

Magnetic properties of isotropic and anisotropic CoFe_2O_4 -based ferrogels and their application as torsional and rotational actuators

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CoFe_2O_4 -based ferrogels were prepared with both isotropic and anisotropic orientation of the magnetic anisotropy axis of the magnetic particles. In contrast to the superparamagnetic properties of the ferrofluid, the ferrogels exhibit hysteresis, indicating that (i) a significant fraction of magnetic particles has volumes beyond the critical value that allows Néelian relaxation, and (ii) a mechanical interaction between the particles and the polymer network exists, which prevents the particles from Brownian relaxation. The contribution of such particles was investigated by field cooling field warming and zero field cooling field warming measurements as well as temperature-dependent magnetization measurements. By application of an external field during gel polymerization, a magnetic texture was induced as confirmed by the angular dependence of m_R/m_S and H_C . The net-magnetic torque, exerted on the magnetic particles in an anisotropic ferrogel in combination with the soft elastic properties of the gel matrix enables the application as torsional soft actuator as demonstrated.

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I. INTRODUCTION

A ferrogel is a chemically crosslinked polymer network gel which is functionalized by using a ferrofluid instead of pure solvent as swelling agent [1]. Ferrogels represent viscoelastic soft materials which are sensitive to an applied external magnetic field. The mechanical force exerted on the magnetic particles in a field gradient is transferred to the polymer network which may result in a significant change of the ferrogel's shape and enables us to perform mechanical work on a macroscopic scale [1–4]. In previous studies, magnetite (Fe_3O_4)-based ferrofluids have been used as a magnetic component. At an average grain size of about 10 nm, magnetite nanoparticles are superparamagnetic and the magnetic properties of derived isotropic ferrogels can be expected to be comparable to the constituting ferrofluid in the sense that they should also exhibit superparamagnetic behavior [3].

It has been demonstrated in several studies that uniaxial magnetic gels can be obtained when the ferrogel precursor is exposed to an external magnetic field prior to polymerization [5–7]. It is well established that in an external magnetic field, the magnetic particles in a ferrofluid are subjected to condensation from a gaslike distribution to dense liquidlike aggregates such as drops or stripes [7–10], which on a microscopic scale manifest linear particle chains [11,12]. While in ferrofluids such structures may dissolve again when the field is removed, they remain conserved in ferrogels due to the suppressed mobility obtained after polymerization of the gel network. The dipolar interaction within these aggregates gives rise to magnetic anisotropy due to the reduced magnetostatic energy for magnetization along a chain of magnetic nanoparticles as compared to a magnetization perpendicular to the chain axis (shape anisotropy). Due to the cross coupling of the anisotropic magnetic and elastic components, such uniaxial magnetic gels exhibit several interesting hydrody-

namic properties such as shear-induced magnetization or oscillating shear strain caused by oscillating magnetic fields [13].

The present study focuses on the preparation of isotropic and uniaxial ferrogels based on a cobalt-ferrite ferrofluid. Due to the much larger magnetocrystalline anisotropy of CoFe_2O_4 as compared to Fe_3O_4 , a significant fraction of particles within the size distribution of such a ferrofluid is ferromagnetic, i.e., the magnetic moments are preferentially oriented along an anisotropy axis of the particles. One yet observes superparamagnetic behavior in the ferrofluid due to Brownian rotational relaxation of the particles in the liquid matrix. The suppression of this relaxation in the ferrogel should give rise to distinctly different magnetization behavior of the ferrogel as compared to the ferrofluid. Additionally, one can assume that the high magnetocrystalline anisotropy should dominate the overall anisotropy of uniaxial magnetic gels. Therefore, this study is mainly focussed on the characterization of the magnetic properties of the CoFe_2O_4 ferrofluid and derived isotropic and anisotropic ferrogels.

II. BASIC CONSIDERATIONS

An approach to understand the magnetism of a ferrogel can be derived from the magnetic properties and the associated microscopic mechanisms of the ferrofluid of which it is synthesized. A ferrofluid is a colloidal suspension of single domain magnetic particles with an average diameter of approximately 10 nm. Depending on the relative magnitude of anisotropy energy to thermal energy, these particles exhibit superparamagnetic behavior, i.e., the magnetization may overcome the anisotropy energy barrier by thermal activation. Furthermore, if the magnetic moment was oriented under the influence of an external field, this orientation is lost when the field is switched off (Néelian relaxation) within a characteristic time scale τ_N which is given by [14]

$$\tau_N = \tau_0 \exp\left(\frac{KV}{k_B T}\right), \quad (1)$$

with $\tau_0 \approx 10^{-9}$ s, the anisotropy constant K and particle volume V . The magnetization of a dilute (i.e., neglecting dipolar

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interaction) ensemble of superparamagnetic particles can be described by the Langevin function $L(\xi)$ [15,16]:

$$M(H) = M_S L(\xi) \quad (2)$$

with

$$L(\xi) = \coth(\xi) - \frac{1}{\xi}, \quad \xi = \frac{\mu_0 m H}{k_B T}. \quad (3)$$

M_S denotes the magnetic fluid's saturation magnetization, μ_0 denotes the vacuum permeability, m is the magnetic moment of a single magnetic particle, H is the applied magnetic field, k_B is the Boltzmann constant, and T is the temperature.

