ESRF	**Experiment title:** Structure and dynamics of ferrogels containing anisotropic magnetic particles	**Experiment number**: SC3947
Beamline: ID02/ID10	**Date of experiment**: from: 12.12.2014 to: 15.12.2014 (ID02) 29.10.2014 03.11.2014 (ID10)	**Date of report**: 7.9.2015
Shifts: 9 / 15	**Local contact(s)**: Johannes Moeller (ID02) / Beatrice Ruta (ID10)	*Received at ESRF:*

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Report:

The mesostructure and dynamics of ferrogels contatining spindle-shaped hematite particles with different aspect ratios is investigated by means of SAXS and XPCS in dependence on an external magnetic field. The hydrogel-matrix consisting of poly-N-Isopropylacrylamide is varied with respect to the volume fraction of the polymer and its crosslinking density.

At the beamline ID02 the experiments are carried out with 12 keV photons at a single detector distance of $d = 20\,\mathrm{m}$ covering $8 \times 10^{-3}\,\mathrm{nm}^{-1} \leq Q \leq 0.2\,\mathrm{nm}^{-1}$. The small angle scattering in presence of magnetic fields perpendicular and parallel to the primary beam is investigated at flux densities up to $B_{\perp} = 1.4\,\mathrm{T}$ and $B_{\parallel} = 1.0\,\mathrm{T}$. The spindle-shaped hematite particles align with their long axis perpendicular to an external field. Due to the high electron density of iron oxide the scattering is dominated by the hematite particles whereas the background resulting from the polymer matrix is extremely weak.

The orientational distribution functions (odf) of aqueous suspensions of the particles are compared to the odfs of identical particles embedded in different hydrogel-matrices. Both in aqueous suspension and in ferrogels a completely reversible, field-induced isotropic-nematic transition occurs. Due to the viscoelasticity of the hydrogels, however, the nematic alignment is hindered in ferrogels. This is already in false-color representations of the scattered intensity visible by the less anisotropic scattering patterns of ferrogels compared to aqueous suspensions at the same flux density (Fig. 1).

The sector-averaged scattered intensity is quantitatively analysed employing Boltzmann approach describing the energy of hematite particles in an external magnetic field via contributions both from permanent dipole moments and induced dipole moments resulting from an anisotropic susceptibility tensor of the particles. With the magnetic moment μ of a particle and the anisotropy $\Delta\chi = \chi_{\parallel} - \chi_{\perp}$of its magnetic susceptibility the energy of non-interacting hematite particles in an external field reads as

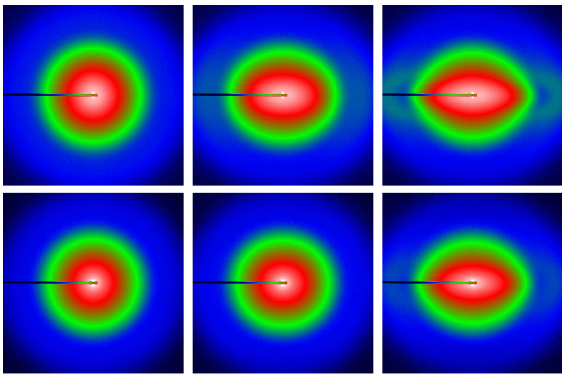

Fig. 1: False color representation of the scattered intensities resulting from aqueous suspensions (upper row) and ferrogels (lower row). The direction of the external field is parallel to the direction of the shadow of the beamstop holder. The scattering patterns of the ferrogels are less anisotropic than those of the aqueous suspensions.

$$V(\vartheta_\mu, \vartheta_P) = \mu B \cos\vartheta_\mu + \frac{\Delta\chi V_P B^2}{2\mu_0}\cos^2\vartheta_P \quad ,(1)$$

where ϑ_μ is the angle between the magnetic moment and the external field and ϑ_P the angle between the particles director and the external field. V_P is the volume of the particle and μ_0 the magnetic field constant. The scattered intensity can consistently be described with the form factor of spindle-shaped particles underlaying a Schulz-Flory size distribution and the odf resulting from the orientation-dependent potential (1). The result of a global fit in dependence on the modulus and the direction of the scattering vector $\mathbf{Q}$ with respect to the direction of the external field and the flux density of the external field is displayed in Fig. 2 (lhs). From the ODF the nematic order parameter $S_2 = \langle 3\cos^2\vartheta - 1\rangle/2$ is determined and compared for different hydrogels (Fig. 2, rhs).

