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Nonlinear contributions to the dynamic magnetic susceptibility of magnetic fluids

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ABSTRACT

Based on our earlier analytical results for the magnetization of magnetic fluids with respect to the magnetic field strength, we propose an expansion method within the framework of mean spherical approximation (MSA) to obtain the coefficients of different nonlinear terms. Through a Fourier expansion of the frequency-dependent magnetic susceptibility the harmonic coefficients corresponding to the linear and nonlinear dynamic susceptibilities are calculated from the field expansion of magnetization. The frequency dependence of the higher order susceptibilities is determined on the basis of the Debye relaxation of magnetic dipoles. Our MSA based results are in line with the corresponding limiting case of the Debye-Weiss theory. We mapped the range of applicability of the expansion method concerning the field strength and frequencies. Our results show that under weak fields a 7th order expansion is sufficient to predict the magnitudes of the susceptibility components up to the 4th harmonic relevant for magnetic fluids.

1. Introduction

One of the fundamental methods to characterize the magnetic behavior of complex magnetic systems is to probe the dynamic (ac) susceptibility, which is the differential response of the magnetization to a perturbative oscillating magnetic field. In the weak field limit the response is linear, and the undisturbed ground state of the magnetic system can be probed without any significant changes induced in the magnetic structure. However, in stronger external fields the magnetization is no longer a linear function of the field strength, and in a sinusoidal exciting field the ac susceptibility response will contain higher order harmonics due to the nonlinearity.

The ac susceptibility (and its linear and nonlinear components) is a very sensitive indicator for the presence of various magnetic ordering, spontaneous magnetization [1], and phase transitions between different magnetic states [2,3], as it shows divergence in the vicinity of the transition temperature [4]. Moreover, the frequency dependence of the ac susceptibility provides an insight into the relaxation processes [5] in a collection of magnetic dipoles.

The relaxation processes, and the ac susceptibility response of complex magnetic systems are exploited in several practical applications. For instance, the colloidal suspensions of single domain magnetic nanoparticles carrying permanent dipole moments (magnetic fluids, ferrofluids) dissipate power under an external alternating magnetic field mainly through the relaxation processes. Such systems are used as localized heat source in medical hyperthermia treatments [6,7]. Furthermore, the harmonic susceptibility response of these systems provides the basis for three-dimensional visualization in the novel biomedical application called magnetic particle imaging (MPI) [8]. During typical applications the amplitude of the alternating field is large enough to drive the system out of the region of linear response – or even into saturation – so the characterization of the nonlinear contribution to the ac susceptibility is essential.

A great variety of theoretical approaches are available to describe the ac susceptibility of an ensemble of magnetic nanoparticles. The frequency dependence of Weiss's mean-field theory can be understand by the application of the Debye theory, when the magnetic dipoles are essentially non-interacting [9]. Recently, Ivanov et al. [10] extended the modified mean field theory of interacting dipoles to describe the frequency dependence of magnetic susceptibility in Brownian relaxation domain. Through the time-dependent distribution function by the help of the Fokker–Planck equation [11] the

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Fourier components of the magnetic susceptibility were also studied.

Starting from Wertheim's mean spherical approximation (MSA) results [12] within the framework of density functional theory (DFT) one of the present authors have proposed an analytical equation for the dc magnetization of monodisperse magnetic fluids [13]. In their publication quantitative agreement was found between DFT results and corresponding canonical Monte Carlo (MC) simulation data. Later this theoretical approach was extended to the description of the magnetization of multi-component systems [14]. As a natural expansion of the multicomponent MSA magnetization to polydisperse systems, in [15] we proposed an equation for the magnetization of polydisperse magnetic fluids. Translated into the analogous electric language on the basis of our theory an implicit analytical equation for the electric field dependence of the polarization was obtained [16]. On the basis of the third-order field strength expansion of polarization we deduced a formula for the nonlinear dielectric permittivity of dipolar fluids. Moreover, we compared our theoretical findings with MC simulation and experimental data, and reasonable agreements were found.

Expanding along the line of our earlier works, the objective here is to calculate the higher order terms of magnetic field power expansion of magnetization in case of magnetic fluids. Starting from these results the frequency components of nonlinear dynamic susceptibility are predicted within the framework of MSA using the well known Debye approximation [17]. The numerical results will be compared with the Langevin and Debye-Weiss (DW) limiting cases.

2. Theory

In the following a monodisperse magnetic fluid is described by the dipolar hard sphere fluid model, where the particles are characterized by the diameter σ, and magnetic dipole moment μ. The number density of the macroscopic system is $\rho = N/V$ with the volume of the system V and number of dipolar spheres N. A highly elongated cylindrical shaped sample of magnetic fluid is considered to ensure the absence of demagnetizing field.

2.1. Magnetization and susceptibility in the framework of MSA

The dependence of the magnetization M of a magnetic fluid on an external magnetic field H is given by an implicit equation [13]:

$$M = \mu\rho\mathscr{L}\left(\frac{\mu H}{k_B T} + 3M\frac{(1-q(-\xi))}{\mu\rho}\right), \tag{1}$$

where $\mathscr{L}(x) = \coth(x) - 1/x$ is the Langevin function, T is the thermodynamic temperature, k_B is the Boltzmann constant, and ξ is the implicit solution of the corresponding MSA equation

$$4\pi\chi_L = q(2\xi) - q(-\xi). \tag{2}$$

In Eqs. (1) and (2) the function $q(x)$ is the reduced inverse compressibility function of hard spheres within the Percus–Yevick approximation:

$$q(x) = \frac{(1+2x)^2}{(1-x)^4}. \tag{3}$$

According to Eq. (1) the zero-field magnetic susceptibility of the system is

$$\chi_0 = \frac{\chi_L}{q(-\xi)}, \tag{4}$$

where $\chi_L = \rho\mu^2/(3k_B T)$ is the Langevin susceptibility.

