Experimental report

23/12/2020

Proposal: **5-32-889**

Council: 4/2019

Title: Magnetic correlations in a liquid magnet

Research area: Soft condensed matter

This proposal is a new proposal

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Samples: BaSc0.5Fe11.5O38/barium hexaferite

Instrument	Requested days	Allocated days	From	To
D33	5	3	10/01/2020	13/01/2020
D22	0	0		

Abstract:

The existence of polar, i.e. ferromagnetic or ferroelectric ordering, in dipolar liquids has been a long standing open question. Some of the models predicted it some not. The main issue is that appropriate nearest neighbors positional and orientational correlations are crucial for the appearance of the polar phase and are difficult to predict correctly. Recently, it has been shown that in a special kind of ferrofluids, made of magnetic nanoplatelets suspended in alcohols above a certain volume fraction a ferromagnetic nematic phase appears. These ferrofluids present a unique model system, in which positional and magnetic correlations between the constituents can be studied and polarization analyzed SANS is an excellent method for such studies.

Magnetic and positional correlation studies for ferromagnetic ferrofluids

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Background

The existence of polar, i.e. ferromagnetic or ferroelectric ordering, in dipolar liquids has been a long standing open question. Some of the models predicted it some not [1]. The main issue is that appropriate nearest neighbours' positional and orientational correlations are crucial for the appearance of the polar phase and are difficult to predict correctly. Recently, it has been shown that in a special kind of ferrofluids, made of magnetic nanoplatelets suspended in alcohols above a certain volume fraction a ferromagnetic nematic phase appears [2,3]. These ferrofluids are therefore liquid magnets and they are sensitive to very small magnetic fields. They present a unique model system, in which positional and magnetic correlations between the constituents can be studied, which are crucial for the existence of ferromagnetic phase. Polarization analysed SANS is an excellent method for such studies.

Aims of the experiment

The aims of the proposed experiment were to fully quantitatively evaluate magnetic correlations using PASANS and gain information on the strength of the local magnetic field and dynamics of the correlations by stroboscopic technique.

Ferrofluids are suspensions of magnetic nanoparticles in various solvent. The ferrofluid planned to use in this study is made of magnetically single domain barium hexaferrite nanoplatelets (with thickness 3 nm and average diameter 50 nm) dispersed in 1-butanol [4,5]. Due to high magnetocrystalline anisotropy, platelets' magnetic moments are perpendicular to the plane of the platelets. The stability of the suspensions is achieved by electrostatic repulsion [6]. Due to anisotropic magnetic interaction and screened electrostatic repulsion the probability that the neighbouring platelets have a given relative position and orientation is larger for some configurations (e.g. parallel dipoles in the direction of the dipoles). Because of the long range nature of the interaction, many body interactions are important, but are difficult to predict. By measurement of positional and magnetic microstructure and its dynamics, information on the magnitude and spatial variations of the local field, which determines average positional correlations and orientation of the dipoles, can be obtained.

Static measurements give information on probability that the neighbouring platelets have a given relative position and orientation, they, however, do not tell anything about the time the neighbours persist in a given configuration. In order to get insight in the strength of the interaction, which determines the dynamical behaviour, we additionally studied the response of the system to small AC fields with different frequencies using stroboscopic approach. In stroboscopic measurement the detector is triggered synchronously with the AC field to obtain information on ferrofluid microstructure at different phases of the field.

Because the suspensions are sensitive to magnetic fields of order of Earth magnetic field, it is necessary to create a zero field sample environment ($B < 1$ μT). This is for example achieved by 3 pairs of coils. The coils are also used for application of small external field in arbitrary direction.

Samples

For the experiments three suspensions of ferrofluid with different volume fractions were used, giving us the opportunity to observe a sample in the nematic state (S3) with $\phi = 5.7$ %, an isotropic sample (S2) with $\phi = 3.0$ % and a diluted sample (S1) with $\phi = 1.0$ %. The samples are sealed between two 1 mm thick fused quartz plates with spacers defining the sample thickness to be 115 μm ± 10 μm.

Results

On the D33 beamline couple of different modes of operation were used. For every experiment neutrons with $\lambda = 6\,\text{Å}$ were used. The sample-detector distance, that was used for most experiments is 4 m and collimation 5.3 m.

SANS with polarized and guided neutrons and ^{3}He analyser gives us the results on Figure 1 (a-d). The sample *S3* was placed in our magnetic chamber used for compensating static external magnetic field and application of small static or oscillating magnetic fields. With this results magnetic correlation studies can be further evaluated.