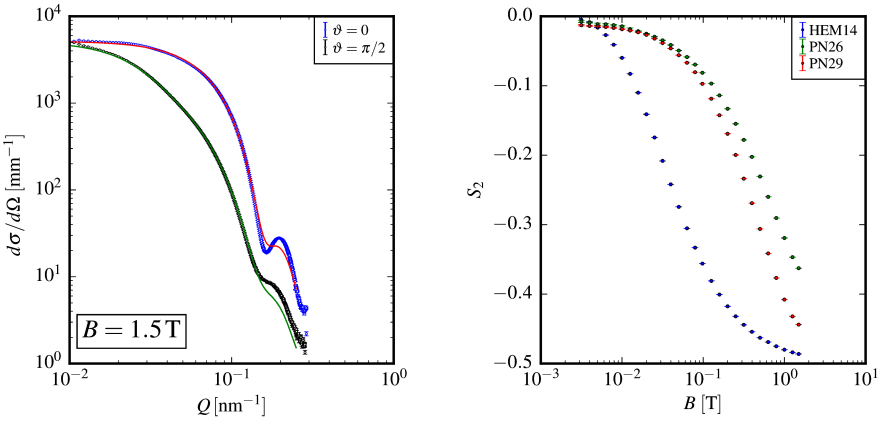

Fig. 2. Simultaneous fit of sector averaged intensities in sectors with an acceptance of $\Delta\vartheta = \pm 2.5°$. Here only the sectors parallel $\vartheta = 0$ and perpendicular ($\vartheta = \pi/2$) to the external field are displayed (lhs). The nematic order parameter S_2 of an aqueous suspension (blue) is compared to those of ferrogels with different crosslinking density (red and green) containing the same particles.

From XPCS experiments in presence of external fields correlation functions in dependence of the moduls and the direction of the scattering vector with respect to the field-direction are calculated. The XPCS experiments at ID10 are carried out at 7 keV to avoid resonant scattering from Fe. For the calculation of the correlation functions the detector is partioned in sectors with an acceptance of $\Delta\vartheta = \pm 15°$. While the contrast strongly depends on the direction of $\mathbf{Q}$ the relaxation rates are nearly independent of the direction due to the confinement of the particles in the hydrogel-network. At large Q, clearly two seperated relaxtion processes are visible (Fig. 3).

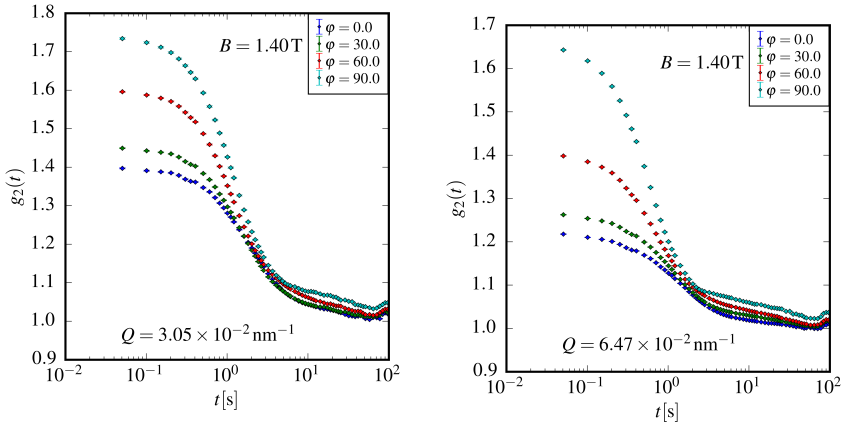

Fig. 3 Correlation functions for different directions of $\mathbf{Q}$ with respect to the direction of the external field.