# Local-field distribution in systems with dipolar interparticle interaction

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It is shown that the interaction (local) field distribution in disordered systems with dipolar interparticle interaction transforms from the Lorentzian to the Gaussian when the particle volume concentration is increased. To evaluate correctly the local-field dispersion $\overline{h^2}$ in systems with randomly oriented dipoles it is sufficient to take into account the nonmonotonic behavior of the particle radial pair distribution function. For aligned dipoles *angular* correlations of particle positions in the first coordination sphere play a dominant role, strongly decreasing $\overline{h^2}$.

The task of calculating the distribution density of the interaction field for the system of particles carrying electric or magnetic dipole moments arises in a broad spectrum of physical problems, including the determination of the line shape for various magnetic resonances,[1] studies of dipolar glasses,[2–5] and fine magnetic particle systems.[6,7] The latter are widely used now as magnetic recording media,[7] so that the influence of the (dipolar) interparticle interaction on both quasistatic properties (hysteresis loop parameters) and dynamic behavior (stability of the information storage) of such systems are of a great practical interest.[6]

Probably the first rigorous result concerning this problem is due to Anderson.[1] Studying the electron-paramagnetic-resonance line broadening in systems of interacting magnetic dipoles, he showed that the distribution of any interaction field component in a system of dilute aligned dipoles is a Lorentzian. This result was generalized by Klein[3] for the system of dipoles with six allowed orientations (corresponding to possible orientations of $OH^-$ defects in cubic KCl crystals[2]) and by Berkov and Meshkov[9] for dilute ensembles of small ferromagnetic particles which moments can have any orientation. A closely related question plays the central role in single-particle theories of the Ruderman-Kittel-Kasuya-Yosida (RKKY) spin glasses.[10] Due to the same $r^{-3}$ scaling of the RKKY interaction the Lorentzian field distribution was obtained for both Ising model[10] and Heisenberg glass with randomly oriented moments.[11] However, we point out that (in contrast to the dipole-dipole interaction) the RKKY interaction is essentially isotropic, so that complete similarity between systems possessing these two kinds of interactions cannot be expected (see below).

On the other hand, based on the numerical simulation results, Faehnle[12,13] claimed that for amorphous binary alloys $A_{1-x}B_x$ where only atoms $A$ carry a dipole moment, the interaction field distribution $\rho(h)$ is Gaussian for all $x$ in the system with aligned moments[12] and for large concentration of magnetic atoms for randomly oriented dipoles.[13] Though the criterion used in Refs. 12 and 13 is insufficient to find out whether the studied distribution is indeed Gaussian, results listed in Ref. 13 indicate some systematic change of $\rho(h)$ by decreasing $x$.

Hence the important problem which still should be solved concerns the behavior of the interaction field distribution when the concentration of dipole moments increases. In this paper we demonstrate that with increasing concentration of dipoles the interaction field distribution transforms from the Lorentzian to the Gaussian one. Furthermore, we show that spatial correlations of particle positions due to the excluded volume effect significantly influence the distribution width even for moderate particle concentrations. And finally we demonstrate that in systems with aligned dipoles *angular* position correlations for particles which form the first coordination sphere around the trial particle strongly reduce the width of the interaction field distribution.

For the definiteness we consider a system of identical spherical particles carrying (classical) *magnetic* moments but all results can be applied to systems of electric dipoles as well. For the material with the magnetization $M_s$ the particle magnetic moment is $\mu=(4\pi/3)a^3M_s\mathbf{m}\equiv V_pM_s\mathbf{m}$, where $a$ is the particle radius, $V_p$ is the particle volume, and $\mathbf{m}$ denotes the unit vector along the magnetization direction. Using the reduced magnetic field $\mathbf{h}=\mathbf{H}/M_s$ we can write the dipole field created by a particle located at the coordinate origin as

$$\mathbf{h}^{\mathrm{sp}}(\mathbf{r})=V_p\frac{3\mathbf{e}_r(\mathbf{e}_r\mathbf{m})-\mathbf{m}}{r^3}\equiv V_p\frac{\tilde{\mathbf{h}}(\mathbf{e}_r)}{r^3}. \tag{1}$$

