Anisotropy effects on the nonlinear magnetic susceptibilities of superparamagnetic particles

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Equilibrium nonlinear susceptibilities of an overdamped three-dimensional rotator in a uniaxial anisotropy potential $\propto\cos^2\theta$ (θ is the angle between the rotator axis and the potential symmetry axis), which apply to independent magnetic particles and other rotationally bistable entities, are derived. In the crossover range from free-rotator to either two-state or plane-rotator regime induced by that potential, the dependences of the nonlinear susceptibilities on T can be steeper than those of the limit inverse-temperature power laws. The nonlinear susceptibilities can then resemble the high-temperature ranges of quantities diverging at low temperature, misleadingly suggesting interparticle interactions. [S0163-1829(97)04002-2]

I. INTRODUCTION

Fine magnetic particles are ubiquitous in naturally occurring and manufactured forms.[1] Along with their technological relevance, they can be considered as model systems for various phenomena in nonequilibrium statistical mechanics, to illustrate: rotational Brownian motion and thermally activated processes in multistable systems,[2] and stochastic resonance;[3] and in condensed-matter physics, e.g., dipole-dipole interaction effects,[4] macroscopic quantum phenomena,[5] and dependence of the properties of solids on their size.

As an important example of how the properties of magnetic particles can differ from those of the bulk material, a sufficiently fine particle consists of a single *domain*, whose magnetic moment, $\mathbf{m}$, can rotate due to thermal agitation, surmounting the magnetic-anisotropy potential barriers.[6] For large $\beta\Delta U$ (barrier height over temperature), the mean time for this process is given by $\tau=\tau_0\exp(\beta\Delta U)$, where $\tau_0(\sim10^{-10}$–10^{-13} s) is related to intrawell relaxation. For $\tau\ll t_m$ (the measurement time), $\mathbf{m}$ maintains the equilibrium distribution of orientations (as $m\sim10^3-10^4\mu_B$ this phenomenon is named *superparamagnetism*), when $\tau\gg t_m$, $\mathbf{m}$ is *blocked* at a potential minimum, and, under intermediate conditions, *nonequilibrium phenomena* are observed.

The extent to which the properties of certain spin glasses can be explained in terms of a progressive blocking of superparamagnetic clusters of spins, has deserved considerably attention.[7] In this frame, the nonlinear magnetic susceptibilities (NLMS's) play a significant role. Thus Bitoh *et al.* have shown $\chi_2(\omega,T)$ [χ_{2n} being the nth order NLMS] as a suitable experimental tool to *distinguish* canonical spin glasses from solid dispersions of noninteracting particles.[8(a)] The marking feature is not the width nor the roundness of the peak on $\chi_2(\omega,T)$, which could just reflect barrier distribution, but the way χ_2 decreases as T increases above the peak temperature: abruptly for the former but slowly for the latter. Besides, for amorphous $Fe_{93}Zr_7$, considered as a superparamagneticlike *spin glass*, χ_2 resembles that of the particles studied by Bitoh *et al.*[9] Finally, the glassy dynamics exhibited by interacting particles[4(c)] suggests an extensive use of χ_{2n} in their study, considering the propriety of these quantities to study collective phenomena in disordered systems.

Despite these interesting issues, it is not available at present a satisfactory theoretical description of the NLMS's of noninteracting particles: merely expressions for χ_{2n} that account for limit cases of magnetic anisotropy can be used. Limit descriptions of anisotropy, however, just hold for the equilibrium magnetic properties of liquid dispersions of particles (anisotropy is uncoupled from the magnetization process *via* particle physical rotation[10]), and solid dispersions in the limits $\beta\Delta U\gg1$ and $\beta\Delta U\ll1$. For these latter systems (e.g., the self-same *magnetic fluids* when the solvent is frozen), the exponential decrease of τ as T increases, yields the wide range $\ln(t_m/\tau_0)$ $(\sim10$–$35)>\beta\Delta U\geq0$ as the superparamagnetic one ($\tau\ll t_m$), turning inadequate limit descriptions of magnetic anisotropy. It is precisely in this range where the mentioned decrease of χ_2 with increasing T occurs. Nevertheless, to our knowledge, the NLMS's have never been derived from the available magnetization vs field formulas that include anisotropy. In fact, these formulas are either not expressly suited to derive χ_{2n},[11(a)] or would yield the NLMS's as series of powers of the anisotropy parameters.[11(b)] The lack of proper formulae for χ_{2n} entails that, e.g., (i) alternate features of χ_2 for noninteracting particles cannot theoretically be compared to those of canonical spin glasses, (ii) the NLMS's of presumed *superparamagneticlike* systems cannot be checked against the superparamagnetic model, and (iii) as it is not known the result from which χ_2 would depart due to interparticle interactions, χ_2 does not inform about them.

