetition rate of a single RDS measurement instead. It is between 650 Hz and 1 kHz for the following measurements. In [2] a short initial pulse is used to orient all magnetic moments prior to the measurement. The presented work uses the fact that a weak offset field oriented in parallel to the rotating plane causes the magnetic moments to align themselves. The offset field is 2 mT and generated using a permanent magnet. It is oriented in parallel to the receive coil. According to simulations based on the equation and assumptions used in [3], which is restricted to magnetic particles that effectively show no Néel rotation, the effect of the offset field becomes negligible as soon as the magnetic field starts rotating again.

The disadvantage of this effect is that it destroys any phase distribution (i.e., the orientation of all particle moments inside the sample) that was imprinted during the prior part of the sequence and essentially restarts the measurement whenever the external magnetic field transitions from rotating to linearly oscillating and vice versa. Therefore, it is not possible to measure signal echo trains in the presence of weak offset fields. The big advantage is the lack of any abrupt field switching that could cause problematic pulse responses in the receiver chain. This allows avoiding the dead time occurring in [2] and therefore allows measuring samples with much shorter relaxation times. The receive chain blocks the induction caused by the rotating magnetic field with a 9-pole Chebychev filter with a cut-off frequency of 10 kHz. The samples were 40 µl suspension put in 5 mm glass tubes. The particle suspensions used were fluidMAG-UC/C 50 nm (chemicell, Berlin, Germany), SHP-30 (ocean NanoTech, San Diego, USA), carbon coated cobalt 50 nm in water (Sigma Aldrich, USA) and magnetotactic bacteria (dead sediments). The fluidMAG-UC/C samples were used in original form and mixed with phosphate-buffered saline (PBS) in order to model basic interactions with particle surface functionalization. The viscosity of the SHP-30 samples (magnetite particles with 30 nm diameter) was changed by adding sugar.

III. Results and discussion

The first example is shown in Fig. 2. It shows SHP-30 with different amounts of sugar. A), B) and C) mark the time points, where the rotating magnetic field changes its rotating direction. The signal shape is expected to be the same at A) and B) for magnetic particles that show no Néel rotation and have a sufficiently long Brownian relaxation time, based on simulations using the equation in [3]. As the maxima marked with A) and B) are not the same, this indicates that there are additional signal features present. In addition, it shows only low frequency features, which indicates a wide distribution of particle properties. The marks at D) show the shift of the minima for increasing amounts of sugar.

Fig. 3 shows a single measurement for the original fluidMAG-UC/C suspension and for fluidMAG-UC/C added with PBS. It also shows a simulation for particle systems of negligible relaxation time. In Fig. 3, the signal at A) and C) is missing. The simulation corresponds to a particle system with a relaxation time of zero and a Langevin parameter of $(mB)/(k_B T) = 30$ with m being the magnetic moment of the particle, B being the amplitude of the rotating magnetic field, k_B the Boltzmann constant and T the temperature. Therefore, the missing repetition of the signal shape at A) and C) is characteristic for particle systems with negligible relaxation time. The difference between A) and B) in Fig. 2 suggests a superposition of particles with finite relaxation time and particles with negligible relaxation time.

IV. Conclusions

The presented work indicates that increasing the parameter space for RDS provides measurements that are highly sensitive to small differences in magnetic nanoparticle systems. This is crucial for, e.g., bio sensing applications based on magnetic nanoparticles. Multiparametric RDS shows promising results for different types of magnetic nanoparticle suspensions, including ones commonly used for MPI. This makes it a very promising aspect for biosensing applications and expanding the molecular imaging capabilities of MPI.

Acknowledgments

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Author’s statement

Conflict of interest: Authors state no conflict of interest.

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