For a given temperature \tilde{T} and a time constant $\tilde{\tau}$ which is characteristic to a specific measurement, there is a critical particle diameter D_{sp} (assuming spherical shape) that discriminates smaller particles $D < D_{sp}$ exhibiting superparamagnetic behavior from larger particles $D > D_{sp}$ which are ferromagnetic. For cubic anisotropy and assuming $\tilde{T} = 298$ K and $\tilde{\tau} = 10$ s, the critical particle size can be estimated [17] as $D_{sp} \approx 60$ nm for Fe_3O_4 ($K = -12$ kJ/m³ [18]) and $D_{sp} \approx 16$ nm for CoFe_2O_4 ($K = +180$ kJ/m³ [18]). Hence, for a typical size distribution as shown in Fig. 4, one can expect that all particles in a Fe_3O_4 -based ferrofluid have superparamagnetic properties whereas in CoFe_2O_4 -based ferrofluids, larger particles within the size distribution are ferromagnetic. The magnetization of dilute CoFe_2O_4 ferrofluids still exhibits Langevin-type behavior due to the Brownian relaxation process, in which the ferromagnetic particles rotate in order to align their magnetic moment along an external field and restore isotropic orientational distribution by thermally activated rotational diffusion as the field is switched off. The Brownian relaxation time is controlled by the viscous drag that the matrix exerts on the particle,

$$\tau_B = \frac{3V\eta}{k_B T}, \quad (4)$$

and depends explicitly on the viscosity η of the matrix [19]. Obviously, the magnetization properties of a ferrofluid and a derived ferrogel are not necessarily identical if a significant volume fraction of the magnetic particles is ferromagnetic. If we, for the sake of clarity, assume that the crosslinked polymer network gel has been functionalized by a ferrofluid made up of Néelian magnetic particles only and with a sufficiently dilute dispersion of magnetic particles so that particle-particle interaction can be neglected, we would consequently expect superparamagnetic behavior of the ferrogel also. However, if the ferrogel has been functionalized by a ferrofluid with a significant amount of ferromagnetic particles which exhibit superparamagnetic behavior only as long as Brownian relaxation is allowed by the matrix, we expect a strong deviation from superparamagnetism whenever those Brownian particles become mechanically linked to the polymer network in the ferrogel. It can further be expected that if the ferromagnetic particles are aligned with their low-energy anisotropy axis along an applied external field prior to polymerization an uniaxial magnetic gel is formed in which the anisotropy of the magnetic particles contribute to the overall magnetic anisotropy of the ferrogel.

In the following the magnetization properties of a CoFe_2O_4 ferrofluid will be investigated. The magnetic behavior of isotropic and anisotropic ferrogels will then be explored and discussed with reference to the underlying ferrofluid. Finally, the potential of an anisotropic CoFe_2O_4 ferrogel as a soft actuator, which is capable to conduct a torsional deformation under the influence of a homogeneous magnetic field, will be demonstrated.

III. EXPERIMENT

The synthesis of the ferrogels follows the route described by Zrínyi *et al.* [1]. The ferrogel was prepared by mixing a solution of polyvinyl alcohol (PVA, MERCK-Suchardt, [PVA]=1,6 mol/l), the crosslinking agent glutardialdehyde (GDA, Lancaster, [PVA]/[GDA]=400) and a ferrofluid based on cobalt ferrite, water and glycerin (Sustech GMBH, Darmstadt). The volume fraction of the ferrofluid in the ferrogel was 5%. By introducing nitric acid, the crosslinking reaction was initiated. For preparation of magnetically anisotropic ferrogels, the solution was exposed to homogeneous magnetic fields of 0.1 T or 1 T during the gelation. The particle size distribution of the ferrofluid has been investigated using a JEOL JEM 200CX transmission electron microscope (TEM). The TEM sample was prepared by immersing a TEM grid into a sample of the ferrofluid diluted by water.

Magnetization measurements at room temperature were performed with a Lakeshore vibrating sample magnetometer (VSM) Model 7400. The available field for this magnetometer is -2 T to 2 T.

For temperature-dependent magnetization measurements an Oxford MagLab VSM CF-12 with a field range from -12 T to 12 T was used.