In this paper, rigorous expressions for the NLMS's of a solid dispersion of noninteracting superparamagnetic particles are derived. Particle magnetic moment is described as an overdamped classical 3D rotator in a uniaxial anisotropy potential $U_A(\theta)=-a\cos^2\theta$ (θ is the angle between the rotator axis and the potential symmetry axis). Consequently, the results apply to the equilibrium nonlinear response of any system made up by independent such entities (rotationally bistable ones for $a>0$). Along with independent fine magnetic particles, a variety of other systems can approximately be described as assemblies of such rotators, among them: superparamagneticlike spin glasses,[12(a)] magnetic molecular materials as those named $Mn_{12}Ac$,[12(b)] certain high-spin dilutely doped glasses (random-axial-anisotropy model for magnetic glasses,[12(c)] and nematic liquid crystals with uniaxial physical properties.[12(d)]

Developing the superparamagnetic model, the derived expressions overcome the aforementioned hurdles and show that (i) the crossover from high-temperature free-rotator regime to low-temperature either two-state ($a>0$) or plane-rotator ($a<0$) ones induced by the potential entails a sizeable departure of $\chi_{2n}(T)$ from the power law dependences $[T^{-(2n+1)}]$ of the limit regimes [Langevin (Heisenberg) and either Ising or XY, respectively]. (ii) Within the crossover range, the rate of change of $\chi_{2n}(T)$ can be steeper than that of $T^{-(2n+1)}$, hence $\chi_{2n}(T)$ can resemble the high-temperature range of a quantity with a low-temperature divergence, misleadingly suggesting interparticle interactions. (iii) Unlike the linear one, the first nonlinear susceptibilities retain these properties when the anisotropy (symmetry) axes are oriented at random.

II. EXPANSION OF THE PARTITION FUNCTION IN A SERIES OF POWERS OF THE MAGNETIC FIELD; THE FIRST NONLINEAR SUSCEPTIBILITIES

The statistical independence of noninteracting particles allows one to express M, the magnetization along the external magnetic field $\mathbf{B}$, as $M=(\Sigma\nu)^{-1}\Sigma\langle\mathbf{m}\rangle\cdot\hat{\mathbf{b}}$, where ν denotes particle volume, $\hat{\mathbf{b}}=\mathbf{B}/B$, and $\langle\cdot\rangle$ stands for thermal average. The expansion of the one-particle contribution $M_p\equiv\langle\mathbf{m}\rangle\cdot\hat{\mathbf{b}}$,

$$M_p=\chi_0^pH+\chi_2^pH^3+\chi_4^pH^5+\chi_6^pH^7+\cdots \tag{1}$$

where $(H=B/\mu_0)$, defines the one-particle linear, χ_0^p, and nonlinear, χ_{2n}^p, susceptibilities, $n=1,2,3,\ldots$ $[\chi_{2n}=(\Sigma\nu)^{-1}\Sigma\chi_{2n}^p]$. For equally oriented, identical particles $M=\nu^{-1}M_p$ and $\chi_{2n}=\nu^{-1}\chi_{2n}^p$; hence the index p will usually be dropped.