2.1.1. Limiting cases

For $q(-\xi) = 1$ Eq. (1) gives the well known Langevin magnetization

$$M = \mu\rho\mathscr{L}\left(\frac{\mu H}{k_B T}\right), \tag{5}$$

and the corresponding magnetic susceptibility is

$$\chi_0 = \chi_L. \tag{6}$$

For $q(-\xi) = 1 - (4\pi/3)\chi_L$ Eq. (1) gives the magnetization in the mean field approximation

$$M = \mu\rho\mathscr{L}\left(\frac{\mu}{k_B T}(H + 4\pi M/3)\right), \tag{7}$$

and the corresponding magnetic susceptibility is

$$\chi_0 = \frac{\chi_L}{1 - 4\pi\chi_L/3}. \tag{8}$$

The other form of this equation is expressed for the Langevin susceptibility:

$$\frac{\chi_0}{4\pi\chi_0/3 + 1} = \chi_L. \tag{9}$$

In the literature both Eq. (8) and Eq. (9) are called Debye-Weiss equation.

2.2. Field strength expansion of MSA magnetization

In order to obtain the magnetic field strength power expansion of implicit magnetization function (see Eq. (1)) the method described in [16] can be applied. The result of the 7th order expansion is:

$$M(H) = m_0 + m_1 H + m_2 H^2 + m_3 H^3 + m_4 H^4 + m_5 H^5 + m_6 H^6 + m_7 H^7 + \dots. \tag{10}$$

Magnetic fluids show no spontaneous magnetization, therefore the coefficient m_0 is zero, and due to symmetry reasons the coefficients of even order terms also vanish. The magnitude of non-zero terms is decreased with increasing order, and their coefficients are:

$$m_1 = \frac{\rho\mu^2}{3k_B T}\frac{1}{q(-\xi)}, \tag{11}$$

$$m_3 = -\frac{\rho\mu^4}{45(k_B T)^3}\frac{1}{q^4(-\xi)}, \tag{12}$$

$$m_5 = -\frac{\rho\mu^6}{4725(k_B T)^5}\frac{11q(-\xi) - 21}{q^7(-\xi)}, \tag{13}$$

$$m_7 = -\frac{\rho\mu^8}{70875(k_B T)^7}\frac{19q^2(-\xi) - 88q(-\xi) + 84}{q^{10}(-\xi)}. \tag{14}$$

2.3. Field-dependent static susceptibility

The definition of the field-dependent static magnetic susceptibility is

$$\chi = \frac{\partial M}{\partial H}. \tag{15}$$

Considering Eq. (10) we can write that

$$\chi = m_1 + 3m_3 H^2 + 5m_5 H^4 + 7m_7 H^6 + \dots, \tag{16}$$

where in MSA m_1 gives back Eq. (8) in zero-field approximation. From m_3 the first nonlinear term can be obtained (see [16]).

2.4. Time-dependent susceptibility

In the following we assume that the external magnetic field is an alternating field:

$$H(t) = H_0 \sin(\omega t), \tag{17}$$

which oscillates along the long axis of the cylindrical sample. H_0 is the amplitude of the field, ω is the angular frequency, and t is the time. The nonlinearity of the time-dependent susceptibility under a sinusoidal field is conveniently characterized experimentally by extracting the magnitude of the higher order harmonics, thus we will use the following formalism. Substituting Eq. (17) into Eq. (16), and collecting the corresponding terms according to the formal Fourier series of χ:

$$\chi(t) = \chi_0 + \chi_{2\omega}\cos(2\omega t) + \chi_{4\omega}\cos(4\omega t) + \chi_{6\omega}\cos(6\omega t) + \dots . \tag{18}$$

For the coefficients of the trigonometric functions we obtain that

$$\chi_0 = m_1 + \frac{3}{2}m_3H_0^2 + \frac{15}{8}m_5H_0^4 + \frac{35}{16}m_7H_0^6, \tag{19}$$

$$\chi_{2\omega} = \frac{3}{2}m_3H_0^2 + \frac{5}{2}m_5H_0^4 + \frac{105}{32}m_7H_0^6, \tag{20}$$

$$\chi_{4\omega} = \frac{5}{8}m_5H_0^4 + \frac{21}{16}m_7H_0^6, \tag{21}$$

$$\chi_{6\omega} = \frac{7}{32}m_7H_0^6. \tag{22}$$

As we can see, even the first coefficient χ_0 contains contribution from the higher order terms of the power expansion of the magnetization function, but the number of terms in $\chi_{n\omega}$ decreases with the harmonic number n. In Eqs. (19)–(22) the following well known trigonometric relations are used:

$$\begin{aligned}\sin^2 x &= \frac{1-\cos(2x)}{2},\\ \sin^4 x &= \frac{3-4\cos(2x)+\cos(4x)}{8},\\ \sin^6 x &= \frac{10-15\cos(2x)+6\cos(4x)-\cos(6x)}{32}.\end{aligned} \tag{23}$$