To collect field cooling field warming (FC-FW) and zero field cooling field warming (ZFC-FW) data, a Quantum Design Physical Properties Measurement System (PPMS) has been utilized. During the FC of the sample a field of 1 T was applied. The FW was performed in a field of 0.01 T.

In addition, the ferrogels macroscopic structure has been investigated by light-microscopy using a Leitz Metallux 3 optical microscope.

IV. CHARACTERIZATION OF THE FERROFLUID

The particle size distribution of the CoFe_2O_4 ferrofluid has been studied by transmission electron microscopy. A typical TEM micrograph of the magnetic particles in the ferrofluid is presented in Fig. 1. By measuring more than 1000 particles, the particle size distribution shown in Fig. 2 was obtained. The particle size distribution can be well described by a log-normal distribution, $P_{LN}(D)$, characterized by a median $D_0 = 7$ nm and a distribution width $\sigma = 1.5$. These results are in good agreement with small angle x-ray scattering (SAXS) measurements on the same ferrofluid, from which values of $D_0 = 7.4$ nm and $\sigma = 1.7$ were obtained [20].

The magnetization measurements at room temperature revealed superparamagnetic behavior, as expected, Fig. 3. By fitting the experimental data with a superposition of Langevin functions, weighted by a log-normal particle size distribution,

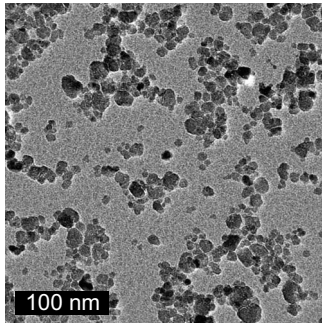


FIG. 1. A typical TEM micrograph of the CoFe_2O_4 -based ferrofluid.

bution, the parameters $D_0=9$ nm and $\sigma=1.5$ were obtained, in reasonable agreement with the TEM and SAXS results.

From these three independently measured parameters, average values $D_0=7.8$ nm and $\sigma=1.57$ were calculated and used to obtain the distribution of the incremental volume fraction of each particle size, $P_V(D) = P_{LN}(D)D^3 / \int P_{LN}(D)D^3 dD$, shown in Fig. 4. With a critical size of $D_{sp}=16$ nm (see previous paragraph) for CoFe_2O_4 , a total volume fraction of $\approx 40\%$ of the particles is expected to be ferromagnetic, but nevertheless exhibits Langevin-type magnetization due to Brownian relaxation in the ferrofluid. Further experimental evidence for a significant contribution of Brownian relaxation is obtained from ZFC-FW and FC-FW measurements, presented in Fig. 5. Upon cooling, the liquid matrix of the ferrofluid solidifies at about 180 K, confirmed by calorimetric measurements, so that the magnetic particles adhere to the matrix. For Néelian particles with $D < D_{cr}$, locking of the particles caused by the solidification of the matrix has no influence on the magnetization behavior. However, the Brownian relaxation of larger particles is severely hampered by an increasing viscosity with decreasing temperature. In fact, below the melting point, the solid matrix prohibits any rotational relaxation of the particles whenever a sufficient coupling to the matrix is given. To verify the loss of rotational degree of freedom, a ferrofluid with random orientation of the particle's magnetic moments at room temperature has been cooled to 4 K without application of a magnetic field (ZFC). The frozen-in ran-

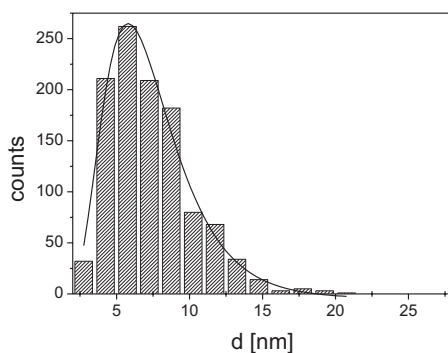


FIG. 2. The particle size distribution obtained from a measurement of more than 1000 particles and the corresponding fit by a log-normal distribution with a median $D_0=7$ nm and a width $\sigma=1.5$.

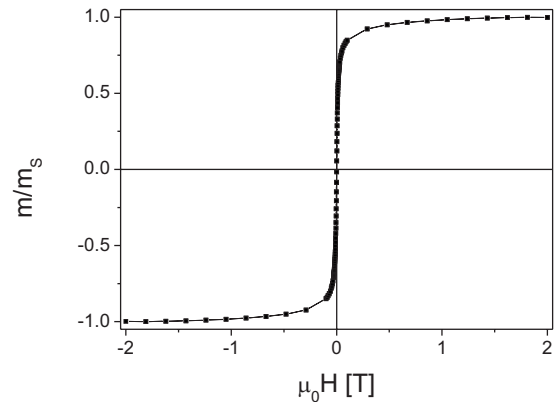


FIG. 3. The superparamagnetic magnetization properties of the ferrofluid can be described by a superposition of Langevin functions, weighted by a log-normal size distribution with a median $d_0=9$ nm and distribution width $\sigma=1.5$ (solid line).

domly oriented magnetic moments will not be able to reorient along an applied field (0.01 T). By approaching the melting point of the matrix material, the locked particles regain their rotational and translational degrees of freedom allowing Brownian relaxation, indicated by the sudden jump in the FW data at 180 K. Particles, which were prealigned nearly parallel during cooling in a high field of 1 T (FC), will reorient according to the equilibrium angular distribution associated with the smaller field of 0.01 T applied during FW, resulting in a steep decrease of the overall magnetic moment at 180 K.