On introducing unit vectors, $\hat{\mathbf{n}}$ along the anisotropy axis and $\hat{\mathbf{e}}=\mathbf{m}/m$, particle total magnetic potential (anisotropy plus Zeeman terms) is given by

$$-\beta U=\sigma(\hat{\mathbf{e}}\cdot\hat{\mathbf{n}})^2+\xi(\hat{\mathbf{e}}\cdot\hat{\mathbf{b}}), \tag{2}$$

where $\xi=\beta mB$ and $\sigma=\beta K\nu$ ($K=a/\nu$, being the anisotropy energy constant). Easy-axis and easy-plane anisotropy correspond, respectively, to $K>0$ and $K<0$. The relevance for magnetic particles of the taken form for the anisotropy term has been discussed in, e.g., Ref. 13. We choose $\hat{\mathbf{n}}$ as the polar axis of the coordinate system; (θ,ϕ) and $(\alpha,0)$ denote the angular coordinates of $\hat{\mathbf{e}}$ and $\hat{\mathbf{b}}$, respectively. On introducing $\xi_\parallel=\xi\cos\alpha$ and $\xi_\perp=\xi\sin\alpha$, the integral over ϕ in the partition function, $Z=\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\sin\theta\exp(-\beta U)/4\pi$, give us a factor $I_0(\xi_\perp\sin\theta)$, where $I_n(\cdot)$ is the modified Bessel function of the first kind of order n (Ref. 14, p. 610). Insertion of the expansions of $I_0(\xi_\perp\sin\theta)$ and $\exp(\xi_\parallel\cos\theta)$ into Z, and the substitution $x=\cos\theta$, yield

$$Z=\sum_{i,k=0}^{\infty}b_{ik}\xi^{2(i+k)}\int_0^1dx\ x^{2i}(1-x^2)^ke^{\sigma x^2},$$

where $b_{ik}=[(2i)!2^{2k}(k!)^2]^{-1}\cos^{2i}\alpha\sin^{2k}\alpha$. On gathering the terms with the same power of ξ and expanding $(1-x^2)^k$, we get

$$Z=F\sum_{i=0}^{\infty}\xi^{2i}\left\{\sum_{k=0}^{i}b_{i-k,k}\sum_{m=0}^{k}(-1)^m\binom{k}{m}\frac{F^{(m+i-k)}}{F}\right\},$$

where $F^{(l)}(\sigma)\equiv\int_0^1dx\ x^{2l}\exp(\sigma x^2)$ is the lth-order derivative of $F(\sigma)\equiv F^{(0)}(\sigma)$.[2(c)] Hereafter, $C_i(\sigma,\alpha)/i!$ will denote the expression into the above curly brackets. In passing, we quote that $F^{(l)}(\sigma)=M(l+1/2,l+3/2;\sigma)/(2l+1)$, where $M(a,c;\sigma)$ is the confluent hypergeometric (Kummer) function (Ref. 14, p. 753).

By dint of the relation $M_p=m\partial\ln(Z)/\partial\xi$, we obtain

$$M_p=m\sum_{i=1}^{\infty}\frac{2\xi^{2i-1}}{(i-1)!}C_i\Bigg/\sum_{i=0}^{\infty}\frac{\xi^{2i}}{i!}C_i. \tag{3}$$

On substituting into this equation, $F^{(l)}/F|_{\sigma=0}=(2l+1)^{-1}$, we recover the Langevin result $\{M_p=m[\coth(\xi)-\xi^{-1}]\}$, $F^{(l)}/F|_{\sigma\to\infty}=1$, M for Ising spins $[M_p=m\cos\alpha\tanh(\xi_\parallel)]$, and $F^{(l)}/F|_{\sigma\to-\infty}=0$ ($l>0$), M for the plane rotator $[M_p=m\sin\alpha I_1(\xi_\perp)/I_0(\xi_\perp)]$. Thus Eq. (3) generalizes these results as a quotient of two series of powers of ξ whose coefficients and, hence, the NLMS's, are expressible in terms of Kummer functions. Note that when m is temperature independent, M depends on B and T via B/T in the three limit models [B/T superposition of M and $\chi_{2n}\propto T^{-(2n+1)}$].

On taking the ξ derivative of the cumulantlike expansion of $\ln(Z)$, we get for M_p

$$\begin{aligned}M_p=m\{&2C_1\xi+2[C_2-C_1^2]\xi^3+[C_3-3C_2C_1+2C_1^3]\xi^5\\&+\tfrac{1}{3}[C_4-4C_3C_1-3C_2^2+12C_2C_1^2-6C_1^4]\xi^7+\cdots\},\end{aligned} \tag{4}$$

which embodies χ_2, χ_4, and χ_6. (χ_{2n} is obtained through the insertion of the appropriate C_i's into the expression for the nth-order cumulant.) Due to the temperature dependence of $C_i(\sigma,\alpha)$ through σ, $\chi_{2n}(T)$ no longer fulfills a $T^{-(2n+1)}$ law. Moreover, as $\sigma=\beta K\nu$, the actual dependence of χ_{2n} on T is determined by the distributions in K and ν occurring in the system.