Another useful mode was the Time-Of-Flight (TOF) mode that gives us the opportunity to observe the positional correlations in the nematic sample for different sections of the period, visible in Figure 1 (e-g). Three different modes are shown on the plots below. The radial plots show that the correlation distance between the constituents are the same – not affected by frequency or amplitude of oscillating magnetic field. The interesting are the azimuthal plots, where the direction of correlation changes in some cases. There exist a range of frequencies and amplitudes of the oscillating magnetic field where correlations in average change direction to perpendicular to the direction of applied magnetic field. With the TOF mode more than only average can be observed – inside one period the direction changes in case (e). Case (f) has a too strong field so the platelets orient faster and no effect is visible and in case (g) correlations exist in both parallel and perpendicular direction at the same time.

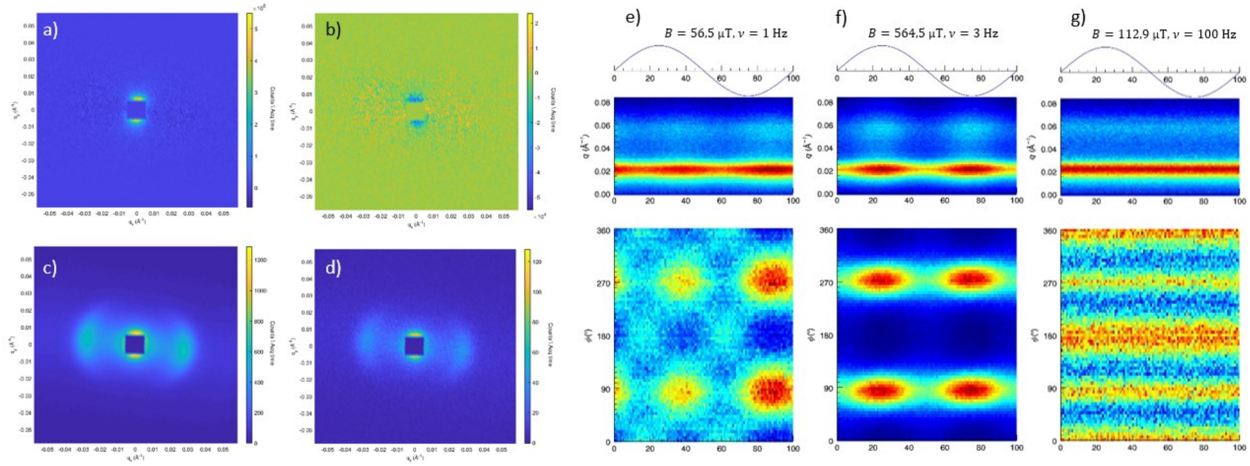

Figure 1: Results of PASANS for nematic sample S3 in the magnetic field of 34 μT in horizontal direction (a-d) corresponding to polarizations 11, 10, 01, 00 respectively. The right part (e-g) are results of time-of-flight SANS showing radial and azimuthal dependency of positional correlations in oscillating magnetic field with amplitude and frequency marked on top. In (e) positional correlations change direction to perpendicular to the direction of the field in (f) the correlations don't change direction and in (g) the correlations are both – vertical and horizontal direction at the same time.

The third mode used were measurements with polarized neutrons. The difference between polarizations up and down is shown on the Figure 2. The results show some asymmetry but otherwise they are inconclusive.

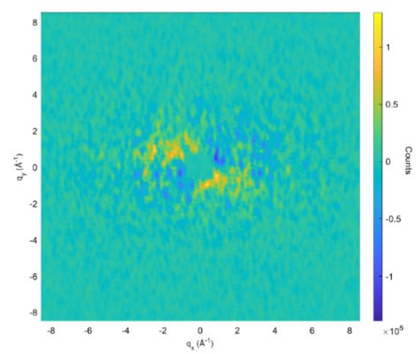

Figure 2: SANSPol difference between polarization up and down in oscillating magnetic field with frequency 1 Hz and amplitude 226 μT.

Conclusions

PASANS gives us more information on magnetic correlations between constituents in ferrofluids, TOF mode allows us to have an overview of behaviour inside one period of oscillation. Positional correlations and their direction gives new input in the study of the stripes regime below the optical limitation. The SANSPol didn't give us enough statistics to extract more data than only the visual anisotropy so the results of SANSPol are inconclusive.

References

[1] B. Huke and M. Lücke, Rep. Prog. Phys. **67**, 1731 (2004).
[2] M. Shuai, A. Klittnick, Y. Shen, G. P. Smith, M. R. Tuchband, C. Zhu, R. G. Petschek, A. Mertelj, D. Lisjak, M. Čopič, J. E. Maclennan, M. A. Glaser, N. A. Clark, Nat Commun **7**, 10394 (2016).
[3] A. Mertelj and D. Lisjak, Liquid Crystals Reviews **5**, 1 (2017).
[4] D. Lisjak and M. Drofenik, Crystal Growth & Design **12**, 5174 (2012).
[5] D. Lisjak and A. Mertelj, Progress in Materials Science **95**, 286 (2018).
[6] P. Hribar Boštjančič, M. Tomšič, A. Jamnik, D. Lisjak, and A. Mertelj, J. Phys. Chem. C **123**, 23272 (2019).