How particle interactions and clustering affect the dynamic magnetic susceptibility of ferrofluids

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Abstract

The effects of magnetic interparticle interactions on the frequency-dependent, dynamic magnetic susceptibility of ferrofluids are summarised with reference to recent theoretical and simulation studies. With weak to moderate interactions, the dynamic magnetic susceptibility qualitatively resembles the classic Debye prediction, but with features shifted to lower frequencies as the particle concentration and/or interaction strength are increased. This shows that the mutual spatial and orientational correlations arising from the interactions lead to slower collective motions. With strong interactions, and at low concentrations, the particles form chain-like and ring-like clusters, and this leads to distinct collective and intracluster motions being apparent in the dynamic magnetic susceptibility. The initial formation of chains increases the static susceptibility, while the subsequent formation of rings decreases it.

Keywords: ferrofluids, dynamic magnetic susceptibility, chain aggregates, ring-like clusters, Brownian dynamics simulations

1. Introduction

One of the most attractive applications of ferrofluids is as heating media in magnetic hyperthermia treatments [1, 2, 3, 4, 5]. Here, the ferrofluid is exposed to an ac external magnetic field, and energy is dissipated as heat. The heating rate is proportional to the imaginary, or out-of-phase, component of the dynamic magnetic susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ [6, 7]. $\chi(\omega)$ characterises the *linear* response of the magnetisation to a *weak* ac field [8]. The peak frequency in the imaginary part is controlled by the intrinsic rotation time of the magnetic nanoparticles [9, 10], and the interactions between the particles. Concerning the interactions, two different types of non-linear collective effects are worth mentioning here. The first one is the hydrodynamic interaction between ferroparticles, arising from perturbations of the hydrodynamic fields influenced by the other rotating particles. Since the solution of this problem is unknown, the common approach [11, 12, 13] is to replace the viscosity η of the carrier liquid in the expression for the Brownian relaxation time $\tau_{\mathrm{B}} \propto \eta$ with some effective viscosity of the suspension, which increases with the ferroparticle concentration. The second effect results from the collective long-range magnetic interparticle interactions between ferroparticle magnetic moments, which is the main focus of this contribution. The theoretical framework is based on Brownian dynamics, i.e., a particle's magnetic dipole moment is fixed within the body frame, and reorientation happens by particle rotation [14]; Néel rotation is not considered here.

2. Model and theoretical methods

The ferrofluid is modelled as a system of N particles in a volume V at temperature T. The number concentration of ferroparticles is $\rho = N/V$. Each particle has hydrodynamic diameter d and magnetic dipole moment m. The ferroparticles interact with an external magnetic field through the Zeeman energy, and with each other through the dipole-dipole interaction potential. The total volume fraction φ, the dipolar coupling constant λ characterising the strength of the interparticle interactions, and the Langevin (initial) susceptibility χ_{L} are given by

$$\varphi = \frac{\pi\rho d^3}{6}, \tag{1}$$

$$\lambda = \frac{\mu_0 m^2}{4\pi d^3 k_{\mathrm{B}} T}, \tag{2}$$

$$\chi_{\mathrm{L}} = \frac{\mu_0 \rho m^2}{3 k_{\mathrm{B}} T} = 8\lambda\varphi, \tag{3}$$

where μ_0 is the vacuum permeability, $k_{\mathrm{B}}T$ is the thermal energy, and k_{B} is Boltzmann's constant.

Only the Brownian relaxation mechanism is considered here, and so the magnetic moment is fixed inside the body of the particle. In other words, the energy of the magnetic crystallographic anisotropy is considered much larger than

*16th International Conference on Magnetic Fluids, Granada, Spain, 2023

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the thermal energy. For a particle in a liquid with viscosity η, the Brownian rotation time of a single particle is given by

$$\tau_{\rm B} = \frac{\pi\eta d^3}{2k_{\rm B}T}. \tag{4}$$

Next, it is assumed that the container of the ferrofluid is shaped like a highly elongated ellipsoid, and a weak linearly polarised probing ac magnetic field $H(t) = H_0 e^{i\omega t}$ is applied along the principal axis of the ellipsoid. The demagnetisation effect can be neglected for this configuration. If the field strength amplitude is weak, meaning $\mu_0 m H_0/k_{\rm B}T \ll 1$, then the magnetisation is within the linear response regime. This is written