III. THE NONLINEAR SUSCEPTIBILITY χ_2

Henceforth some of the parallel properties of the NLMS's will be illustrated on χ_2. On defining $G\equiv F'/F[(\cdot)'\equiv d(\cdot)/d\sigma]$, using the relation $F''/F=G'+G^2$, and then inserting C_1 and C_2 into Eq. (4), we get for χ_2^p

$$\begin{aligned}\chi_2^p=\mu_0^3m^4[&\tfrac{1}{6}(G'-2G^2)\cos^4\alpha-\tfrac{1}{2}G'\cos^2\alpha\sin^2\alpha\\&+\tfrac{1}{16}(G'-G^2+2G-1)\sin^4\alpha]\beta^3.\end{aligned} \tag{5}$$

The part of χ_2^p embodying nonelementary functions, $\chi_2^p/(\mu_0^3m^4\beta^3)$, can be computed, e.g., through the numerical integration[15] of the differential equation

$$G'=(2\sigma)^{-1}(1-3G)+G(1-G), \tag{6}$$

which is derived from the recurrence relations $F^{(l)}=[e^\sigma-2\sigma F^{(l+1)}]/(2l+1)$. The limit cases of χ_2 are $\chi_2|_{\text{Langevin}}=-(\mu_0^3m^4/45)\beta^3$, $\chi_2|_{\text{Ising}}=-(\mu_0^3m^4\cos^4\alpha/3)\beta^3$,

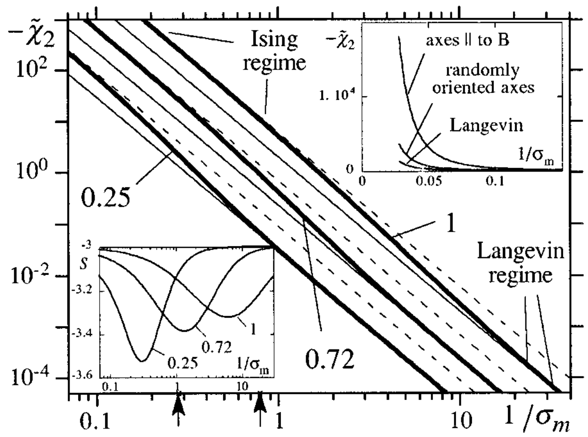

FIG. 1. Log-log plot of the nonlinear susceptibility $-\tilde{\chi}_2$ vs $1/\sigma_m$ (dimensionless T) for a system with randomly oriented anisotropy axes (thick solid lines). Straight lines correspond to Langevin (thin solid) and Ising (dashed) results. The numbers mark the width ρ of the barrier distribution. Within the arrows, the mean slope of the $\rho=0.72$ curve is compared with the experiment of Bitoh *et al.* [Ref. 8(a)]. Bottom inset: logarithmic slopes S vs $1/\sigma_m$. Top inset: effect of the alignment of the axes towards **B** on $-\tilde{\chi}_2$ vs $1/\sigma_m$ for $\rho=0.25$.

and $\chi_2|_{\text{plane rotator}}=-(\mu_0^3 m^4 \sin^4\alpha/16)\beta^3$. For an assembly of identical particles with randomly oriented anisotropy axes, χ_2 is given by

$$\langle\chi_2\rangle_{\text{ran}}=\nu^{-1}\mu_0^3 m^4[\tfrac{1}{30}(2G-3G^2-1)]\beta^3, \tag{7}$$

which, unlike $\langle\chi_0\rangle_{\text{ran}}$ that is given by the Curie law, still depends on K. For a fixed T, χ_2 can largely deviate from those of the limit models: for $k_BT=K\nu/5$ and randomly oriented axes, $(\chi_2-\chi_2|_{\text{Langevin}})/\chi_2\approx 45\%$ and $(\chi_2|_{\text{Ising}}-\chi_2)/\chi_2\approx 60\%$, whereas, for axes collinear with **B**, these values shift to 90% and 80%, respectively.