If we neglect any spatial correlations in the positions of other particles with respect to the trial one except the restriction $r>r_{\min}=2a$, the interaction field dispersion $\sigma^2_{\alpha\beta}=\langle h_\alpha h_\beta\rangle$ for the system with the particle volume fraction $\eta=NV_p/V$ ($N$ being the particle number, $V$ is the system volume) is given by[9] ($\alpha,\beta=x,y,z$)

$$\sigma^2_{\alpha\beta}=NV_p^2\int_{2a}^{\infty}\frac{\langle\tilde{h}_\alpha(\mathbf{e}_r)\tilde{h}_\beta(\mathbf{e}_r)\rangle}{r^6}d^3r$$
$$=\frac{2\pi^2\eta}{15}\left\{\delta_{\alpha\beta}+\frac{\langle m_\alpha m_\beta\rangle}{3}\right\}. \tag{2}$$

 

The standard Margenau method[8–10] to evaluate the distribution $\rho(h_\alpha)$ of some interaction field component $h_\alpha$ uses its definition as

$$\rho(h_\alpha)=\int_{r>2a} P(\{\mathbf{r}_i\})\,\delta\left(h_\alpha-\sum_{i=1}^{N} h_\alpha^{\rm sp}(\mathbf{m}_i,\mathbf{r}_i)\right)\prod_{i=1}^{N} d^3r_i, \tag{3}$$

where $P(\{\mathbf{r}_i\})$ denotes the probability density to find the $i$th particle at the point $\mathbf{r}_i$ if the trial particle is placed at the coordinate origin. For the dilute system all spatial correlations can be neglected, so that $P(\{\mathbf{r}_i\})=1/V^N$. Using the well-known integral representation of the $\delta$ function, the Fourier transform of, for example, $\rho(h_z)$ can be rewritten in the limit $V\to\infty$ via the one-particle integral[1,9,10]

$$I(k_z)=1-\frac{1}{V}\left\langle\int[1-\exp(-ik_z h_z^{\rm sp}(\mathbf{m},\mathbf{r}))]d^3r\right\rangle \tag{4}$$

as $F(k_z)=I(k_z)^N$. For large $k_z\gg k_{\min}(\sim 1)$ the explicit evaluation of (4) gives $I=|k_z|A(\mathbf{m})V_p/V$, where the function $A(\mathbf{m})$ can be calculated numerically and shows only a weak dependence on the magnetic moment orientation.[9] This $\sim|k_z|$ dependence of $I$ leads directly to the Lorentzian form of $F(h_\alpha)$,[1,9,10] since in this case its Fourier transform takes the form $F(k_z)=I(k_z)^N\sim\exp(-|k_z|NV_p/V)\sim\exp(-\eta|k_z|)$.

The assumption $k\gg k_{\min}$ means that the Lorentzian distribution obtained this way is valid for $h\ll h_{\rm nn}$, where $h_{\rm nn}$ is the field created by the single nearest neighbor ($r_{\min}=2a$), so that some cutoff should be introduced by large field values.[9] For most applications it is sufficient to take the interaction field distribution in the form

$$\rho(h_z)=\frac{C_N}{1+(h_z/\Delta)^2},\quad |h_z|\leqslant h_c \tag{5}$$

and $\rho(h_z)\equiv 0$ for $|h_z|>h_c$. Here $C_N$ denotes the normalization constant and $\Delta\approx 4.54\,\eta$ (Ref. 9) is the distribution width. The cutoff value $h_c$ should be determined from the condition $\langle h_z^2\rangle=\sigma_{zz}^2$ which ensures that the distribution (5) has the correct dispersion (2).

For dilute systems ($\eta\ll 1$) the condition $h\ll h_{\rm nn}$ holds for almost the whole field region of interest, because the probability to find even one neighbor on the distance $r\sim r_{\min}=2a$ is very small. But by increasing the volume concentration this condition is violated for most particles for two reasons: (i) the mean interparticle distance $\bar{r}$ tends to the minimal distance $2a$ and (ii) the spatial correlations in the particle positions become important, so that the assumption $P(\{\mathbf{r}_i\})=1/V^N$ is no longer valid.