$$M(t) = \chi(\omega)H(t) = M_0 e^{i(\omega t-\delta)}, \tag{5}$$

where $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ is the (initial) dynamic magnetic susceptibility. The magnetic response has an in-phase component $H_0\chi'(\omega) = M_0\cos\delta$, and an out-of-phase component $H_0\chi''(\omega) = M_0\sin\delta$ due to the fact that the ferroparticles cannot rotate infinitely fast. The phase lag is $0 \le \delta = \arctan(\chi''/\chi') \le \pi/2$ (the magnetisation lags behind the field). The power loss is proportional to the imaginary part of the dynamic magnetic susceptibility, $\chi''(\omega)$ [6].

Linear response theory leads to an important relationship between $\chi(\omega)$ and the time correlation function $C(t)$ of the equilibrium magnetisation fluctuations in zero field [15]:

$$\frac{\chi(\omega)}{\chi(0)} = 1 + i\omega\int_0^\infty C(t)e^{i\omega t}dt. \tag{6}$$

$C(t)$ is defined by

$$C(t) = \frac{\langle \boldsymbol{M}(t)\cdot\boldsymbol{M}(0)\rangle}{\langle \boldsymbol{M}(0)\cdot\boldsymbol{M}(0)\rangle}, \tag{7}$$

where $\boldsymbol{M}(t) = \sum_{j=1}^N \boldsymbol{m}_j(t)$ is the instantaneous magnetisation of the sample, $\boldsymbol{m}_j(t)$ is the dipole moment on particle j, and of course $\langle \boldsymbol{M}(t)\rangle = 0$. This provides a convenient route to calculating $\chi(\omega)$ from Brownian dynamics (BD) simulations, as has been demonstrated in recent work [16, 17, 18, 19].

2.1. Debye model

In the ideal case of non-interacting particles – which corresponds to the original Debye theory [20, 21] – the time correlation function is simply

$$C_{\rm D}(t) = e^{-t/\tau_{\rm B}}, \tag{8}$$

and the real and imaginary parts of the dynamic magnetic susceptibility are then given by the well-known Debye expressions

$$\chi'_{\rm D}(\omega) = \frac{\chi_{\rm L}}{1+\omega^2\tau_{\rm B}^2}, \tag{9a}$$

$$\chi''_{\rm D}(\omega) = \frac{\chi_{\rm L}\omega\tau_{\rm B}}{1+\omega^2\tau_{\rm B}^2}, \tag{9b}$$

where the static susceptibility $\chi_{\rm D}(0)$ is equal to the Langevin value $\chi_{\rm L}$. Note that $\chi''_{\rm D}(\omega)$ shows a maximum at the frequency $\omega_{\max} = \tau_{\rm B}^{-1}$, which suggests a useful way of measuring the effective relaxation time(s) in other situations.

Obviously, the Debye model is applicable only for very dilute ferrofluids, under conditions where the interparticle distance greatly exceeds the particle size, and the effects of interparticle interactions may be ignored. In this case, the susceptibility appears to be a linear function of the ferroparticle concentration, as in Eq. (3). Taking the magnetic interparticle interactions into account results in a non-linear concentration dependence of the susceptibility, and this dependence is the subject of the following discussion.

2.2. Modified mean-field model

There have been many attempts to include the effects of magnetic interactions [11, 12, 22, 23, 24, 25, 26]. The basic phenomenology is that interactions between particles lead to more collective, and hence slower, motions. The correction term to the Debye formulae, quadratic in the ferroparticle concentration, was proved mathematically in Ref. [27] using the method known as the 'modified mean-field model' (MMF), although it is actually a perturbative approach. The result for the dynamic magnetic susceptibility can be expressed entirely in terms of $\chi_{\rm D}(\omega)$ [13, 27]:

$$\chi'_{\rm MMF}(\omega) = \chi'_{\rm D}(\omega) + \frac{1}{3}\left\{[\chi'_{\rm D}(\omega)]^2 - [\chi''_{\rm D}(\omega)]^2\right\}; \tag{10a}$$

$$\chi''_{\rm MMF}(\omega) = \chi''_{\rm D}(\omega)\left[1+\frac{2}{3}\chi'_{\rm D}(\omega)\right]. \tag{10b}$$

Note that the static susceptibility is given by $\chi_{\rm MMF}(0) = \chi_{\rm L}(1+\chi_{\rm L}/3)$, which is a well-known result derived long ago [28, 29, 30]. These results have been tested against both experimental [13, 27, 31] and simulation data [16, 17, 32], and they are reliable as long as the Langevin susceptibility $\chi_{\rm L} \lesssim 1$.