To assess the effect of the magnetic anisotropy on $\chi_2(T)$, the spontaneous magnetization $M_s(m=\nu M_s)$ will be assumed independent on T. This condition, fulfilled far below the ordering temperature of the magnetic material constituting the particles, yields also temperature independent K's. Facing the subsequent computation of $\chi_2(T)$ for various experimental systems, we consider here the occurrence of distribution in ν [with density $f(\nu)$] but fixed $K>0$ and M_s (i.e., neither distribution in shape, nor size effects on M_s will be accounted). Besides, when the axes of particles with the same ν are randomly oriented, $\chi_2=\int_0^\infty\langle\chi_2\rangle_{\text{ran}} f(\nu)d\nu$. The computed quantity will be $\tilde{\chi}_2=\chi_2[K^3/(\mu_0^3 M_s^4)]$ (dimensionless χ_2) and we shall employ a log-normal distribution for $f(\nu)$ with ν_m as median and ρ as standard deviation of $\ln(\nu)$.

Figure 1 displays in a log-log plot $-\tilde{\chi}_2$ and the corresponding Ising and Langevin results vs the dimensionless temperature $1/\sigma_m=(\beta K\nu_m)^{-1}$ for a number of values of ρ. As the influence of the anisotropy decreases with increasing T, $-\tilde{\chi}_2$ undergoes a smooth crossover from the low-temperature Ising regime to the high-temperature Langevin one. Logarithmic slopes, $S=d\ln(-\tilde{\chi}_2)/d\ln(1/\sigma_m)$, -3 occur in the asymptotic ranges (T^{-3} dependence), but values lesser than -3 emerge in the transitional one, where the departure of $\tilde{\chi}_2$ from a T^{-3} law is sizeable. As ρ increases, the crossover region widens and shifts to higher temperatures owing to the function $\nu^3 f(\nu)$, which determines the particles making the most substantial contribution to χ_2, broadens and moves to larger volumes, whose contribution is Ising like over a wider temperature range. It is pertinent to emphasize here that, henceforth, arguments discarding superparamagnetism based on the departure of $\chi_2(T)$ from a T^{-3} law, as those of Ref. 16, should be carefully scrutinized.

As Fig. 1 shows, the increase of $-\chi_2(T)$ with decreasing T, can be steeper than that of T^{-3}. Hence, when observed over limited temperature windows (e.g., imposed by finite measurement time, t_m), $\chi_2(T)$ can resemble the high-temperature range of a quantity with a low-temperature divergence. This misleadingly suggests presence of interparticle interactions and then, one could fit, e.g., $\chi_2(T)\propto(T-T_c)^{-3}$ (mean-field critical behavior), obtaining false ordering temperatures. The rate of change of $\chi_2(T)$, moreover, enlarges as the axes are aligned towards **B** (see the top inset of Fig. 1): for $\rho=0.25$, the maximum S shifts from -3.53 for axes oriented at random to -3.98 for axes collinear with **B**. ($Mn_{12}Ac$ and textured frozen magnetic fluids are systems with parallel axes.) The mentioned fit of the χ_2 computed for the most diluted sample of Ref. 4(c) over 100 K$\leqslant T\leqslant$180 K (χ_2 was not measured in that work) yields $T_c\approx 17.3$ K (regression 0.99992), whereas experimentally, e.g., for $\omega/2\pi=125$ Hz, t_m effects begin at $T\approx 100$ K. Finally, although less dramatic, those effects also occur when $K<0$, being magnified now as the axes lay perpendicular to **B**.

