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## Local Field Distributions in Systems with Dipolar Interaction \*

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We investigate systematically the local field distribution functions of up-spins for systems of dipolar interaction. with particular emphasis on Ising-type lattice systems. It is found that as the fraction increases, the shape of the distribution function changes from Lorentzian to Gaussian. In addition, sub-peaks can be induced in the distribution function by non-cubic lattice structures. This is in stark contrast with a dilute gas system, where the distribution has only one Lorentzian peak for any up-spin fraction.

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The dipolar field interaction is an important issue.<sup>[1-12]</sup> In this Letter, we provide a thorough study of the local field distribution function in Ising-type systems with particular emphasis on the lattice system.

For Ising-type spins, e.g., molecular magnets Fe<sub>8</sub> and  $Mn_{12}$ ,<sup>[1-3]</sup> the dipolar interaction between them is

$$d(\boldsymbol{r}) = \frac{E_D(1 - 3\cos^2\theta)\Omega_0}{r^3},\qquad(1)$$

where r is the displacement between spins,  $\theta$  the angle between r and the easy axis, and  $E_D$  the strength of the interaction. For a lattice,  $\Omega_0$  is the volume of the unit-cell; for a gas,  $\Omega_0$  is the volume of a particle.

When there is randomness in either spin configuration or spatial configuration or both, each spin feels a different field strength. In this kind of situation, rather than to know exactly the field strength felt by each individual spin, it is usually more important to find the distribution function of the local fields felt by the spins. The distribution function can be computed with the Margenau method, [13-15] where one chooses a test spin and computes how the local field felt by the test spin changes with different spin, spatial, or both configurations. If there is no correlation between space and spin degrees of freedom, the distribution function can be written as

$$P(\xi) = \sum_{\{r_j\}} \mathcal{P}(\{r_j\}) \sum_{\{s_j\}} \mathcal{Q}(\{s_j\}) \delta\left(\xi - \sum_{j=1}^N s_j d_j\right), \quad (2)$$

where  $\mathcal{P}(\{r_i\})$  and  $\mathcal{Q}(\{s_i\})$  are the probabilities of a given spatial configuration and spin configuration, respectively; the summations are over all possible configurations;  $d_j = d(\mathbf{r}_j)$  is the dipole field generated by a spin at  $r_i$ ;  $s_i = \pm 1$  indicates the spin up or down; and N is the number of particles in the system.

In a dilute gas system, the spatial distribution configuration function is uniform. In a lattice system,

however, because the spins are fixed in space, the distribution function of the system does not depend on  $\mathcal{P}(\{r_i\})$  anymore. In both dilute gas systems and spin lattice systems, the probabilities related to the spin degree of freedom can be characterized by the fraction of up-spin states and we use F to indicate this fraction. Due to the symmetry of the system, we shall focus only on the case of F < 1/2 and treat the downspins as the background.

We compute the distribution functions of local field for a dilute gas system firstly. This serves for two purposes. On the one hand, the results for a dilute gas may find applications in ultra-cold molecule systems;<sup>[5]</sup> on the other hand, these results can be compared to the previous known results for dipolar gases and our results for lattice systems to be presented later.

We define a concentration parameter as

$$\eta = \frac{N\Omega_0}{V},\tag{3}$$

where V is the system volume. For a dilute gas, we have  $\eta \ll 1$ . In this dilute limit, the correlation between the particle positions can be safely ignored and the particles can be regarded as randomly scattered in the space. Therefore, for a dilute dipolar gas, when the fraction of up-spin is F, Eq. (2) takes the following form,

$$P(\xi, F) = \frac{1}{V^N} \int \prod_{j=1}^N d^3 r_j \sum_{n=0}^N F^n (1 - F)^{N-n} \\ \times \sum_{\{s_j\}_n} \delta(\xi - \sum_{j=1}^N s_j d_j),$$
(4)

where the summation is over all possible spin configurations when each spin is fixed in space. To compute **6**Y