The simplest possible way to take these spatial correlations into account is to neglect all except the pair correlations. This requires the introduction of the pair distribution function[14] (PDF) $g(r)$ into (4) leading to

$$I(k_z)=1-\frac{1}{V}\left\langle\int[1-\exp(-ik_z h_z^{\rm sp}(\mathbf{m},\mathbf{r}))]g(r)d^3r\right\rangle. \tag{6}$$

Already for a moderate volume concentration ($\eta\sim 0.1$) $g(r)$ starts to exhibit a peak for $r\approx 2a$ signaling the formation of the short-range order — the first coordination sphere (see, e.g., Ref. 14, Chap. 2). This means that the probability to find a neighboring particle at the distance $r=r_p\approx 2a$ is strongly enhanced, which leads to the increase of the interaction field also.

A quite large maximal number of the nearest neighbors $N_{\max}=12$ (achieved for the perfect hexagonal lattice) together with the trend $\bar{r}\to r_{\min}=2a$ mentioned above allows us to assume that for sufficiently high volume concentration $\eta$ the opposite assumption $|h_z|\gg h_{\rm nn}$ is valid for most particles. Hence we can expand the exponent in (6) up to the second order in the small quantity $k_z h_z^{\rm sp}$ ($k_z\sim 1/h_z\ll 1/h_z^{\rm sp}$). Then the real part of this single-particle integral takes the form

$$I(k_z)=1-\frac{1}{2V}\left\langle\int[k_z h_z^{\rm sp}(\mathbf{m},\mathbf{r})]^2 g(r)d^3r\right\rangle, \tag{7}$$

so that $F(k_z)=I^N(k_z)\sim k_z^2$, which obviously leads to the Gaussian distribution $\rho(h_z)$ with the dispersion

$$\sigma_z^2=\frac{4\pi}{3}\eta\int_{2a}^{\infty}\frac{g(r)dr}{r^4}\int_\Omega \tilde{h}_z^2(\mathbf{m})d\Omega_{\mathbf{m}}. \tag{8}$$

The reasons given above are nothing but a statement that the number of particles $M$ essentially contributing to the interaction field on the given particle is large ($M\gg 1$), so that the central limit theorem can be applied for the evaluation of $\rho(h_\alpha)$. As it can be seen, the dispersion (8) coincides with (2) except that the PDF $g(r)$ is introduced.

To check whether the interaction field distribution for higher particle concentrations becomes indeed Gaussian, we performed direct numerical simulations. For each volume concentration of interest 50 configurations each consisting of 2048 particles were generated choosing particle positions randomly and rejecting particles which would overlap with those already created. Then the interaction field on each particle was calculated assuming periodical boundary conditions and zero demagnetizing field.

To investigate the finite-size effects we have also performed the same simulations for systems consisting of $N=256$, 512, and 1024 particles. With the increasing particle number no systematical changes in the final results were observed for systems with $N\geqslant 512$ particles, so that results presented below (for which $N=2048$) seem to be representative in the thermodynamic limit.

Two examples of the interaction field distributions obtained this way are shown in Fig. 1, where the histograms of simulated distributions for systems with randomly oriented moments are compared with (i) Gaussian distributions with dispersions (8) (solid line) and (ii) the restricted Lorentzian distribution (5). It can be seen, that for $\eta=0.01$ the simulated $\rho(h_z)$ coincides with the corresponding Lorentzian, whereas for $\eta=0.25$ — with the Gaussian distribution. As a quantitative criterion of the difference $D$ between the simulated $\rho_{\rm sim}(h_\alpha)$ and the Gaussian $\rho_G(h_\alpha)$ distributions we have used the integrated absolute value of their difference

$$D=\int_{-\infty}^{\infty}|\rho_{\rm sim}(h_\alpha)-\rho_G(h_\alpha)|dh_\alpha. \tag{9}$$

