# RANDOM LOCAL FIELD METHOD FOR THE DESCRIPTION OF PHYSICAL PROPERTIES OF DISORDERED SOLIDS\*

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We suggest an approach to describe the physical properties of disordered dielectric and/or magnetic systems. These systems are characterized by randomly positioned and oriented spins (dipoles) in a host crystal lattice. The ensemble of these spins or dipoles create the random magnetic or electric fields in a host lattice. Their distribution function, defined as an average (over spatial and orientational fluctuations) of Dirac delta contributions of each spin (dipole), enables us to obtain the self-consistent equations for order parameters like average magnetization (polarization)  $\langle S \rangle$ , and/or general quantities like  $\langle S^n \rangle$ . We calculate explicitly the above distribution functions for different types of interactions and show that, in general, they are not Gaussian. Our theory delivers pretty good description of experiments in disordered ferroelectrics, multiferroics, magnets and diluted magnetic semiconductors.

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## 1. Introduction

Many physical systems can be well described in terms of the properties of a certain fictitious particle under the action of random fields, originating from its environment [1, 2, 3]. One example of such complex systems is the disordered solids having localized dipole (disordered ferroelectrics) and/or spin (disordered magnets, diluted magnetic semiconductors) moments embedded in otherwise ordered host crystal lattice, see *e.g.* [4, 5].

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The problem of cooperative behavior of above impurities is not fully understood and attracts much attention of the scientists. The main problem here is the randomness in the impurity subsystem. The impurity effects, which may be of static (like smearing of phase transition into the phase with long-range order like ferroelectric or ferromagnetic) or dynamic (like long-time relaxation of the order parameter) nature are inherent in disordered ferroelectrics [4], magnets [6], ceramic superconductors (see [7] and references therein) and other complex systems [3]. For example, in highly polarizable dielectrics such as KTaO<sub>3</sub> with dipole impurities (off-center ions Li<sup>+</sup>, Nb<sup>5+</sup> or Na<sup>+</sup>) the ferroelectric phase transition occurs at low temperatures if impurity concentration n exceeds some critical value  $n_{\rm cr}$  (see [8] and references therein). At  $n < n_{\rm cr}$  these substances are in the dipole glass phase ("dipole analog" of spin glass). The mixed ferro-glass phase exists in the intermediate range of concentrations [9]. Such a rich phase diagram suggests many anomalous (as compared to ordered ferroelectrics) properties, the most intriguing one is the unusual long-time relaxation of the order parameter [10].

The disordered dielectrics is a typical example of the systems, where impurities interact via known potential  $J(\vec{r})$ . In this case the existing methods of spin glass theory (like Sherrington–Kirkpatrick or Edwards–Anderson approaches based on the so-called replica formalism [11]) cannot be applied directly since the explicit form of interaction between impurity dipoles is important for the description of their observable behavior (see [8] and references therein). Although replica formalism (see, *e.g.* [11]) operates directly with a free energy of a disordered system as a self-averaging quantity, it cannot incorporate the predefined interaction potential  $J(\vec{r})$ . Our random field approach is able to incorporate  $J(\vec{r})$  (see, *e.g.* [8,12,13]) and it can get the free energy (see, *e.g.* [14,15,16]) via self-consistent equations for the order parameters. Latter equations, in turn, depend explicitly on the distribution function of random fields, for which a nonlinear integral equation is obtained within the framework of suggested random field approach.

In this paper, we present the random local field method from the point of view of its probabilistic sense. We will derive explicitly the distribution functions of random fields and relaxation times for different disordered solids and show the correspondence between different interaction (between disorder constituents like impurity spins or dipoles) potentials and the character of the distribution function.

The presented formalism can be generalized for any kind of disordered systems with predefined interactions between impurities. Another (than disordered dielectrics) example of such systems may be the semimagnetic semiconductors, where localized spins interact with each other indirectly via RKKY interaction (see *e.g.* [17] and references therein).

#### 2. General formalism

There are many sources of randomness in the disordered solids. The substitutional disorder, vacancies, off-center ions (dipoles), impurity spins and other imperfections are the sources of random electric, magnetic and elastic fields in the system. All random field sources can be regarded as the impurities in some (ideal) host lattice, called reference phase. For this reason hereafter we call them "impurities".