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the distribution function, it is convenient to compute first its Fourier transform,

$$\bar{P}(k,F) = \int P(\xi,F) \exp(-ik\xi) d\xi$$
$$= \left[\frac{1}{V} \int [F \exp(-ikd_j) + (1-F) \exp(ikd_j)] d^3r_j\right]^N.$$
(5)

In the limit of  $V \to \infty$ , for a fixed gas concentration  $N/V = \eta$ , using Fresnel sine and cosine integral,<sup>[16]</sup> we obtain

$$\bar{P}(k,F) = \lim_{V \to \infty} \left[ 1 - \frac{1}{V} (\mathcal{A}_1 | k | \Omega_0 - i \mathcal{A}_2 k \Omega_0) \right]^{V \frac{\eta}{\Omega_0}}$$
$$= \exp\left( -\mathcal{A}_1 | k | \eta + i \mathcal{A}_2 k \eta \right),$$
(6)

with

$$\mathcal{A}_{1} = \frac{8\pi^{2}E_{D}}{3^{5/2}},$$

$$\mathcal{A}_{2} = \frac{(1-2F)2\pi E_{D}}{3} \left(\frac{4}{3} - \frac{4}{9}\sqrt{3}\ln\left(2+\sqrt{3}\right)\right).$$
(8)

After a simple reverse Fourier transform, we find that the distribution function  $P(\xi, F)$  is Lorentzian

$$P(\xi, F) = \frac{A_1 \eta / \pi}{A_1^2 \eta^2 + (\xi + A_2 \eta)^2}.$$
 (9)

This recovers the result by Berkov and Meshkov.<sup>[11,12]</sup> However, we want to emphasize that our result here is in a sense more general than Berkov and Meshkov's although we consider only Ising-type spin. In Refs. [11,12], the orientation of magnetic moment of the particle is random, equivalent to F = 1/2 in our system. Consequently, it is unclear how the distribution function would change if the magnetic moment (or spin) has a favored direction. In contrast, it is clear from our result in Eq. (9) that the distribution does not change its height and width as F changes, only the peak position shifts with F.

In deriving Eq. (9), we have implicitly assumed that the shape of the system is spherical. For other shapes, the peak position is shifted and the distribution function becomes

$$P(\xi, F) = \frac{\mathcal{A}_1 \eta / \pi}{\mathcal{A}_1^2 \eta^2 + (\xi + \mathcal{A}_2 \eta + 2CE_D \eta (2F - 1))^2},$$
(10)

where the parameter C is the shape coefficient of the sample (Generally,  $C = 2\pi(\frac{1}{3} - N_g)$  in which  $N_g$  is the demagnetization parameter of the sample. For ellipsoid sample with three axes a b and c located in the x, y and z axes respectively, the demagnetization parameter  $N_g = \frac{1}{2}abc \int_0^\infty \frac{dx}{(x+c^2)\sqrt{(x+a^2)(x+b^2)(x+c^2)}}$  for the magnetic field along the z axis).

We have also investigated the distribution function with Monte Carlo simulation and they match almost perfectly with our analytical results in Eq. (9).

For the general case, the shape of the system only shifts the position of the peak and we find perfect agreement between our theoretical results<sup>[17]</sup> and the Monte Carlo simulation.



Fig. 1. The local dipolar field distribution of a dilute gas with F = 0.015 and  $\eta = 0.01$ . The red solid line is the theoretical result of Eq. (9) and the blue area is the Monte Carlo simulation result. The inset shows an enlarged portion of the distribution peak, clearly indicating that the peak center deviates from zero.