IV. COMPARISON WITH AVAILABLE EXPERIMENTAL DATA AND PROPOSED EXPERIMENTS

Bitoh *et al.*[8] measured $\chi_2(\omega,T)$ for cobalt particles precipitated in a $Cu_{97}Co_3$ alloy, obtaining from the equilibrium part of the χ_2 vs T curve a mean logarithmic slope -3.17,[8(a)] whose departure from -3 [subsequently unnoticed in Ref. 8(b)] remains unexplained. Their sample appears suitable to check the predicted deviation from $\chi_2\propto T^{-3}$ since (i) the high Curie temperature of the particles ($\approx$1400 K) yields M_s feebly dependent on T in the range of the experiment ($\leqslant$280 K) and (ii) the Curie law fits the equilibrium χ_0 (mean logarithmic slope -1.01), suggesting absence of dipole-dipole interaction effects. [The ascription of the extra $T^{-0.17}$ factor in χ_2 to $M_s^4(T)$ implies the occurrence of its square root in the Curie law ($\chi_0\propto M_s^2$), yielding an incongruent total exponent -1.085 for χ_0.] However, the high amplitude of the ac field (B_0) employed in their experiment ($\nu_m M_s B_0/k_B\approx 17$ K) might have induced saturation effects on the measured χ_2 and χ_0 at low temperatures, moving the $f(\nu)$ derived[8(b)] from χ_0 from the actual volume distribution. Even so, on specializing the above calculation of $\chi_2(T)$ to such a $f(\nu)$ (see Fig. 1), we get a mean slope -3.25 (within 2.6% of -3.17), whose size makes mandatory the inclusion of anisotropy effects to achieve a complete understanding of their experiment.

In addition to search for deviations from $T^{-(2n+1)}$ laws, the angular dependence of χ_{2n} could be measured in systems with oriented axes. Into a polar plot, χ_2 will undergo an increasing deformation as T decreases from a circle at high temperature (isotropic χ_2) towards the two-looped shape of [the Ising regime ($\chi_2|_{\rm Ising} \propto \cos^4\alpha$). In a magnetic fluid, moreover, $\chi_2(T)$ must undergo a discontinuous change at the freezing point (T^*) of the solvent, albeit, if the axes become randomly immobilized at T^*, χ_0 will be continuous there. (For liquid suspensions $\chi_2 = \chi_2|_{\rm Langevin}$ irrespective of K. Hence, as T decreases, χ_2 evolves by a Langevin line of Fig. 1 and, owing to the onset of anisotropy effects at T^*, it abruptly rises to the corresponding anisotropy-dependent curve.) This jump could be smeared out around T^* due to effects related to the immediacy of the critical point of the carrier. The relative size of the discontinuity, $\Delta\chi_2/\chi_2$, depends on T^*, K's, and $f(\nu)$. Hence, $\Delta\chi_2/\chi_2$ would be small for the most diluted sample of Ref. 4(a) (T^* is close to the Langevin regime) whereas, for the corresponding one of Ref. 4(c), it would be about 90%. Once more, if the axes are frozen collinear with **B**, the effect will be even more dramatic.

V. CONCLUSIONS

In summary, (i) applying to a number of systems, formulas for the equilibrium nonlinear susceptibilities of independent overdamped three-dimensional rotators with anisotropy potential $U_A(\theta) = -a\cos^2\theta$, have been derived. (ii) This potential entails crossover from free-rotator regime to either two-state ($a>0$) or plane-rotator ($a<0$) ones, with the departure of the non-linear susceptibilities from the $T^{-(2n+1)}$ laws of these limit models. (iii) The consequent effects, not ruled out by the random average of the anisotropy axes, are large enough to be observable in standard experiments. Hence, it is mandatory to take the presented results into account in any analysis of $\chi_{2n}(T)$ to avoid misinterpretations about the imprints of interparticle interactions in quantities widely used to study collective phenomena in disordered systems.

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APPENDIX: APPROXIMATE FORMULAS FOR χ_2

1. Small-anisotropy range

In order to derive an approximate formula for χ_2 valid in the small-anisotropy range, we shall seek a solution of Eq. (6) in the form of a power series $G = \Sigma_{n=0}^{\infty} a_n \sigma^n$ ($G \equiv F'/F$). For the coefficients a_n, we obtain from Eq. (6), $a_0 = 1/3$ and, for $n \geq 1$, $(n+3/2)a_n = a_{n-1} - \Sigma_{k=0}^{n-1} a_k a_{n-1-k}$. On computing the first few a_n's, G takes the form

$$G \approx \frac{1}{3}\left(1 + \frac{4}{15}\sigma + \frac{8}{315}\sigma^2 - \frac{16}{4725}\sigma^3 - \frac{32}{31185}\sigma^4\right), \tag{A1}$$

where the expansion has been carried out through terms in σ^4 since χ_2 will be obtained up to terms of order σ^3 and Eq. (5) also involves G'. From the power series for F and its derivatives,[2(c)] Eq. (A1) can also be established. Nevertheless, we consider that the procedure employed here is less laborious to obtain further terms in the expansion.