Consider a system of impurities chaotically distributed over the sites of the host crystal lattice with concentration n = N/V (N is a number of impurities, V is a crystal volume). For the case of dipole impurities in a dielectric host the Hamiltonian has the form

$$\mathcal{H}_{e} = \sum_{ij\alpha\beta} \mathcal{K}_{\alpha\beta}(\boldsymbol{r}_{ij}) l_{i}^{\alpha} l_{j}^{\beta} + \sum_{i\alpha} \left[ \mathcal{E}_{\alpha}(\boldsymbol{r}_{i}) + \mathcal{E}_{\alpha} \right] l_{i\alpha} , \qquad (1)$$

where  $l_i$  is the unit vector pointing along the direction of impurity dipole  $d_i$  at the point  $r_i$  ( $d_i = dl_i$ ),  $\mathcal{E}$  is an external electric field in energy units,  $\mathcal{E}(r_i)$  is a random internal field (in energy units) of nondipole impurities (*i.e.* of point defects *etc.*),  $\alpha$ ,  $\beta = x, y, z$  and  $r_{ij} = r_j - r_i$ . Summation in (1) is performed over the host lattice sites occupied by impurities. In dielectrics, the interaction potential  $\mathcal{K}_{\alpha\beta}(r)$  signifies the interaction of dipoles through transversal optical phonons of a host lattice and has following general form [18]

$$\mathcal{K}_{\alpha\beta}(\boldsymbol{r}) = -\frac{1}{2} \frac{d^2}{\varepsilon_0 r^3} \left[ f_1(r/r_c) \delta_{\alpha\beta} + (3m_\alpha m_\beta - \delta_{\alpha\beta}) f_2(r/r_c) \right], \quad \boldsymbol{m} = \frac{\boldsymbol{r}}{r}, \ (2)$$

where  $\varepsilon_0$  and  $r_c$  is a host lattice static dielectric permittivity and correlation radius (two impurities at a distance less or equal  $r_c$  are correlated). The physical reason for appearance of  $r_c$  and functions  $f_{1,2}(r/r_c)$  is an indirect interaction of impurity dipoles via the host lattice soft phonon mode (see, *e.g.* [18]). If the latter interaction is absent, we have  $r_c \to 0$ ,  $f_1(r/r_c) \to 0$ ,  $f_2(r/r_c) \to 1$  so that (2) gives the ordinary dipole–dipole interaction. The functions  $f_{1,2} \sim \exp(-r/r_c)$ ; their detailed form is unimportant for present consideration.

For the case of magnetic impurities with localized spins, the interaction term in the Hamiltonian (1) becomes isotropic due to its exchange nature

$$\mathcal{H}_m = \sum_{ij} J(\boldsymbol{r}_{ij}) \boldsymbol{S}_i \boldsymbol{S}_j + \sum_i \boldsymbol{H}_0 \boldsymbol{S}_i \,. \tag{3}$$

Here  $S_i$  is a spin operator (it also can be large classical spin) at the point  $r_i$  and  $H_0$  is the external magnetic field. The Hamiltonian (3) describes disordered magnetic systems and so-called diluted magnetic semiconductors [14], in particular.

For latter substances the interaction between the impurity spins occurs via conduction electrons and has the Ruderman–Kittel–Kasuya–Yosida (RKKY) form [19]

$$J(\mathbf{r}) = J_0 F(2k_{\rm F}r), \qquad F(y) = \frac{y\cos y - \sin y}{y^4},$$
 (4)

where interaction constant  $J_0$  is expressed via conduction electron effective mass and electron-ion exchange constant,  $k_{\rm F}$  is Fermi wave vector. We note here that the reciprocal Fermi wave vector  $1/k_{\rm F}$  plays a role of correlation radius in this case, *i.e.* two spins on the distance less then  $1/k_{\rm F}$  are correlated. In other words, in dilited magnetic semiconductors, those spins are correlated which have wave vectors  $k > k_{\rm F}$ .