We now turn to lattice systems, where each lattice site has one Ising-spin. Molecular magnets such as Fe<sub>8</sub> and Mn<sub>12</sub> are this type of spin lattice system:<sup>[1-3]</sup> at low temperature, only two ground states  $|s_z = \pm 10\rangle$ are occupied in these systems. In these molecular systems,  $\Omega_0$  is the volume of the unit cell of the lattice and  $E_D = \frac{2\mu_0}{4\pi} \frac{(gS\mu_B)^2}{\Omega_0}$ , where  $\mu_B$  is Bohr magneton. For Fe<sub>8</sub> and Mn<sub>12</sub>, S = 10 and  $g \approx 2$ . Similar spin lattice systems have been investigated for the dipolar line width.<sup>[9]</sup>

Because all spins are fixed in space in a lattice system, there is only one spatial configuration. As a result, the distribution function Eq. (2) becomes<sup>[3]</sup>

$$D(\xi, F) = \sum_{m=0}^{N} F^m (1-F)^{N-m} \sum_{\{s_j\}} \delta\Big(\xi - \sum_{j=1}^{N} s_j d_j(\mathbf{r})\Big).$$
(11)

We use  $D(\xi, F)$  to distinguish the distribution function in a spin lattice system from the one in a dilute gas system. Similarly, we compute its Fourier transform first

$$\bar{D}(k,F) \equiv \int d\xi D(\xi,F) \exp(-ik\xi)$$

$$= \prod_{j=1}^{N} [F \exp(-ikd_j) + (1-F) \exp(ikd_j)].$$
(12)

We have used this equation to compute numerically how the distribution function  $D(\xi, F)$  changes with F. The results are shown in Fig. 2. When F is very small, the distribution function is a Lorentzian. As the F increases, the distribution function is broadened with the appearance of side-peaks. When F is close

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to 1/2, the side-peaks merge with the main peak and the distribution function is again a one-peak function. However, this one-peak function has a Gaussian shape, instead of Lorentzian. In the following, we analyze in detail the two limiting cases:  $F \ll 1$  and  $F \approx 1/2$ .



Fig. 2. The change of the local dipolar field distribution function  $D(\xi, F)$  of  $Mn_{12}$  with the up-spin fraction F. From (a) to (f), F = 0.012, 0.05, 0.10, 0.15, 0.30, and 0.50, respectively. All the red solid lines in (b)–(e) are obtained with the reverse Fourier transform of Eq. (12) and the blue area are Monte Carlo simulation results. In (a), the red dashed line is obtained from Eq. (16); in (f), the red dashed line is obtained from Eq. (19). The black solid lines in (a) and (f) are obtained from Eq. (12). To show clearly the fitting between the black lines and the red lines in (a) and (f), the Monte Carlo results for F = 0.012 and F = 0.50 are not shown.

We consider the limiting case,  $F \ll 1$ , which means that most of the spins in the lattice system are downspins. For this case, using Fresnel sine and cosine integral,<sup>[16]</sup> we obtain

$$\bar{D}(k,F) \approx \exp\left[ik\sum_{j=1}^{N} d_j - F\sum_{j=1}^{N} (1 - e^{-2ikd_j})\right]$$
$$= \exp\left[-|k|A_1 + i\left(\sum_{j=1}^{N} d_j + A_2\right)k\right],$$
(13)

where

$$A_{1} = \frac{16\pi^{2}E_{D}F}{3^{5/2}},$$

$$A_{2} = \frac{4\pi F E_{D}}{3} \left( -\frac{4}{3} + \frac{4}{9}\sqrt{3}\ln\left(2 + \sqrt{3}\right) \right).$$
(15)

With a reverse Fourier transform, we arrive at

$$D(\xi, F) = \frac{A_1/\pi}{A_1^2 + (\xi + \sum_{j=1}^N d_j + A_2)^2}.$$
 (16)

This shows that when  $F \ll 1$ , the distribution function  $D(\xi, F)$  is a Lorentzian with its half-width proportional to the up-spin fraction F. This confirms the numerical result in Fig. 2(a). The summation term,  $\sum_{j=1}^{N} d_j$ , is the background field created by a lattice where all spins are down.

It is not a coincident that the small F limit of the lattice system has a Lorentzian distribution similar to the gas system. When F is small, the averaged

distance between up-spins is much larger than the lattice constant. Consequently, they can be viewed as a gas floating in the background of a down-spin lattice. When this small fraction F of up-spins is viewed as a gas, this "gas" has all the spin pointing up and the fraction F should be identified with the gas concentration parameter  $\eta$ . With these facts in mind, one can easily derive  $D(\xi, F \ll 1)$  from  $P(\xi, F)$ .