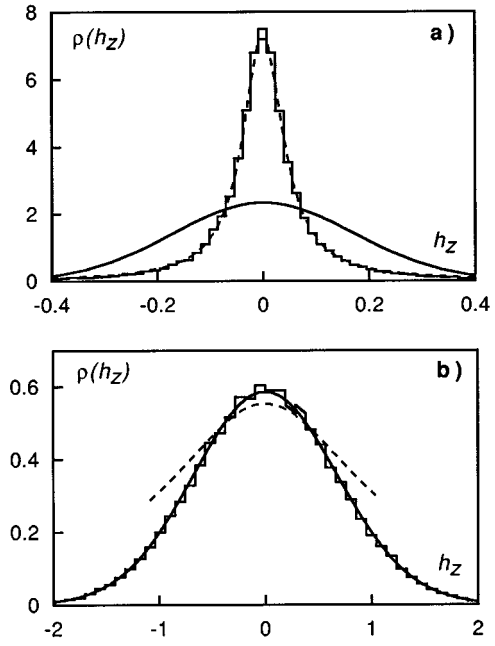


FIG. 1. Distribution densities $\rho(h_z)$ of the $z$ projection of the interaction field for particle volume fractions $\eta=0.01$ (a) and $\eta=0.25$ (b). Histograms are the numerical simulation results, solid lines are the Gaussian distributions with the same dispersion, dotted lines are the restricted Lorentzian distributions (5).

In our case the mean of the corresponding Gaussian distributions ($\bar{h}_\alpha=0$) is known in advance, whereas its dispersion was obtained from the simulated data. Values of $D$ for $\rho(h_z)$ are shown in Fig. 2 as a function of the particle concentration $\eta$ for randomly oriented (triangles) and aligned (crosses) dipole moments. With increasing particle concentration the simulated distribution converges to the Gaussian more rapidly for the randomly oriented system, which is obviously due to the additional disorder arising from the random orientation of dipole moments in comparison with the aligned system.

To find out, for which concentrations the difference between the simulated and the Gaussian distributions are statistically significant, we applied the $\omega^2$ criterion.[15] It allows us to calculate the probability that the difference between the

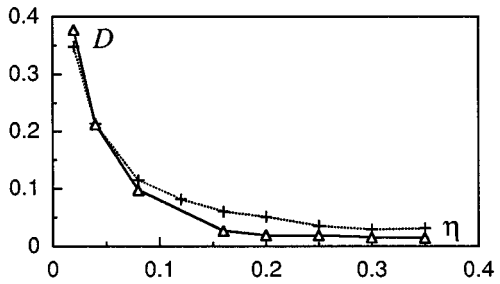


FIG. 2. Integrated differences $D$ [Eq. (9)] between the simulated and the corresponding Gaussian distribution densities as functions of the particle volume fraction $\eta$ for randomly oriented (triangles) and aligned (crosses) moments.

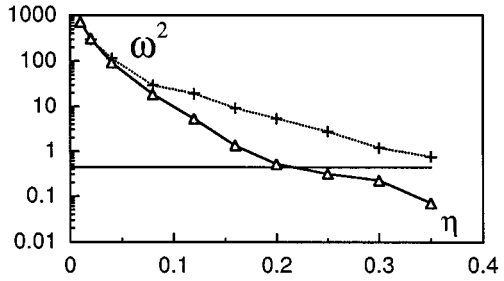


FIG. 3. Values of the $\omega^2$ criterion (calculated to check whether the distribution obtained numerically is a Gaussian one) versus particle volume fraction $\eta$ for randomly oriented (triangles) and aligned (crosses) moments. $\omega^2$ values above the horizontal line (corresponding to the 95% probability level) indicate statistically significant deviations from the Gaussian distribution.

known distribution function (Gaussian in our case) and the distribution function derived from some empirical data set exceeds some prescribed value if the distribution law of these data is indeed Gaussian.

The dependences of $\omega^2$ values in the particle volume fraction $\eta$ are shown in Fig. 3 for randomly oriented (triangles) and aligned (crosses) dipoles. The horizontal line represents the usual probability level $P=95\%$, which for the $\omega^2$ criterion calculated using the Gaussian distribution with the dispersion derived from the empirical data is $\omega^2(P=0.95)=0.442$.[15] All $\omega^2$ values greater than this one should, strictly speaking, lead to the conclusion that the studied distribution is *not* Gaussian, i.e., deviations from the Gaussian distribution are statistically significant. It can be seen that for randomly oriented moments the distribution can be treated as Gaussian for $\eta\geqslant 0.2$, whereas for aligned moments small (see Fig. 2), but statistically significant deviations from the Gaussian distribution occur up to the highest concentration $\eta=0.35$ available by such simulations.

Unfortunately, it was not possible to perform the same $\omega^2$ test for the Lorentzian distribution for all values of $\eta$, except the smallest ones ($\eta<0.03$), where the distribution was clearly Lorentzian. The reason is that the analytical form (5) does not describe the distribution tails correctly even for moderate volume concentration. This leads to the wrong values of the $\omega^2$ criterion which is very sensitive to such details.