The Hamiltonians (1) and (3) incorporate two types of randomness. The first is random spatial position of the impurity spin or dipole in a host lattice, constituting the so-called spatial disorder. The second is random orientation of dipole (or projection of spin) in its site due to thermal fluctuations. The averaging over two above types of disorder just gives the actual shape of distribution function of random electric or magnetic fields. Proceeding formally, we rewrite the Hamiltonians (1) and (3) in the following equivalent form

$$\mathcal{H}_{e} = \sum_{i\alpha} E_{i\alpha} l_{i\alpha}, \qquad \mathcal{H}_{m} = \sum_{i} \boldsymbol{H}_{i} \boldsymbol{S}_{i},$$

$$E_{i\alpha} = \sum_{j\beta} \mathcal{K}_{\alpha\beta}(\boldsymbol{r}_{ij}) l_{j}^{\beta} + \mathcal{E}_{\alpha}(\boldsymbol{r}_{i}) + \mathcal{E}_{\alpha},$$

$$H_{i\alpha} = \sum_{j} J(\boldsymbol{r}_{ij}) \boldsymbol{S}_{j} + \boldsymbol{H}_{0} \qquad (5)$$

and introduce the distribution function of random (electric or magnetic) field E defined as the average value of Dirac-delta contributions  $E_i$  to the effective field E

$$f(\boldsymbol{E}) = \left\langle \overline{\delta \left( \boldsymbol{E} - \sum_{i} \boldsymbol{E}_{i} \right)} \right\rangle, \qquad (6)$$

where bar denotes the averaging over random spatial positions of impurities (spatial averaging) and angular brackets denote the thermal averaging over possible dipole (or spin) orientations. Without loss of generality hereafter we use "dielectric" notations for random fields keeping in mind that for magnetic case it is easy to restore the proper notations. Our next step is to perform the spatial and thermal averagings in Eq. (6).

The spatial averaging procedure in (6) can be done with the help of  $\delta$ -function integral representation

$$f(\boldsymbol{E}) = \frac{1}{(2\pi)^3} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \exp\left(i\sum_{\alpha} E_{\alpha}\rho_{\alpha}\right) \left\langle \exp\left[-i\sum_{i\alpha} \rho_{\alpha}E_{i\alpha}\right] \right\rangle d^3\rho \,.$$
(7)

To perform the averaging we assume that the impurities do not form clusters [20], *i.e.* we consider single impurity in the (fluctuating) field of the rest of them but not pair in the field of pairs, triple in the field of triples *etc.* In other words, the probability for single impurity dipole (spin) to be in some position (*i.e.* in the infinitesimal volume  $V_1$ ) in a host lattice is uniform with probability density equal to  $1/V_1$ . It can be shown that this assumption gives pretty good description of the disordered dielectric and magnetic systems everywhere except a narrow region near the phase transition temperatures.

The realization of above scenario yields

$$\left\langle \overline{\exp\left[-i\sum_{i\alpha}\rho_{\alpha}E_{i\alpha}\right]}\right\rangle \approx \left[\frac{1}{V}\int_{V}\overline{\left\langle \exp\left[-i\sum_{i\alpha}\rho_{\alpha}E_{i\alpha}\right]\right\rangle}dV\right]^{N}$$
$$= \left[1+\frac{1}{V}\int_{V}\overline{\left\langle \exp\left[-i\sum_{i\alpha}\rho_{\alpha}E_{i\alpha}\right]-1\right\rangle}dV\right]^{N}.(8)$$

Introducing then the impurity concentration n = N/V (N is the number of impurities, V is system volume) and passing to thermodynamic limit  $N \to \infty, V \to \infty, n = \text{const}$ , we obtain finally

$$\left\langle \exp\left[-i\sum_{i,\alpha}\rho_{\alpha}E_{i\alpha}\right]\right\rangle = \exp\left[n\int_{V}\left\langle \exp\left[-i\sum_{\alpha}\rho_{\alpha}E_{\alpha}(\boldsymbol{r})\right] - 1\right\rangle d^{3}\boldsymbol{r}\right] . (9)$$

To perform further averagings in Eq. (9), we should expand the exponential in power series and average term by term the corresponding configurations. This procedure leads to following self-consistent integral equation

for distribution function  $f(\mathbf{E})$ 

$$f(\boldsymbol{E}) = \int_{\rho} \exp\left[i\rho(\boldsymbol{E}-\mathcal{E}) - n\int_{V} \Psi(\boldsymbol{r})d^{3}r\right] \frac{d^{3}\rho}{(2\pi)^{3}},$$
  
$$\Psi(\boldsymbol{r}) = \int_{E} \left\langle 1 - \exp\left[-i\sum_{\alpha,\beta} \mathcal{K}_{\alpha\beta}(\boldsymbol{r})\rho_{\alpha}l_{\beta}\right] \right\rangle_{E} f(\boldsymbol{E})d^{3}E, \qquad (10)$$

where  $\langle .. \rangle_E$  means the averaging with single particle Hamiltonian  $\mathcal{H}_1 = \sum_{\alpha} l_{\alpha} E_{\alpha}$ .