In order to independently estimate the fraction of Brownian particles based on their magnetic properties, temperature-dependent magnetization measurements below the melting point of the matrix were performed. As shown in Fig. 6 the magnetization exhibits a temperature-dependent hysteretic

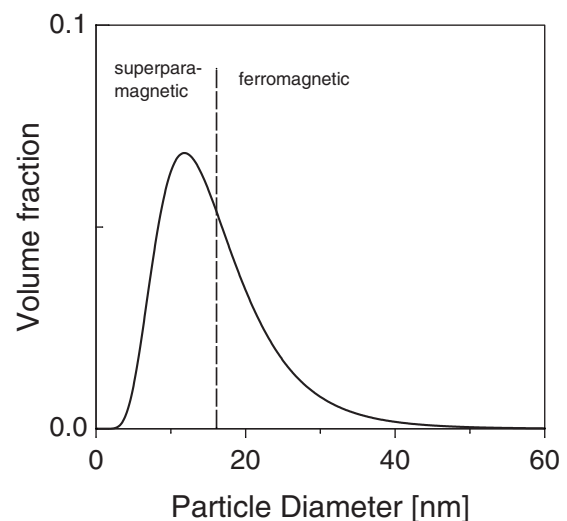


FIG. 4. Incremental volume fraction as a function of particle diameter for a log-normal size distribution with median $D_0=7.8$ nm and distribution width $\sigma=1.57$. The critical diameter of 16 nm for CoFe_2O_4 , which separates the superparamagnetic (≈ 60 Vol %) from the ferromagnetic (≈ 40 Vol %) regime, is indicated.

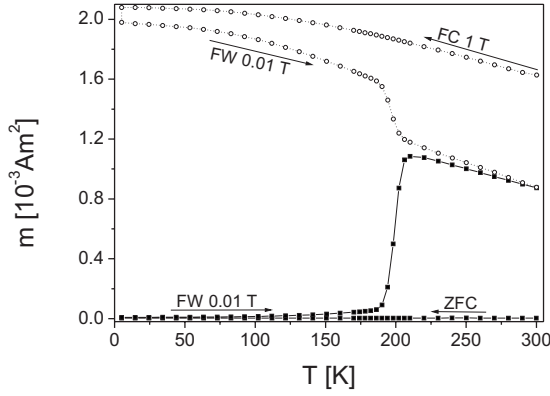


FIG. 5. FC-FW (open circles) and ZFC-FW (full squares) measurements of the ferrofluid. Field cooling was performed at $\mu_0 H = 1$ T followed by the field warming at $\mu_0 H = 0.01$ T. The ZFC-FW measurement is approaching the FC-FW data at 180 K which is the melting point of the matrix.

behavior. From these measurements, the temperature dependence of the reduced remanent magnetization $\alpha(T) = m_R(T)/m_S(T)$ was obtained, shown in Fig. 7. Superparamagnetic particles do not exhibit remanence, $\alpha|_{sp} = 0$, implying that the fraction of smaller particles with Néelian relaxation do not contribute to the remanent magnetization at all. However, the larger particles exhibit ferromagnetic behavior with finite remanence since Brownian relaxation is prohibited by the solid matrix. For ideal single domain particles, the reduced remanence can be calculated as $\alpha(T=0 \text{ K}) = 0.831$ for cubic anisotropy and $K > 0$ [21] as reported for CoFe_2O_4 [18]. By extrapolation to 0 K, where all particles are expected to be ferromagnetic, we obtained a reduced remanence of $\alpha = 0.75$, which is slightly below the theoretical value given above, indicating that a contribution of surface effects [22] and/or particle-particle interactions [23,24] might be present. Hence, this value should be regarded as an effective $\alpha(T=0)$ value characteristic of the ferrofluid under investigation. At any temperature, the volume fraction of ferromagnetic particles X_{fm} can now be