2.3. Modified Weiss model

The so-called 'modified Weiss model' (MW) is an extension of the classic Weiss model in a way that removes a divergence of the susceptibility at $\chi_{\rm L} = 3$, which does not occur in real ferrofluids. The resulting MW expression for the dynamic susceptibility is

$$\chi_{\rm MW}(\omega) = \frac{\left(1+\frac{1}{3}\chi_{\rm L}\right)\chi_{\rm D}(\omega)}{1+\frac{1}{3}\chi_{\rm L}-\frac{1}{3}\chi_{\rm D}(\omega)}, \tag{11}$$

from which the real and imaginary parts can be separated out easily. Importantly, the static low-frequency limits of the MMF and the MW models coincide. Comparisons with computer-simulation results show that the MW theory is marginally better than the MMF theory, and that the predictions are reliable for systems with $\chi_{\rm L} \lesssim 3$ [17].

The main prediction of both dynamic models is the shift of the maximum of the imaginary part, $\chi''(\omega)$, towards frequencies lower than the Debye model prediction $\omega_{\max} = \tau_{\rm B}^{-1}$. This effect, elucidated by computer simulation and demonstrated in Fig. 1, leads to the conclusion that the collective magnetic correlations in a ferroparticle ensemble slow down the dynamic magnetic response; essentially, $\tau_{\rm B}$ should be replaced by an effective rotation time $\tau_{\rm eff} > \tau_{\rm B}$. For monodisperse ferroparticles, the MMF prediction for the effective collective relaxation time is $\tau_{\rm eff} = \tau_{\rm B}\,(1 + \chi_{\rm L}/3)$, which is linear in the ferroparticle concentration.

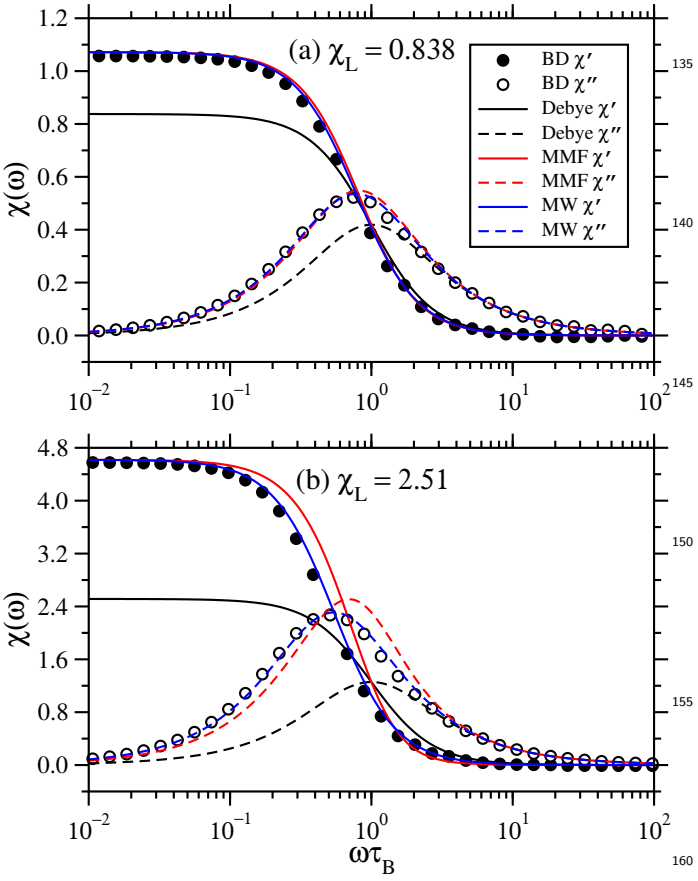

Figure 1: The real and imaginary parts of the dynamic magnetic susceptibility of monodisperse ferrofluids with $\lambda = 1$. The computer simulation data are shown with symbols for the volume concentrations (a) $\varphi = 0.105$, $\chi_{\rm L} = 0.838$) and (b) $\varphi = 0.314$, $\chi_{\rm L} = 2.51$. The lines are the predictions from the Debye (black), MMF (red), and MW (blue) theories.