2.4.1. Complex susceptibility, Debye approximation

To study the dynamic magnetic properties we introduce the complex magnetic susceptibility. Instead of Eq. (17) we prescribe a complex exciting magnetic field as:

$$H(t) = H_0 e^{i\omega t}, \tag{24}$$

where i is the complex unit. In this case the magnetization response and the susceptibility are also complex quantities. For the complex magnetic susceptibility the following sign convention is used:

$$\hat{\chi} = \chi'(\omega) - i\chi''(\omega), \tag{25}$$

where χ' is the real and χ'' is the imaginary part of the magnetic susceptibility. In the classical Debye approximation Eq. (9) can be obtained from the average component of dipole moment in the direction of the magnetic field:

$$\langle \mu \cos\theta \rangle \simeq \frac{\mu^2 H}{3k_BT}, \tag{26}$$

where the Boltzmann distribution function is used for the approximate calculation of the angle average. Assuming an alternating external field (see Eq. (24)) the approximate distribution function is modified [17], and therefore the expression of the average dipole moment in the direction of the ac field is also altered:

$$\langle \mu \cos\theta \rangle \simeq \frac{\mu^2 H_0}{3k_BT}\frac{1}{(1+i\omega\tau)}, \tag{27}$$

where τ is the microscopic relaxation time. We assume that the relaxation of the magnetic dipoles occurs only by the Brownian mechanism, where the dipole moment rotates with the whole par-

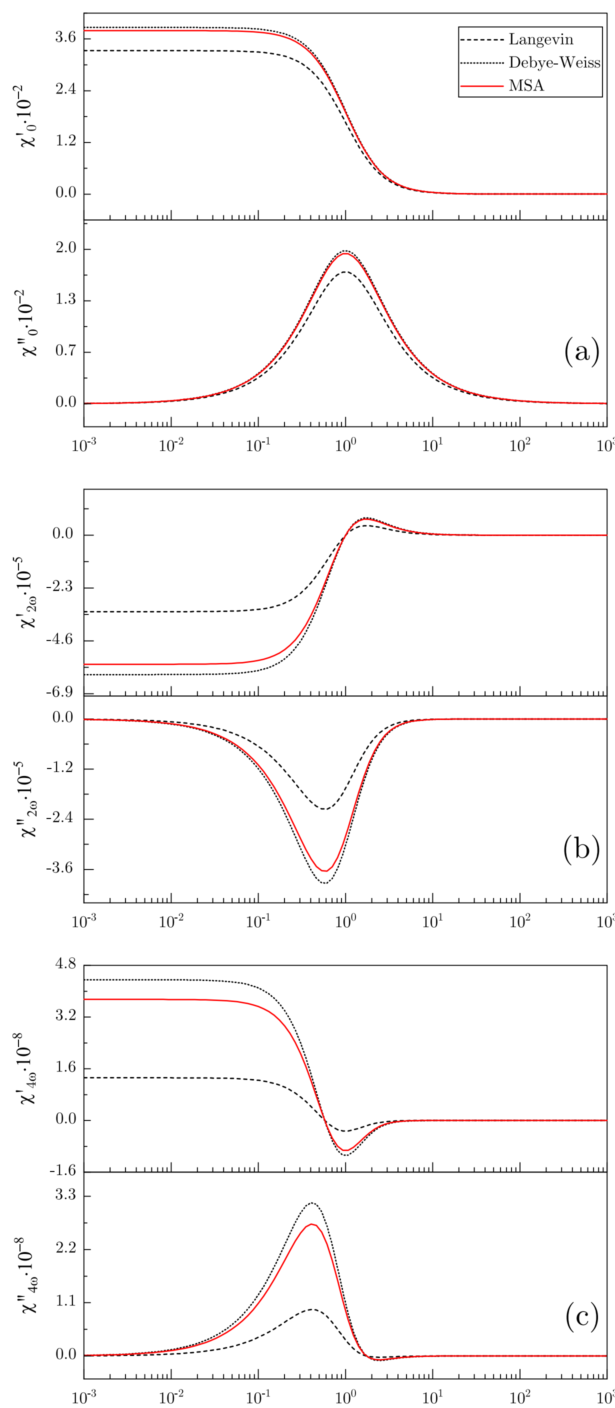


Fig. 1. Spectra of the linear $\hat{\chi}_0$ (a), and the higher order nonlinear $\hat{\chi}_{2\omega}$ (b) and $\hat{\chi}_{4\omega}$ (c) susceptibilities according to the MSA theory in comparison with the corresponding Langevin and Debye-Weiss approximations under a weak field of $H^* = 0.1$ ($\rho^* = 0.1, \mu^* = 1$).

ticle. According to [17] from the static equation of Debye-Weiss (Eq. (8)) the dynamic (frequency-dependent complex expression) can be obtained by the substitution of:

$$\frac{\mu^2 H}{3k_B T} \rightarrow \frac{\mu^2 H_0}{3k_B T}\frac{1}{(1+i\omega\tau)}. \tag{28}$$