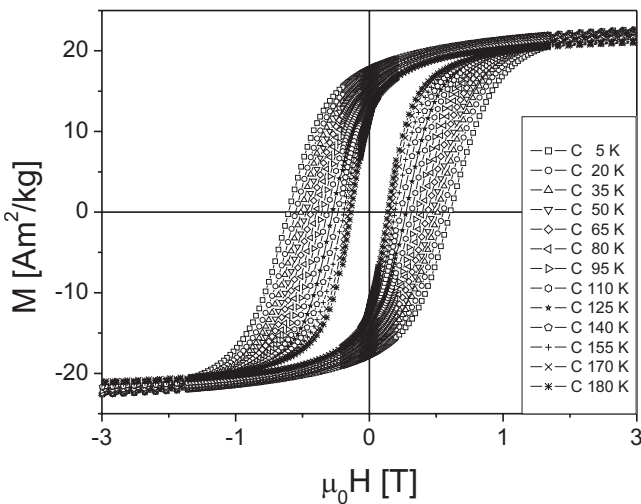


FIG. 6. VSM magnetization measurements of the ferrofluid for different temperatures below the melting point.

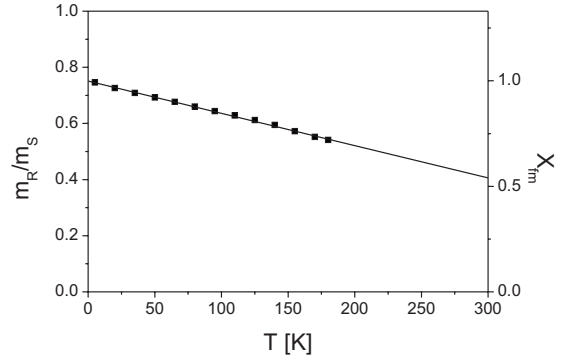


FIG. 7. Linear fit of the temperature-dependent reduced remanence data. The analysis of the presented temperature-dependent magnetization data leads to a simple assessment of the Brownian contribution on the magnetization at room temperature, leading to a volume fraction of about 0.54.

roughly estimated from the reduced remanence α_T , using

$$X_{fm} = \frac{\alpha(T)}{\alpha(0 \text{ K})}. \quad (5)$$

As shown in Fig. 7, the reduced remanence decreases linearly in the given temperature range, which—in this simplified model—implies that the volume fraction of ferromagnetic particles (i.e., particles in the size regime of Brownian relaxation) apparently decreases linearly with temperature. If we assume that this tendency persists, we can extrapolate to the volume fraction of Brownian particles at 300 K obtaining a value of 0.54. According to the calculations by Walker *et al.* [17], the reduced remanence will not continue to decrease linearly but will show concave-up curvature so that the extrapolated value of 0.54 is a lower bound for the volume fraction of ferromagnetic particles.

The temperature dependence of the coercive field is shown in Fig. 8 with an extrapolated value $\mu_0 H_c(T=0 \text{ K}) = 0.91$ T. This result is significantly smaller than the theoretical coercivity $\mu_0 H'_c(0 \text{ K}) = 0.321 \times 2K/M_s = 2.5$ T [17] with $K(0 \text{ K}) = 1.96 \times 10^6 \text{ J/m}^3$ and $M_s(0 \text{ K}) = 5 \times 10^5 \text{ A/m}$ [25], indicating that magnetization reversal is not determined by coherent rotation of the magnetic moments as assumed in the

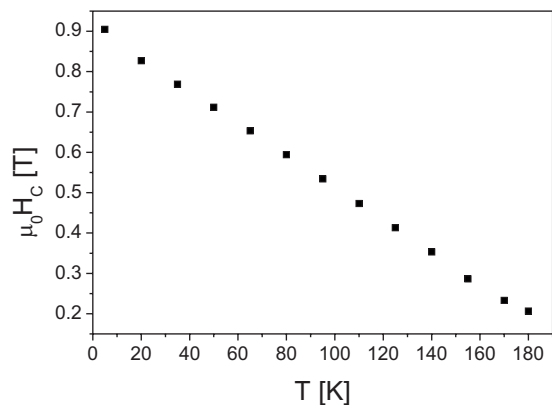


FIG. 8. The temperature-dependent coercivity of the ferrofluid below the melting point.

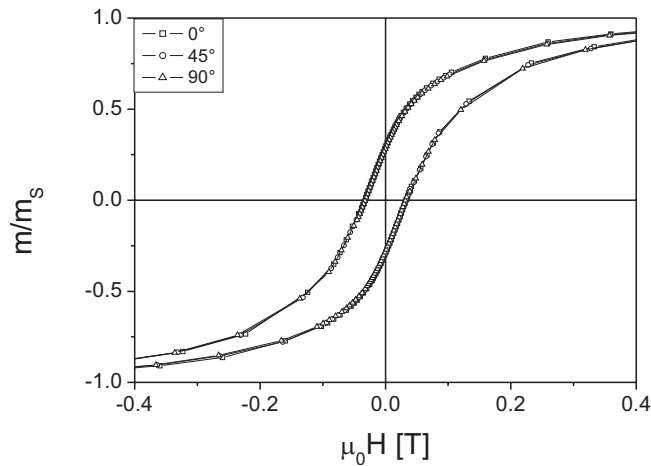


FIG. 9. The magnetization measurements of the CoFe_2O_4 -based ferrogel exhibit a hysteretic behavior. Furthermore, the hysteresis is independent of the orientation between the sample and the magnetic field which indicates isotropic behavior due to the random orientational distribution of the magnetic particles.

model. As a further result, the coercive field decreases much faster with increasing temperature than the reduced remanence, in agreement with [17].