The general conclusion of this Section is that the magnetic dipole interactions reduce the relaxation rate due to increased correlations between magnetic nanoparticles, leading to collective motions. This conclusion is supported by several other studies [33, 34, 35, 36].

3. Structural 'chain–ring' crossover

The perturbative MMF and MW theories are valid only for weak to moderate interparticle interactions, when the corrections to the Debye model are dependent on the product of the dipolar coupling constant and the volume concentration, i.e., the Langevin susceptibility $\chi_{\rm L}$. By contrast, the magnetic properties of ferrofluids become explicitly dependent on λ for strongly interacting ferroparticles [16]. At low concentrations $\varphi \ll 0.1$, and with high dipolar coupling constant $\lambda > 4$, dipolar particles self-assemble to form flexible chain-like clusters [37, 38, 39, 40, 41]. The physical reason for this effect is the energetically favourable configuration of neighbouring ferroparticles with a 'head-to-tail' orientation of their magnetic moments. The association energy prevails over the loss in entropy, and for each φ and λ, a distinct equilibrium distribution of chains is established [42]. At low concentrations, and with increasing λ to about 6, the impact is that the formation of chains increases the static initial magnetic susceptibility $\chi(0)$ faster than $\chi_{\rm L}$, because of the strong orientational correlations between particles in a given chain. This effect is illustrated in Fig. 2, where theoretical predictions are compared with BD simulation results at $\varphi = 5.24 \times 10^{-4}$, but it has also been confirmed experimentally [43]. Above $\lambda \simeq 7$, the chains start to close up and form ring-like clusters. The driving force of the crossover from chains to rings (for four or more particles) is the association energy when the chain ends are closed, and the magnetic flux becomes closed inside the ring-like cluster [44]. The chain closing is accompanied by a loss of conformational entropy of the free chain ends, but the energy change prevails with strong dipolar coupling. This structural crossover, elucidated by means of computer simulations [41], was studied theoretically in detail in Refs. [45, 46]. A ring-like cluster has a very low net dipole moment, it behaves like a 'magnetic hole', and so it does not respond to a weak applied magnetic field. Thus, the crossover to ring-like clusters is accompanied by a rapid decrease of $\chi(0)$ [19, 45], as is shown in Fig. 2.

A recent study on very dilute systems with $\varphi = 5.24 \times 10^{-4}$ and $1 \le \lambda \le 8$ uncovered pronounced effects of clustering on the dynamic magnetic susceptibility as well [19]. Examples are shown in Fig. 3. $\chi(\omega)$ is divided by the Langevin susceptibility, which with the stated value of φ, and $5 \le \lambda \le 8$, is in the range $0.0209 \le \chi_{\rm L} \le 0.0335$. With $\lambda = 5$, there is a single peak in $\chi''(\omega)$ in the region of $\omega\tau_{\rm B} = 1$ [Fig. 3(a)]. The dynamic response is essentially that of weakly interacting but unclustered particles, and it is close to the Debye behaviour (9). With increasing λ, the formation of chains leads to an increase in the effective rotation time, and hence $\chi''(\omega)$ shows a low-frequency peak, well below the single-particle frequency. This is quite clear in Fig. 3(b) with $\lambda = 6$, where two clear peaks in $\chi''(\omega)$ are in evidence: one at $\omega\tau_{\rm B} \simeq 0.03$ and the other at $\omega\tau_{\rm B} \simeq 1$. The obvious interpretation is that the lower-frequency peak arises from short chains, and the higher-frequency peak arises from unclustered but interacting particles. With $\lambda = 7$ [Fig. 3(c)], the predominant peak in $\chi''(\omega)$ is at $\omega\tau_{\rm B} \simeq 0.002$, which again must be due to chains, but now longer ones. It is worth mentioning