In Debye-Weiss approximation, at zero field strength for the complex susceptibility our theory gives:

$$\frac{\hat{\chi}_0}{4\pi\hat{\chi}_0/3+1} = \frac{\chi_L}{(1+i\omega\tau)}, \tag{29}$$

which is in harmony with Eq. (9). Considering the real and imaginary parts we obtain the classical Debye-Weiss expressions. We note that this microscopic relaxation time τ differs from the macroscopic relaxation time of Debye (see [17]). To obtain the frequency dependence of the field-dependent terms in complex $\hat{\chi}$, and $\hat{\chi}_{n\omega}$ (n = 2, 4, 6) coefficients Eq. (28) is applied again. For simplicity, in the following we summarize the complex $\hat{m}_k$ (k = 1, 3, 5, 7) MSA terms to derive the complex MSA $\hat{\chi}_0$ and $\hat{\chi}_{n\omega}$ expressions by substituting them into Eqs. (19)–(22):

$$\hat{m}_1 = \frac{\rho\mu^2}{3k_B T}\frac{1}{q(-\xi)}\frac{1}{(1+i\omega\tau)}, \tag{30}$$

$$\hat{m}_3 = -\frac{\rho\mu^4}{45(k_B T)^3}\frac{1}{q^4(-\xi)}\frac{1}{(1+i\omega\tau)^2}, \tag{31}$$

$$\hat{m}_5 = -\frac{\rho\mu^6}{4725(k_B T)^5}\frac{11q(-\xi)-21}{q^7(-\xi)}\frac{1}{(1+i\omega\tau)^3}, \tag{32}$$

$$\hat{m}_7 = -\frac{\rho\mu^8}{70875(k_B T)^7}\frac{19q^2(-\xi)-88q(-\xi)+84}{q^{10}(-\xi)}\frac{1}{(1+i\omega\tau)^4}. \tag{33}$$

The coefficients $\hat{\chi}_{n\omega}$ (n = 2, 4, 6) are the nonlinear susceptibilities and correspond to the amplitudes of the nth harmonic. In the zero-field limit, where the response is linear the higher order susceptibilities vanish, and only the coefficient $\hat{\chi}_0$ is present, which corresponds to the linear ac susceptibility. In case of typical magnetic fluids the amplitudes of the $n \geqslant 6$ harmonics are so small that their accurate detection by experimental methods is difficult, so usually those are neglected. Therefore, we will consider the harmonic susceptibilities only up to n = 4. It is worth to mention, that if a symmetry breaking dc bias field is superimposed on the ac field, then the odd harmonics will appear besides the even ones.

3. Numerical results and discussion

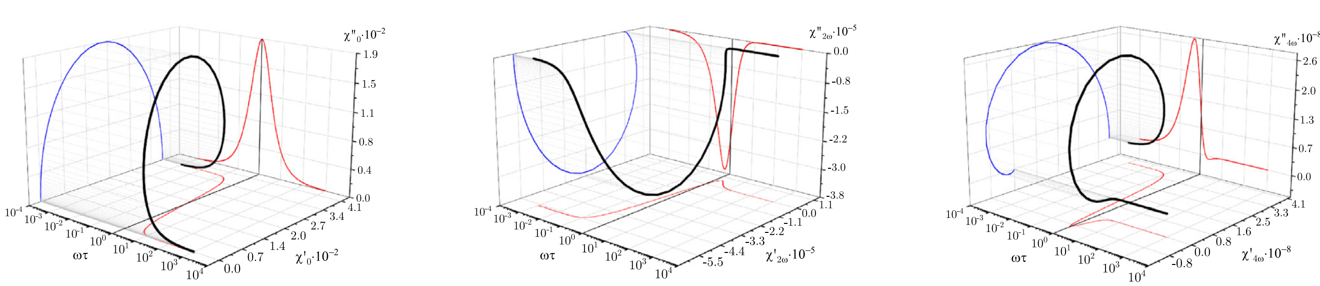
In the following we use reduced quantities: $\rho^* = \rho\sigma^3$ is the reduced density, $\mu^* = \mu/\sqrt{\sigma^3 k_B T}$ is the reduced dipole moment, and $H^* = H_0\sqrt{\sigma^3/(k_B T)}$ is the reduced magnetic field strength. The numerical results were calculated for a reference system with ρ^* = 0.1, and μ^* = 1, so $4\pi\chi_L \approx 0.415$. The reduced parameters were chosen to represent a typical dilute magnetic fluid containing spherical magnetite particles with a magnetic core diameter of $\sigma \approx 10$ nm in a carrier liquid. The field amplitude, and frequency dependence of the linear susceptibility $\hat{\chi}_0$, and the higher order nonlinear susceptibilities ($\hat{\chi}_{2\omega}$ and $\hat{\chi}_{4\omega}$) were calculated. The results are presented as the complex functions, and the spectra of the real and imaginary parts of the quantities $\hat{\chi}_0 = \chi'_0 - i\chi''_0$ and $\hat{\chi}_{n\omega} = \chi'_{n\omega} - i\chi''_{n\omega}$ ($n = 2, 4$).

First, we compare the prediction of the current expansion based MSA theory under weak fields with the corresponding Langevin and Debye-Weiss approximations as limiting cases. As it is shown in Fig. 1 all of the three considered theories give qualitatively similar results at H^* = 0.1. However, as the MSA and DW data show, the interparticle interactions increase the linear – and especially the nonlinear – susceptibilities compared to the non-interacting magnetic dipoles of the Langevin theory. The difference becomes larger with the order of the harmonics n in case of the real and the imaginary part as well. The predictions of the MSA and DW theories for the effect of interactions on the magnitude of $\hat{\chi}_{2\omega}$ and $\hat{\chi}_{4\omega}$ are close to each other; the agreement is reasonable in weak fields. The DW theory slightly overestimates both nonlinear susceptibilities compared to the MSA results. The difference between these two theories widens as the order of the susceptibility is increased. It is known that the DW theory gives accurate results, but it predicts that the linear and nonlinear susceptibilities diverge at $4\pi\chi_L = 3$ [9]. In case of the chosen system χ_L remains well below the point of divergence, thus the comparison with the DW theory is reasonable.