By means of Eqs. (5) and (A1), we obtain

$$\chi_2 \approx -\mu_0^3 m^4 \frac{1}{45}\left[1 + \frac{8}{21}(3\cos^2\alpha - 1)\sigma + \frac{8}{105}(4\cos^4\alpha - \cos^2\alpha)\sigma^2 + \frac{32}{10395}(21\cos^4\alpha - 18\cos^2\alpha + 4)\sigma^3\right]\beta^3, \tag{A2}$$

which yields a good approximation for the exact χ_2 when $|\sigma| \leq 2$. Note that, when the anisotropy axes are oriented at random, the first correction to the $\sigma = 0$ (Langevin) result vanishes.

2. Large-anisotropy ranges

To obtain approximate formulas for χ_2 in the $|\sigma| \gg 1$ ranges, we make in Eq. (6) the substitution $\lambda = 1/\sigma$, getting $-\lambda^2 dG/d\lambda = [(\lambda/2)(1-3G)] + G(1-G)$. On seeking solutions of this equation in the form of a series $G = \Sigma_{n=0}^{\infty} b_n \lambda^n$, we get for the coefficients: $b_0(1-b_0) = 0$, $b_1 = (1/2)(3b_0 - 1)/(1-2b_0)$, and, for $n \geq 2$, $(1-2b_0)b_n = (5/2-n)b_{n-1} + \Sigma_{k=1}^{n-1} b_k b_{n-k}$. [The same method was employed in Ref. 2(c) to derive an asymptotic series for $F(\sigma)$.] Depending on the choice of b_0 we obtain two different solutions

$$G_1 = -1/(2\sigma) \quad (b_0 = 0), \tag{A3a}$$

$$G_2 \approx 1 - 1/\sigma - 1/(2\sigma^2) - 5/(4\sigma^3) \quad (b_0 = 1). \tag{A3b}$$

Owing to $F'/F|_{\sigma\to-\infty} = 0$ and $F'/F|_{\sigma\to\infty} = 1$, we conclude that G_1 and G_2 correspond, respectively, to $\sigma \ll -1$ and $\sigma \gg 1$. [Note that G_1 is an exact solution of Eq. (6) although, as it diverges at $\sigma = 0$, it is not the self-same F'/F.]

On introducing Eqs. (A3) into Eq. (5), we obtain the approximate results

$$\chi_2 \approx -\mu_0^3 m^4 \tfrac{1}{16}\sin^4\alpha[1 + 1/\sigma + (16\cot^2\alpha - 1)/(4\sigma^2)]\beta^3 \quad (\sigma \ll -1), \tag{A4a}$$

$$\chi_2 \approx -\mu_0^3 m^4 \tfrac{1}{3}\cos^4\alpha[1 - 2/\sigma + (3\tan^2\alpha - 1)/(2\sigma^2) + (3\tan^2\alpha - 4)/(2\sigma^3)]\beta^3 \quad (\sigma \gg 1), \tag{A4b}$$

which fit the corresponding exact ones for $|\sigma| \geq 5$.

Finally, we point out that the results (A2) and (A4) together almost cover the entire σ range. Besides, the use of the small-anisotropy approximation, swapped at some point between $|\sigma| = 2$ and $|\sigma| = 4$ by the corresponding large-anisotropy one, would lead to rather acceptable results.

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[15] To compute χ_{2n}, we could also use the relation between F and *error* functions: $F=(\pi/4\sigma)^{1/2}\mathrm{erfi}(\sigma^{1/2})$ for $\sigma>0$ and $F=(\pi/4|\sigma|)^{1/2}\mathrm{erf}(|\sigma|^{1/2})$ for $\sigma<0$. $\mathrm{Erfi}(x)=(4/\pi)^{1/2}\int_0^x \exp(t^2)dt$, and it is related with the Dawson integral $D(x)=\exp(-x^2)\int_0^x \exp(t^2)dt$, a tabulated function which is also available in certain mathematical libraries of computers.

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