As a first conclusion, the preceding characterization provides ample evidence that a volume fraction of approximately 50% of the magnetic particles in the CoFe_2O_4 ferrofluid are ferromagnetic and exhibit Brownian relaxation behavior at room temperature, which should result in different magnetization properties of derived ferrogels as compared to the ferrofluid.

V. MAGNETIC PROPERTIES OF A COBALT-FERRITE-BASED FERROGEL

In Fig. 9 the magnetization versus field behavior of a CoFe_2O_4 -based ferrogel is presented. In contrast to the superparamagnetic behavior of the ferrofluid (Fig. 3), a hysteresis with $\mu_0 H_C = 33$ mT and $m_R/m_S = 0.29$ is observed. In analogy to the measurements of the ferrofluid below the melting point of the matrix in the previous chapter, we suggest that this hysteresis is caused by a partial inhibition of free rotation of larger particles ($D > D_{cr}$) due to a mechanical interaction with the polymer gel network. By applying an external homogeneous field, the individual ferromagnetic particles start to rotate into the direction of the field. This rotation will proceed until the magnetic torque is balanced by an increasing countertorque due to the elastic deformation of the gel matrix. As a consequence, the magnetization of the ferrogel can be assumed to reflect the elastic properties of the gel matrix. However, the reduced remanence (measured in absence of an external field and hence of magnetic torque) is significantly smaller than the extrapolated value 0.41 obtained from the temperature-dependent measurements of the ferrofluid. A possible explanation for this discrepancy could be an increased dipolar interaction between the particles caused by an aggregation during preparation of the ferrogel. In Fig. 10, the magnetization of the ferrofluid and a derived

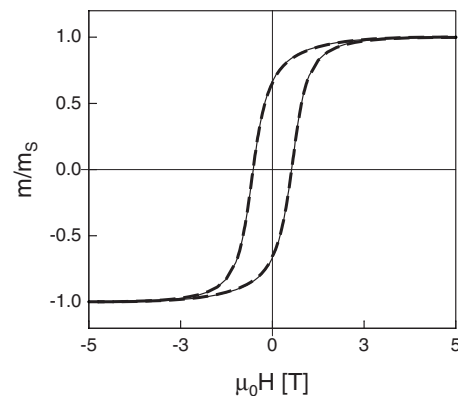


FIG. 10. Magnetization measurements of the CoFe_2O_4 based ferrofluid (solid line) and the isotropic ferrogel (dashed line) at a temperature of 77 K.

ferrogel are shown for comparison, both at $T = 77$ K. At this temperature, the matrix in both samples is frozen and prevents any particle rotation irrespective of its nature (i.e., fluid or gel). Within experimental error, the magnetization curves of both samples are identical so that a significant change in the dipole-dipole interaction caused by aggregation during the preparation of the ferrogel compared to the ferrofluid can be ruled out. We therefore conclude, that the reduced remanence of the ferrogel has its origin in the elastic properties of the matrix. As a matter of fact, in the limit of an extremely soft gel matrix, one would expect that the magnetic behavior of the gel should approach the behavior of the ferrofluid.

As shown in Fig. 9, the magnetization behavior of the ferrogel is independent of the orientation of the gel relative to the magnetic field, which confirms the expected isotropic behavior of randomly oriented magnetic particles. However, if the ferrogel is exposed to a homogeneous magnetic field during polymerization, a growing magnetic anisotropy is introduced at increasing field, Fig. 11 and Fig. 12. In the

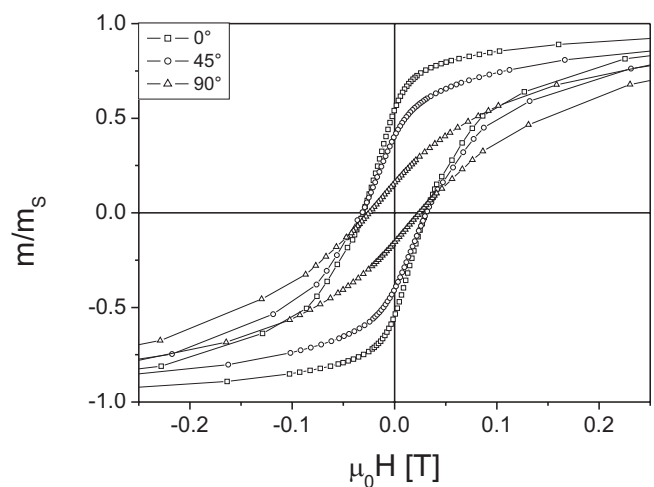


FIG. 11. A CoFe_2O_4 ferrogel, which was polymerized under an applied homogeneous field of 1 T, exhibits different hysteretic curves for measurements with different angles (0° , 45° , 90°) between the direction of the sample's magnetic texture and the magnetic field.