The frequency-dependent complex function of $\hat{\chi}_0, \hat{\chi}_{2\omega}$, and $\hat{\chi}_{4\omega}$ together with the projected spectra of the real and imaginary parts are shown in Fig. 2. The following features can be pointed out. The linear susceptibility $\hat{\chi}_0$ is well described by the Debye relaxation, and displays a relaxation peak of χ''_0 at $\omega\tau$ = 1. A slower relaxation of the higher order susceptibilities is observed as the global extrema of the imaginary parts occur at $\omega\tau < 1$ frequencies. The shift of relaxation into the lower frequencies increases with the order of the harmonics, and simultaneously the spectra of the imaginary part becomes asymmetric with a broadened lower frequency side. The real part of $\hat{\chi}_{2\omega}$ and $\hat{\chi}_{4\omega}$ exhibit a local extremum near $\omega\tau$ = 1. The results are in qualitative agreement with the spectra derived by Kuznetsov and Pshenichnikov [18] for the first three components of the nonlinear susceptibilities by a theoretical approach based on the Fokker–Planck equation.

3.1. Field strength dependence

Let us now examine the effect of increasing magnetic field strength on the linear and nonlinear susceptibilities. The magnetic



(a) (b) (c)

Fig. 2. 3D representation of the complex functions of $\hat{\chi}_0$ (a), $\hat{\chi}_{2\omega}$ (b), and $\hat{\chi}_{4\omega}$ (c) in the frequency range of $\omega\tau = 10^{-3} - 10^3$ under weak fields ($H^* = 0.1$). The linear component shows the classical Debye relaxation at $\omega\tau$ = 1 (marked by the solid line on the real and imaginary planes), while the relaxation of the higher order susceptibilities is shifted toward lower frequencies.

field strength dependence of the spectra of $\hat{\chi}_0$, $\hat{\chi}_{2\omega}$, and $\hat{\chi}_{4\omega}$ according to the expansion based MSA theory is shown in Fig. 3. The reduced magnetic field strength was increased up to $H^* = 2$. A limitation of the current theory can be seen at larger field strengths: above $H^* = 1.6$ unphysical features begin to appear in the spectra of χ_0' and χ_0'' in the form of local maxima. This is due to the truncated nature of the expansion series of the magnetization function (Eq. 10). The limitation can be overcome by continuing the current