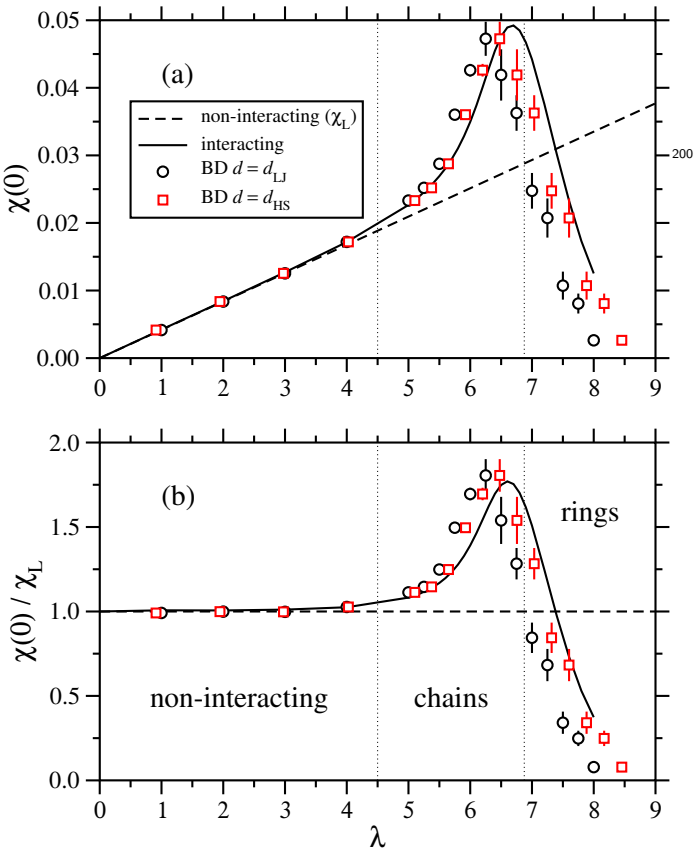

Figure 2: (a) The static susceptibility $\chi(0)$, and (b) $\chi(0)$ divided by the Langevin susceptibility χ_L, as functions of the dipolar coupling constant λ, for ferrofluids with particle volume fraction $\varphi = 5.24 \times 10^{-4}$. The dashed black lines are for non-interacting particles [$\chi(0) = \chi_L$], the solid black lines are the predictions of the chain–ring structural transformation theory [45], and the symbols are from BD simulations [19]. The simulation results are shown in two forms, with λ (2) calculated using either a Lennard-Jones particle diameter (d_{LJ}) or an effective hard-sphere diameter (d_{HS}). The dotted black lines show the approximate boundaries between states where the structure is primarily unclustered (non-interacting) particles, chains, and rings.

that the main peak at $\omega\tau_B \simeq 1$ has actually disappeared, which means that the portion of unclustered particles can be neglected at $\lambda = 7$. As chains start to close up to form rings at $\lambda = 8$ [Fig. 3(d)], the low-frequency peak at $\omega\tau_B \simeq 0.002$ becomes much less pronounced, due to there being a smaller number of long chains. Interestingly, a high-frequency peak in $\chi''(\omega)$ appears unexpectedly, which turns out to be associated with intracluster, single-particle oscillations. A large dipolar coupling constant means that there are strong torques which act to keep the dipole moments aligned. In conjunction with the Brownian forces on the particle, the time scale of the orientational fluctuations is short as compared to τ_B, and the corresponding signature in the dynamic magnetic susceptibility spectrum appears at high frequency. A dynamical, one-particle theory, including the average torque arising from neighbouring particles, yields an accurate estimate of the characteristic time [19], which is in the region of $0.05\tau_B$ when $\lambda = 8$, and gives a peak at $\omega\tau_B \simeq 20$. Such high-frequency motions must be present whenever there are clusters, but they are only visible in $\chi''(\omega)$ when all of the chains have closed up to form rings, since the chain contribution to $\chi(\omega)$ is so much larger than that of individual particles or rings.