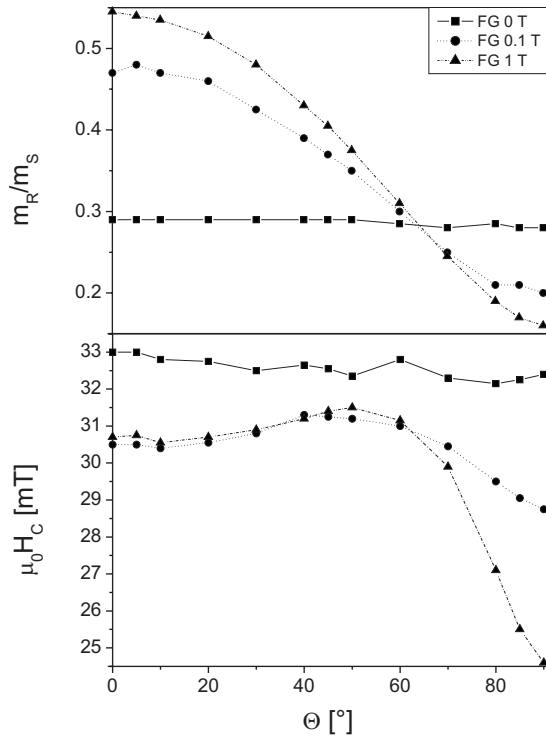


FIG. 12. m_R/m_S and H_C as function of angle Θ between the sample and the magnetic field are shown for the isotropic ferrogel (squares) and anisotropic ferrogels, exposed to a homogeneous field of 0.1 T (circles) and 1 T (triangles) prior to polymerization. With increasing field, a significant magnetic anisotropy is introduced.

uniaxial ferrogel (1 T), the reduced remanence of $m_R/m_S = 0.54$ at $\Theta = 0$ is much larger as compared to the isotropic ferrogel and decreases continuously to 0.16 at $\Theta = 90^\circ$. The coercive field of the uniaxial ferrogel is smaller at all angles and exhibits a shallow maximum at $\Theta = 50^\circ$ followed by a decrease to a minimum at 90° . Such a maximum is not expected for coherent rotation of a single domain particle. However, a similar angular dependency has been predicted for the magnetization reversal of an isotropic ellipsoid and has been attributed to the crossover in the critical nucleation field from incoherent (curling mode) to coherent rotation of the magnetization [26]. Although the physical conditions in the uniaxial ferrogel are certainly far away from the ideal situation assumed in theory (cubic anisotropy, unknown angular distribution, and dipolar interaction) it can be assumed that a variety of yet unknown microscopic processes of mag-

netization reversal exist for the CoFe_2O_4 particles in the ferrogel. Yet, it is not possible to further analyze the angular dependence of the coercive field beyond the qualitative identification of the induced macroscopic magnetic anisotropy. The angular dependence of the reduced remanence resembles the behavior expected for uniaxial anisotropy, despite the cubic anisotropy of CoFe_2O_4 . A possible explanation would be the growth of linear chains of magnetic particles in the applied field prior to polymerization of the gel which would give rise to uniaxial shape anisotropy. In Fig. 13, micrographs of an isotropic and two anisotropic ferrogels, obtained by light microscopy, are shown. Obviously, the ferrogel is inhomogeneous on a macroscopic scale, as has been reported for magnetite-based ferrogels before [7,9,10]. While spherical aggregates are found in the isotropic gel, the magnetic phases were highly elongated in the uniaxial ferrogels. Hence, we may expect—though there is no direct evidence given—that the magnetic nanoparticles may also have formed linear chains which give rise to a uniaxial magnetic anisotropy. An upper limit for the magnitude of this anisotropy energy can be estimated, assuming a homogeneously magnetized rod of CoFe_2O_4 with infinite length. The expected shape anisotropy constant has a value of $K_{\text{rod}} = \mu_0 M_s^2 / 4 = 5.7 \times 10^4 \text{ J/m}^3$ which is however much smaller than the magnetocrystalline anisotropy constant of $K = 3.5 \times 10^5 \text{ J/m}^3$. Therefore, we can assume that irrespective of a possible chain formation in the magnetic gel, the anisotropy in the uniaxial magnetic gel is dominated by the magnetocrystalline anisotropy of the CoFe_2O_4 particles.