When the lattice is populated roughly with equal numbers of up-spins and down-spins, that is,  $F \sim 1/2$ , Eq. (12) is approximately

$$\ln \bar{D}(k,F) \approx \sum_{j=1}^{N} \ln \left\{ \cos(kd_j) \right\}, \tag{17}$$

where terms proportional to |F-1/2| are omitted. In a cubic lattice such as molecular magnet Mn<sub>12</sub>, we argue that  $kd_j$  can always be regarded as small. There are two reasons. First, when k is large, each cosine function in  $\overline{D}(k, F)$  oscillates very fast and  $\overline{D}(k, F)$ becomes essentially zero. This is confirmed by our numerical result. Secondly, our computation shows  $\{d_j\}_{\max} \approx 0.64$ . With these considerations, we can further approximate  $\overline{D}(k, F)$  with a Taylor expansion,

$$\ln \bar{D}(k,F) \approx -\frac{1}{2}k^2 \sum_{j=1}^{N} d_j^2.$$
 (18)

This leads to a Gaussian distribution function

$$D(\xi, F \sim 1/2) = \frac{1}{\sqrt{2\pi \sum_{j=1}^{N} d_j^2}} \exp\Big(-\frac{\xi^2}{2\sum_{j=1}^{N} d_j^2}\Big),$$
(19)

which is already shown in our numerical result Fig. 2(f).

There is an intuitive way to understand why the distribution function becomes Gaussian when F approaches 1/2. These up-spins can be divided into many small fractions with each fraction contributes a Lorentzian to the overall distribution. According to the central limit theorem, the overall distribution then should be Gaussian. We note that Berkov has found that the distribution function for a dense gas is Gaussian<sup>[12]</sup> due to the spatial correlation. Although a lattice can be viewed as correlated in space, it is not apparent that the Gaussian distribution in a spin lattice system has the same origin as the Gaussian distribution in a dense gas system. The above intuitive understanding seems to suggest that these two Gaussian distributions have different origins.

We have also checked our results with the Monte Carlo simulation. Again, we find excellent agreement with our theoretical results from Eq. (12).

The shape effect of the sample only shifts the peak in distribution function and we find nearly perfect agreement between our theoretical calculation<sup>[17]</sup> and Monte Carlo simulation.



**Fig. 3.** The local dipolar field distributions of the Fe<sub>8</sub> system. The blue areas indicate the the Monte Carlo simulation results and the red solid line is the numerical result obtained with Eq. (12). (a) F = 0.30, (b) F = 0.50, (c) F = 0.60, and (d) F = 0.80.

In the above, we have focused on the cubic lattice system. Here we consider non-cubic lattice systems, and use molecular magnet Fe<sub>8</sub> as an example. Fe<sub>8</sub> has a triclinic lattice with its shortest axis almost parallel to the easy axis.<sup>[17]</sup> We have computed the distribution functions of Fe<sub>8</sub> for different fractions, just as shown in Fig. 3. It is clear that in Fe<sub>8</sub> system, all the distributions have three peaks centered at the same positions and they differ only by the sizes of each peaks. This is different from the distribution function of Mn<sub>12</sub>, where the peaks change their positions with F and even merge into one peak when F is either small or close to 1/2.

The peaks in Fig. 3 are the result of the triclinic lattice structures of Fe<sub>8</sub> and the sub-peaks position can be explained by the nearest spins. We find that all of these peaks are Gaussian when F is not very small. We have also compared these results with the Monte Carlo simulation and there is a very good agreement.

For the cubic  $Mn_{12}$  spin lattice system, the situation becomes different. In this case, all axes in three directions have the same length and all 64 nearest spin configurations have to be considered since none of the 64 configurations dominate. The results of the competition among these 64 spin configurations are shifting side-peaks seen in Fig. 2.