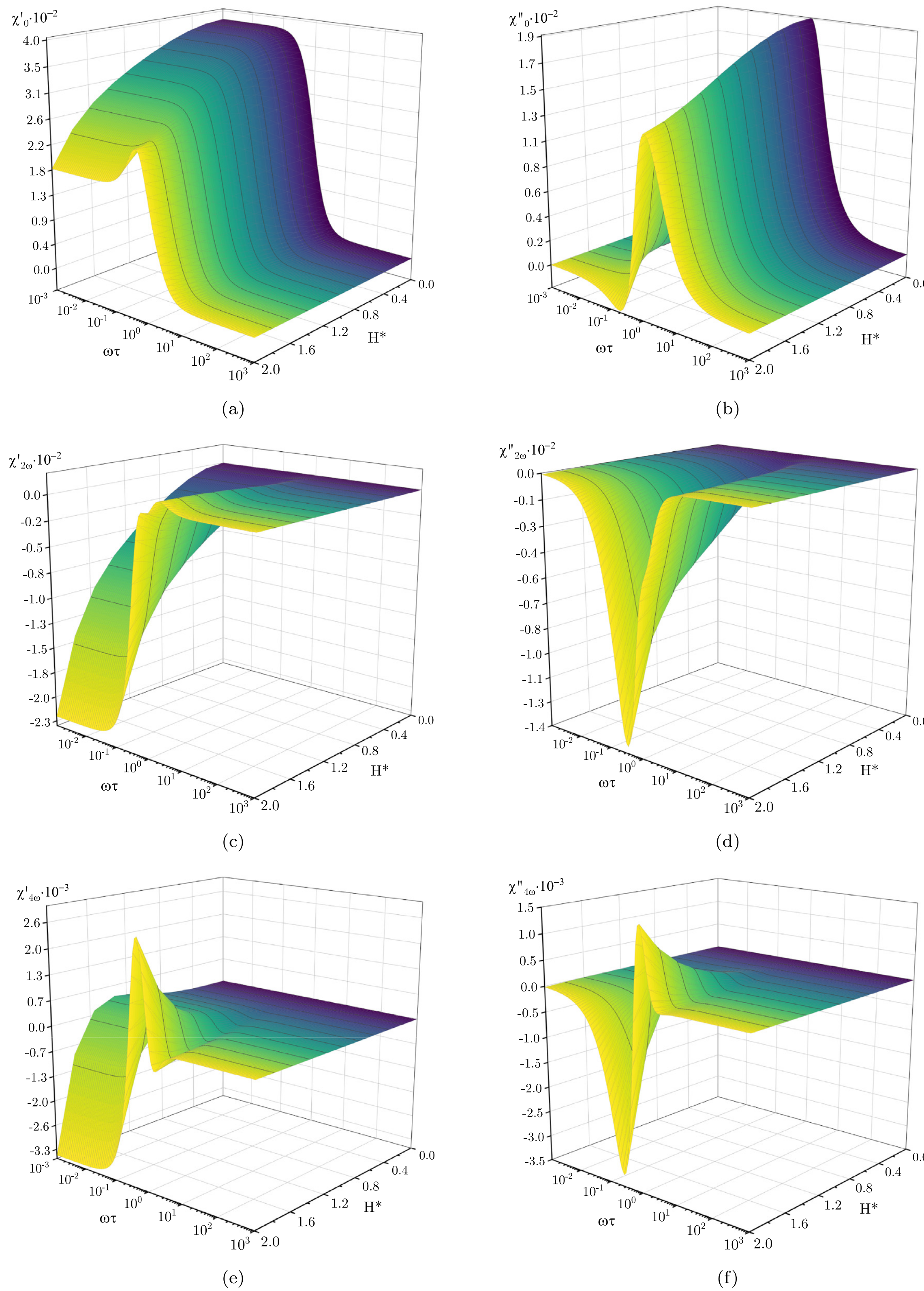


Fig. 3. The effect of increasing magnetic field strength (up to $H^* = 2$) on the spectra of the real and imaginary parts of $\hat{\chi}_0$ (a, b), $\hat{\chi}_{2\omega}$ (c, d), and $\hat{\chi}_{4\omega}$ (e, f) susceptibilities according to the present MSA theory ($\rho^* = 0.1, \mu^* = 1$).

expansion beyond the 7th order, which would incorporate further terms into Eqs. (19)–(21) of the susceptibilities, and enhance their convergence.

The spectra of $\hat{\chi}_0, \hat{\chi}_{2\omega}$, and $\hat{\chi}_{4\omega}$ around the relaxation in the field strength range below $H^* \leqslant 1$, where our MSA theory gives reliable results is shown in Fig. 4. In case of $\hat{\chi}_0$, both real and imaginary parts become smaller as H^* is increased. Simultaneously, the relaxation peak of χ_0'' shifts towards higher frequencies in agreement with other theoretical [19,20] and experimental results [21]. On the other hand, the magnitude of the real and imaginary parts of the nonlinear susceptibilities increase significantly with the increase of H^*. This behavior results from the enlarged nonlinearity in the magnetization of the system as it is driven towards saturation by the increasing field strength. The relaxation of higher order components behaves similarly to the case of $\hat{\chi}_0$: the peak of $\chi_{2\omega}''$ shifts slightly, while the peak of $\chi_{4\omega}''$ to a grater extent into the higher frequency region with larger H^*.

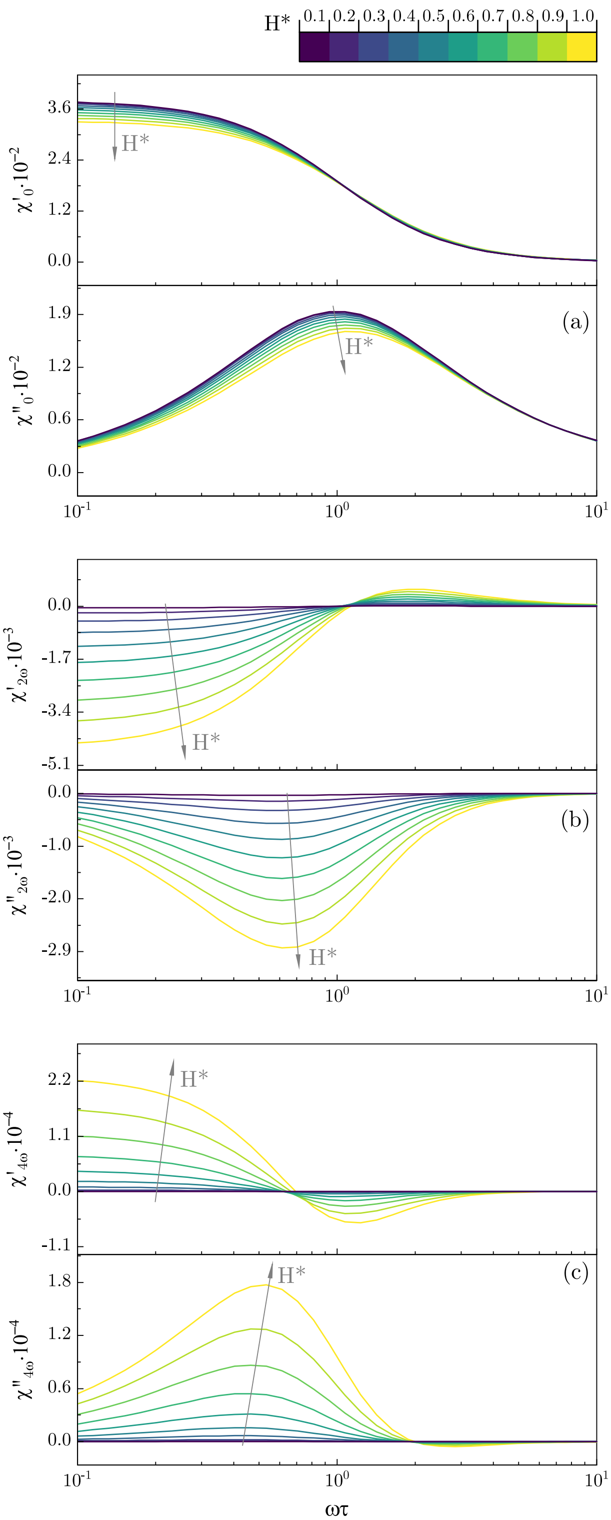


Fig. 4. With increasing magnetic field strength (in the range of $H^* = 0.1 - 1.0$, marked by the arrows) χ_0' and χ_0'' (a) decreases, while the contribution of the nonlinear components $\hat{\chi}_{2\omega}$ (b), and $\hat{\chi}_{4\omega}$ (c) become larger. Simultaneously, the relaxation peaks shift towards higher frequencies.