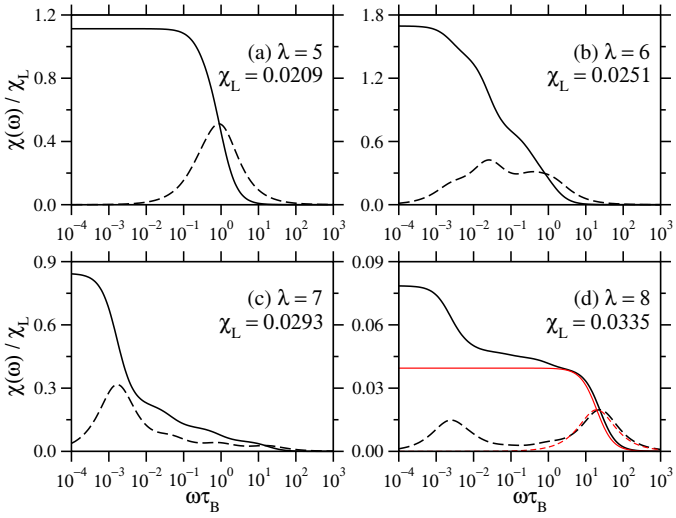

Figure 3: The dynamic magnetic susceptibility $\chi(\omega)$, divided by the Langevin susceptibility χ_L, for ferrofluids with volume fraction $\varphi = 5.24 \times 10^{-4}$: (a) $\lambda = 5$; (b) $\lambda = 6$; (c) $\lambda = 7$; (d) $\lambda = 8$. The solid black line represents the real part, and the dashed black line the imaginary part, from BD simulations. In (d), the solid red line and dashed red line represent the Debye functions with an effective Brownian time $0.0510\tau_B$, and scaled to give the peak height in $\chi''(\omega)$.

4. Summary

In the case of weak to moderate interactions, meaning $\lambda \lesssim 2$ and $\chi_L \lesssim 3$, the peak in the frequency spectrum of the imaginary part of the susceptibility shifts towards lower frequencies, which can be interpreted as the collective response of the ferroparticles being slower than the response of individual, non-interacting particles. In this regime, the effective collective relaxation time is a linear increasing function of the static Langevin susceptibility, meaning that it is proportional to both λ and φ.

Strong magnetic interactions, $4 \lesssim \lambda \lesssim 6$, in a diluted magnetic suspension, $\varphi \lesssim 0.05$, lead to the formation of ferroparticle chains, the presence of which results in a significant increase of the static magnetic susceptibility, and the appearance of an additional peak in the imaginary susceptibility at frequencies much lower than τ_B^{-1}. This peak corresponds to the hydrodynamic rotation of a flexible chain, and it is obviously dependent on the chain length; the longer the chain, the lower the frequency.

Further strengthening of the interparticle interactions, $\lambda \gtrsim 7$, is accompanied by the 'chain–ring' structural crossover, during which the energy decrease on joining the chain ends compensates for the loss in the conformational entropy of

the free chain ends. The magnetic flux becomes closed inside the ring-like cluster, the magnetic moment is approximately zero, and so the ring-like clusters do not contribute to the static magnetic susceptibility. Hence, the 'chain–ring' transformation is accompanied by a huge decrease in the static magnetic susceptibility. The formation of rings with very strong interparticle interactions results in the appearance of a high-frequency peak in the imaginary part of the susceptibility spectrum, due to the strong torques between neighbouring particles, and the disappearance of chains in the cluster distribution.

5. Conclusions

The effects of interparticle dipole-dipole interactions and clustering on the dynamic magnetic susceptibility $\chi(\omega)$ of ferrofluids have been summarised. Examples were taken from recent theoretical and simulation studies, which means that the key physical parameters were well controlled.

Regarding the relevance to experiments, it should be possible to use the theories outlined here to fit functions to measured susceptibility spectra, and determine the particle-size distribution – so-called dynamic magnetogranulometry [47, 48]. Here, the application of models accounting for the interparticle interactions is of major importance for obtaining correct data, since the characteristic relaxation times are dependent not only on the particle size, but also on the sizes of the magnetic moments and the interactions between them. Both the dipolar coupling constant λ and the Brownian rotation time $\tau_{\rm B}$ are proportional to the particle volume ($\sim d^3$), and the effects of particle size on the dynamic magnetic susceptibility are complicated because of the interactions between particles, and the concomitant formation of self-assembled structures with complex dynamics. One way of isolating the effects of interactions would be to compare particles of a given size, so that $\tau_{\rm B}$ remains constant, but synthesised from materials with different values of the saturation magnetisation $M_{\rm s}$, so that $\lambda \sim M_{\rm s}^2$ changes substantially. The theoretical work described here provides concrete predictions which can be tested in experiments.

Acknowledgements

A.O.I. gratefully acknowledges research funding from the Russian Science Foundation, Grant No. 23-12-00039.

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