If the particles and their magnetic moments are randomly oriented in the ferrogel, the magnetic torque on all individual particles cancel each other. However, in a uniaxial magnetic ferrogel, the magnetic torques applied to the individual particles and their rotation as well as the induced deformation of the surrounding matrix is expected to add to a macroscopic torsion of the ferrogel. To demonstrate this effect, a ferrogel cylinder was prepared and polymerized in a magnetic field of 1 T. The sample was attached at the top end but otherwise free to move and was exposed to a homogeneous magnetic field (up to 0.2 T) orthogonal to the magnetic texture, Fig. 14. A torsion around the cylinder axis by about $\pm 90^\circ$ was observed in a field of 0.2 T, depending on the direction of the applied field, Fig. 14. Furthermore, the torsion reversibly disappeared when the applied field was removed again, Fig. 14. In a similar way, a magnetically textured ferrogel, filled in the gap between a rigid hollow cylinder and a centered rod exhibited rotational actuator behavior in a homogeneous

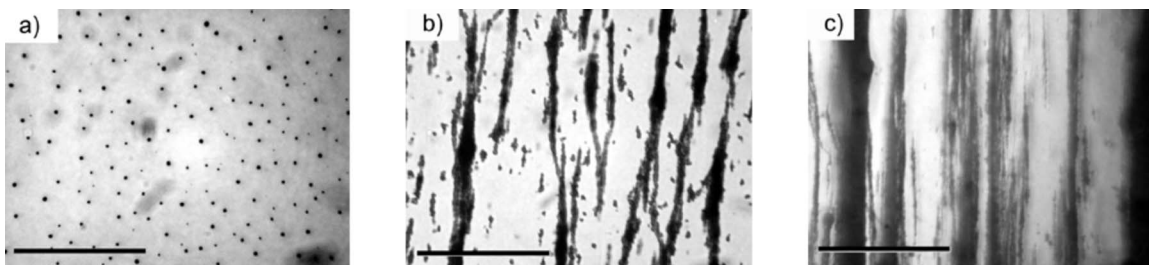


FIG. 13. Optical microscopy images of the CoFe_2O_4 ferrogel which was polymerized (a) without external field and with a homogeneous magnetic field of (b) 0.1 T and (c) 1 T. Scale bars indicate $100 \mu\text{m}$.

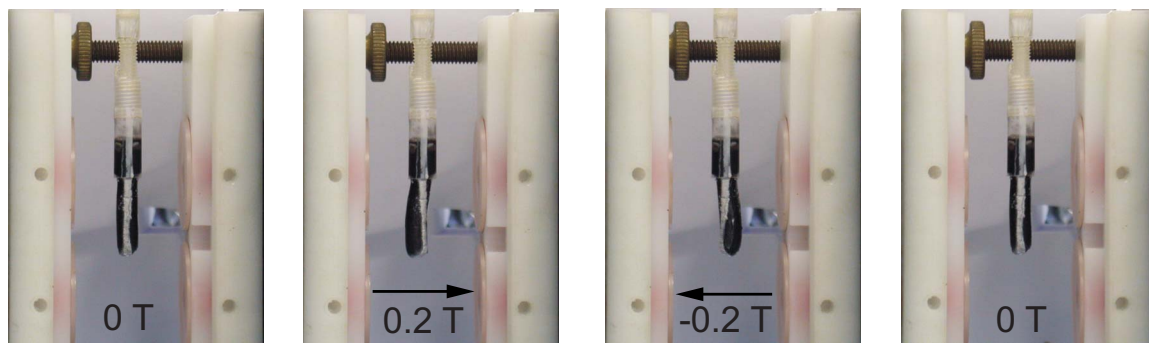


FIG. 14. (Color online) The four pictures present a magnetically textured ferrogel cylinder under the influence of various magnetic fields. The net magnetic moment is directed perpendicular to the image plane and to the applied magnetic field.

magnetic field perpendicular to the anisotropy axis. The combination of a soft elastic matrix and magnetic texture of the dispersed magnetic particles provides the potential of large torsional and rotational deformation. This property adds a degree of freedom—in addition to the translational force in a magnetic field gradient—to the design of ferrogel-based soft matter actuators.

VI. CONCLUSION

The magnetic properties of CoFe_2O_4 -based ferrogels are distinctly different from the properties of the corresponding ferrofluid since a significant fraction of magnetic particles has volumes too large for Néelian relaxation, but Brownian

relaxation—allowed in the ferrofluid—is restricted by the mechanical coupling to the ferrogel network. This particular property enables the synthesis of magnetically textured ferrogels by applying an external field during gel polymerization. The combination of magnetic texture and soft elastic behavior of the gel matrix can be used for application as torsional and rotational soft actuators.

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