To emphasize the relative contribution of the linear and higher order susceptibilities the magnetic field dependence of their absolute values is shown in Fig. 5. In the zero-field limit of $H^* \rightarrow 0$ $|\hat{\chi}_0|$ approaches the initial dynamic susceptibility, while both higher order susceptibilities vanish. As H^* grows the component $|\hat{\chi}_0|$ decreases, while $|\hat{\chi}_{2\omega}|$ and $|\hat{\chi}_{4\omega}|$ show an increasing trend according to a power law in the investigated range of H^*. Some of our preliminary experimental data for the magnetic field dependence of the magnitude of $\hat{\chi}_{2\omega}$ and $\hat{\chi}_{4\omega}$ in magnetic fluids show the same behavior [22]. Similar findings were described in [18], but they showed that within the Fokker–Planck approach the power law increase in weak fields turns into a hyperbolic decrease at large field strength, when the magnetization approaches saturation. The field strength range required to reach the vicinity of saturation is not accessible by the current (7th order) expansion of the MSA approach, as it was mentioned earlier.

If we consider the magnitude of the nonlinear susceptibilities, it is clear that the overall nonlinear contribution is dominated by the magnitude of the second harmonics. The $\hat{\chi}_{4\omega}$ component is an order of magnitude smaller than $\hat{\chi}_{2\omega}$, which is in line with the predictions of other theories [23]. Within the framework of the MSA theory the nonlinearity is the result of the normal saturation of the magnetization only, thus the real part of $\hat{\chi}_{2\omega}$ has a negative sign (see Fig. 4), and with that the overall nonlinear contribution is also negative. This is in agreement with experimental data for magnetic fluids in weak fields [24,25]. Nonlinear susceptibility with a positive sign was obtained by Wang and Huang [23] using a perturbation expansion method. They attributed the positive effect to the anomalous saturation, which stems from the shifting of equilibrium between the structures with different dipole moments (single particles and different sized particle chains). Structural changes as large, that the anomalous saturation overcomes the negative effect of the normal saturation can be expected only at large field strength. However, the present MSA theory can not describe the anomalous saturation, and the nonlinear contribution will remain negative even under a strong field. We note that the positive contribution of the anomalous saturation could be described within the framework of MSA, if the polarizabilities of the dipolar spheres are included, but such an attempt has not been made yet.

Convergence of the expanded MSA model

As Eqs. (19)–(21) show the linear and nonlinear susceptibilities are composed of a sum of terms containing the coefficients m_{n+1}, which stem from the power expansion of the magnetization function. This is a frequently used formalism, and in the domain of weak fields it is generally assumed that $\chi_{n\omega}$ is determined mostly by the first component of its series [26]. So, if H_0 is small, then χ_0 contains only the linear static susceptibility m_1, $\chi_{2\omega}$ is proportional to m_3, and $\chi_{4\omega}$ is connected mainly to m_5. In the following, we will examine the limits of this assumption within the MSA theory.

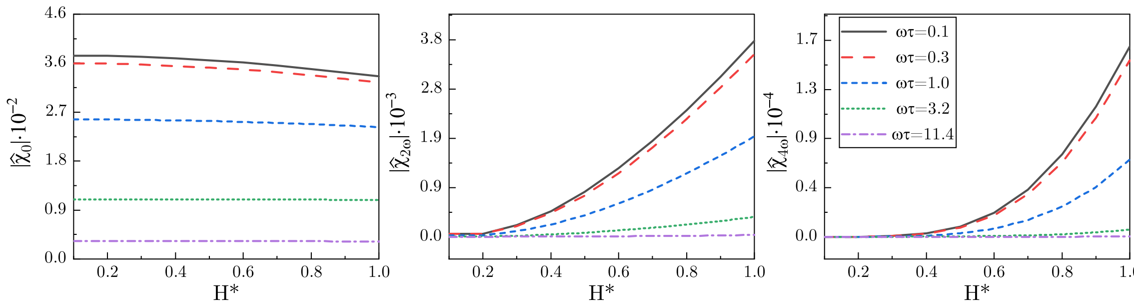


Fig. 5. Magnetic field strength dependence of the magnitude of the first three susceptibility component ($\hat{\chi}_0, \hat{\chi}_{2\omega}$, and $\hat{\chi}_{4\omega}$) at different frequencies in the weak field range ($\rho^* = 0.1, \mu^* = 1$).