We found that the dispersion of the interaction field distribution strongly depends on the orientation degree of the particle moments. In Fig. 4 dependences $\overline{h_z^2}(\eta)$ obtained using different methods are shown for randomly oriented [Fig. 4(a)] and aligned [Fig. 4(b)] dipoles. Crosses indicate values obtained by numerical simulations, straight solid lines represent linear dependences (2) which neglect all spatial correlations except the condition $r>2a$ and open triangles stand for $\overline{h_z^2}$ values obtained by (8), i.e., taking into account radial pair correlations only. The corresponding PDF $g(r)$ was accumulated for all configurations obtained during numerical simulations. It can be seen that the latter approach provides good agreement with the numerical simulation results for the randomly oriented dipoles [Fig. 4(a)], whereas for the aligned system [Fig. 4(b)] it fails to predict even the correct trend of the $\overline{h_z^2}(\eta)$ dependence when compared with the ideal case

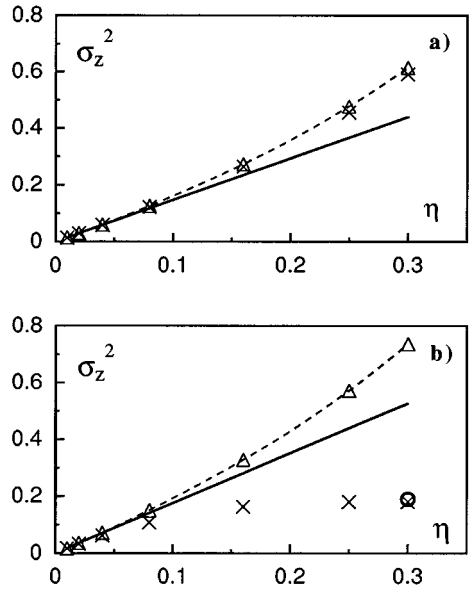


FIG. 4. Interaction field dispersion $\overline{h_z^2}$ as function of the particle volume fraction $\eta$ for randomly oriented (a) and aligned (b) dipoles. Crosses are the numerical simulation values, solid lines are the ideal dependences (2), triangles are the values calculated from (8), i.e., taking into account radial correlations only. Dashed lines through triangles are a guide for the eye. The open circle on (b) represents the value obtained for $\eta=0.3$ when perfect short-range order is assumed (see text for details).

(straight line). This behavior differs also from the corresponding dependence in RKKY spin glasses, where $\overline{h_z^2}$ was found to be larger for the aligned system than for the chaotic one.[11]

We assumed, that for the aligned system $\overline{h_z^2}$ is strongly reduced due to the *angular* correlations of the *positions* of particles forming the first coordination sphere around the trial particle. These particles make the dominant contribution to $\overline{h_z^2}$ and correlations of their angular positions would reduce $\overline{h_z^2}$ for the system with aligned moments because the only disorder left is due to the random orientation of this first coordination shell relative to the given particle. To test this assumption we calculated $\overline{h_z^2}$ for $\eta=0.3$ assuming that the nearest neighbors of the trial particle form the perfect hexagonal lattice and the positions of other neighbors are uncorrelated except that they are outside the first coordination shell $(r>4a)$. To compute the contribution to $\overline{h_z^2}$ from this hexagonal shell we performed numerical averaging over its possible orientations given by the corresponding Euler angles. The result [total $\overline{h_z^2}(\eta=0.3)\approx 0.189$] is shown in Fig. 4(b) by the open circle. It is in good agreement with the value $\overline{h_z^2}\approx 0.179$ obtained numerically.

In conclusion, we have shown that in the disordered system of particles with dipolar interaction the distribution density of the local (interaction) field transforms from the Lorentzian to the Gaussian form when the particle volume fraction $\eta$ increases. The transition for randomly oriented dipoles is completed by $\eta\approx 0.2$, whereas for the aligned system even for the highest studied particle volume fraction $\eta=0.35$ statistically significant deviations from the Gaussian distribution have been found. Already for moderate particle concentrations $\eta\geqslant 0.1$ the local-field dispersion is strongly influenced by the radial and — for systems of aligned dipoles — angular correlations of the particle positions, so that these correlations should be explicitly taken into account.

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