In summary, we find that the distribution function is Lorentzian when the up-spin fraction is very small in both systems. In a lattice system, it changes to a Gaussian when the fraction approaches one half. In addition, the asymmetry in lattice can induce multiple Gaussian peaks in distribution function.

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- 017302 Effect of Substrate Temperature on the Structural and Raman Properties of Ag-Doped ZnO Films WANG Li-Na, HU Li-Zhong, ZHANG He-Qiu, QIU Yu, LANG Ye, LIU Guo-Qiang, QU Guang-Wei,
- 017303 Structural, Electronic and Optical Properties of KTa<sub>0.5</sub>Nb<sub>0.5</sub>O<sub>3</sub> Surface: A First-Principles Study

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- 017701 The Comprehensive Retrieval Method of Electromagnetic Parameters Using the Scattering Parameters of Metamaterials for Two Choices of Time-Dependent Factors HOU Zhi-Ling, KONG Ling-Bao, JIN Hai-Bo, CAO Mao-Sheng, LI Xiao, QI Xin
- 017801 Nanostructured Metal-Enhanced Photoluminescence of Micro- $Sr_2Si_5N_8:Eu^{2+}$  Phosphors LIU Ling, XU Xiao-Liang, LEI Jie-Mei, YIN Nai-Qiang
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- 017804 The Annealing-Induced Shape Deformation of Hydrothermal-Grown ZnO Nanorods ZHENG Zhong-Kui, DUANMU Qing-Duo, ZHAO Dong-Xu, WANG Li-Dan, SHEN De-Zhen
- 017805 The Morphological Change of Silver Nanoparticles in Water WANG Peng, WANG Rong-Yao, JIN Jing-Yang, XU Le, SHI Qing-Fan

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- 018101 The Growth of Semi-Polar ZnO (1011) on Si (111) Substrates Using a Methanol Oxidant by Metalorganic Chemical Vapor Deposition SANG Ling, WANG Jun, SHI Kai, WEI Hong-Yuan, JIAO Chun-Mei, LIU Xiang-Lin, YANG Shao-Yan, ZHU Qin-Sheng, WANG Zhan-Guo
- 018102 A Study of GaN Grown on SiH<sub>4</sub> Pre-Treated 6H-SiC Substrates SONG Shi-Wei, LIANG Hong-Wei, LIU Yang, XIA Xiao-Chuan, SHEN Ren-Sheng, LUO Ying-Min, DU Guo-Tong
- 018103 The Field Emission Characteristics of Titanium-Doped Nano-Diamonds YANG Yan-Ning, ZHANG Zhi-Yong, ZHANG Fu-Chun, DONG Jun-Tang, ZHAO Wu, ZHAI Chun-Xue, ZHANG Wei-Hu
- 018104 The Photovoltaic Properties of BiFeO<sub>3</sub>La<sub>0.7</sub>Sr<sub>0.3</sub>MnO<sub>3</sub> Heterostructures LUO Bing-Cheng, CHEN Chang-Le, FAN Fei, JIN Ke-Xin
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- 018501 Reactive Radiofrequency Sputtering-Deposited Nanocrystalline ZnO Thin-Film Transistors LI Shao-Juan, HE Xin, HAN De-Dong, SUN Lei, WANG Yi, HAN Ru-Qi , CHAN Man-Sun, ZHANG Sheng-Dong
- 018701 Temporal Correlation-Based Spatial Filtering of Rician Noise for Functional MRIs Amir. A. Khaliq, I. M. Qureshi, Jawad. A. Shah
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- 019601 An Internal Heating Model to Elucidate the Shape of a Small Planetary Body LI Gen, CHEN Chu-Xin
- 019701 Reanalysis of the Isotopic Mixture of Neutron-Capture Elements in the Metal-Poor Star HD 175305

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### ERRATA AND OTHER CORRECTIONS

019901 Erratum: Multilayer Antireflection Coating for Triple Junction Solar Cells [Chin. Phys. Lett. 28 (2011) 047802] ZHAN Free WANG Hei Li, HE Li Free WANG here HUANG the Same NI Hei Give NIII Zhi Glo

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