In Fig. 6 the calculated spectra of $\hat{\chi}_0, \hat{\chi}_{2\omega}$, and $\hat{\chi}_{4\omega}$ are shown, when these are gradually approximated by their expanding series up to the term containing $\hat{m}_7$. The spectra are given for H^* = 0.1, and at a larger, H^* = 1 field strength. At H^* = 0.1 the contributions of the higher order terms in the linear and nonlinear susceptibilities are so small, that even the first term gives an excellent approximation. This is confirmed by the overlapping curves in Fig. 6a-c. At larger field strength the situation changes, because the relative contributions of the higher order terms increase rapidly with H^*, and even with the harmonic number n. In this case considering only the first term becomes an inadequate approximation (see Fig. 6d-f). The convergence is satisfactory for the first three susceptibility components (especially in the case of $\hat{\chi}_0$), but deteriorates with increasing n, as the number of terms in the series of the higher order susceptibilities decreases. E.g. $\hat{\chi}_{4\omega}$ has only two terms, and using just the first one will cause large error. The number of terms in the higher order susceptibilities, and with that the accuracy of the convergence can be increased by continuing the power expansion of the magnetization function beyond the 7th order, as it was pointed out earlier.

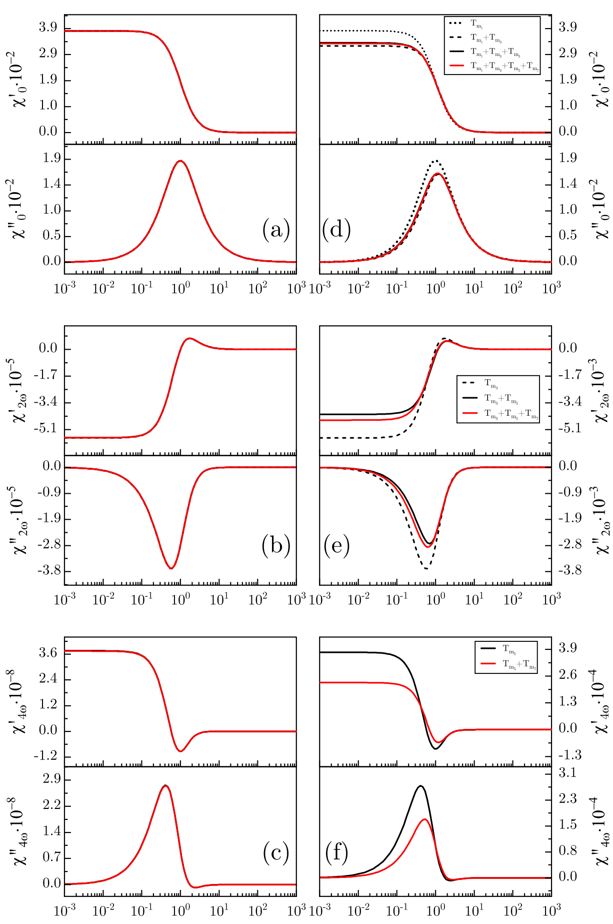


Fig. 6. Approximation of the linear and nonlinear susceptibilities by the sum of increasing number of terms ($T_{m_k}, k = 1, 3, 5, 7$) in their expanding series (Eqs. (19)–(21)) under a weak field of H^* = 0.1 (a, b, c) and at larger field strength H^* = 1 (d, e, f). In weak fields even the first T_{m_1} term is an adequate approximation, but at larger field strengths the convergence deteriorates with increasing harmonic number.

4. Conclusions

We have given a theoretical description of the nonlinear dynamic susceptibility response of interacting magnetic dipoles within the framework of MSA theory. A power expansion based approach was used to calculate the frequency and magnetic field strength dependence of the linear and higher order harmonic susceptibilities. From the obtained results the following conclusions have been drawn:

- An advantage of the current theoretical approach with the expansion based treatment of the ac susceptibility is that simple, analytical equations can be derived for the linear and nonlinear components, which directly correspond to the experimentally detectable magnitude of the higher harmonic susceptibilities.
- The predictions of MSA for the frequency dependence of the first three ac susceptibility components agree reasonably with the well tested Debye-Weiss limiting case under weak fields, however the difference between the two theories increases with the order of the harmonics. We found qualitative agreement for the spectra of $\hat{\chi}_0, \hat{\chi}_{2\omega}$, and $\hat{\chi}_{4\omega}$ with the results in Ref. [18] derived from the more complicated solution of the Fokker–Planck equation.
- We calculated the spectra of the susceptibility components in a range of field strengths, and found that due to the truncated nature of the series expansion of the magnetization function used in the present theory, the MSA predicts unphysical features in the spectra above $H^* > 1.6$.
- In weak fields the 7th order expansion applied here is sufficient to determine the susceptibility component up to the 4th harmonic with acceptable accuracy. With longer power expansion the applicable field strength range can be expanded, and the accuracy of the higher order susceptibilities would be improved further.
- We tested the convergence of the series of the first three susceptibility components, which was satisfactory even in case of $\hat{\chi}_{4\omega}$. Our results showed that the generally accepted approximation of $\hat{\chi}_0$ and higher susceptibilities just by the first term is justified only under weak fields ($H^* \sim 0.1$) for the considered dilute magnetic fluids.

In future works we will try to extend the applicability of the expansion based MSA method to cover the domain of strong fields near saturation, and extend the theory to include the case, when a dc bias field is also applied. We also plan to test the predictions of the theory for the dynamic susceptibility response of magnetic fluids against simulations, and experimental results.

Data availability statement

All data generated and analyzed during this study are available from the corresponding author on reasonable request.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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