Expression (10) is the integral equation for distribution function  $f(\mathbf{E})$ . In general case, this equation can be solved only numerically. However, in many cases (*e.g.* those considered below) it is possible to avoid a solution of the integral equation, since in these cases it is exactly reducible to the set of transcendental equations for order parameters, *i.e.* for macroscopic quantities like  $\langle \mathbf{l}^n \rangle$  (or  $\langle \mathbf{S}^n \rangle$ ),  $n \geq 1$  of the system. In such cases it is possible to perform the thermal averagings explicitly.

We note here, that since nondipole impurities have no internal degrees of freedom (like several possible orientations of an impurity dipole or spin in a host lattice), they are not subject to thermal averaging in (9). Moreover, it can be shown (see, *e.g.* [13]) that distribution function of many random field sources is a convolution of distribution functions of individual sources.

Thermal averaging in (9) depends on particular number of impurity dipole orientations (or impurity spin projections) in a host lattice. As an example, we consider the case of impurity dipole with six possible orientations. This corresponds to the particular case of Li<sup>+</sup> in KTaO<sub>3</sub> lattice. Let us first calculate auxiliary thermal averages  $\langle l_{\alpha} \rangle_E$ ,  $\langle l_{\alpha}^2 \rangle_E$ ,  $\alpha = x, y, z$ . We have

$$\langle l_{x,y,z} \rangle_E = -\frac{\sinh\beta E_{x,y,z}}{\cosh\beta E_x + \cosh\beta E_y + \cosh\beta E_z} , \langle l_{x,y,z}^2 \rangle_E = \frac{\cosh\beta E_{x,y,z}}{\cosh\beta E_x + \cosh\beta E_y + \cosh\beta E_z} ,$$
(11)

 $\beta = (k_{\rm B}T)^{-1}$ . Here, we used the fact that  $l_{\alpha} = \pm 1$  for  $\alpha = \pm x, \pm y, \pm z$  (6 directions of [100] type). From equation (9), we have now

$$\left\langle \exp\left[-i\sum_{\alpha,\beta}K_{\alpha\beta}\rho_{\alpha}l_{\beta}\right] - 1\right\rangle_{E}$$

$$= \frac{\operatorname{Tr}\exp\left[-i\sum_{\alpha,\beta}\mathcal{K}_{\alpha\beta}\rho_{\alpha}l_{\beta} - \beta\sum_{\alpha}l_{\alpha}E_{\alpha}\right]}{\operatorname{Tr}\exp\left[-\beta\sum_{\alpha}l_{\alpha}E_{\alpha}\right]} - 1$$

$$= \frac{\cosh(\beta E_{x} - iQ_{x}) + \cosh(\beta E_{y} - iQ_{y}) + \cosh(\beta E_{z} - iQ_{z})}{\cosh\beta E_{x} + \cosh\beta E_{y} + \cosh\beta E_{z}} - 1, (12)$$

where

$$Q_{x} = \mathcal{K}_{xx}\rho_{x} + \mathcal{K}_{xy}\rho_{y} + \mathcal{K}_{xz}\rho_{z},$$

$$Q_{y} = \mathcal{K}_{xy}\rho_{x} + \mathcal{K}_{yy}\rho_{y} + \mathcal{K}_{yz}\rho_{z},$$

$$Q_{z} = \mathcal{K}_{xz}\rho_{x} + \mathcal{K}_{yz}\rho_{y} + \mathcal{K}_{zz}\rho_{z},$$

$$\mathcal{K}_{\alpha\beta} \equiv \mathcal{K}_{\alpha\beta}(\boldsymbol{r}),$$
(13)

and  $\mathcal{K}_{\alpha\beta} \equiv \mathcal{K}_{\beta\alpha}$  (see (2)). Converting the hyperbolic functions of the imaginary argument into trigonometric functions with respect to the auxiliary averages (11), we have finally

$$\left\langle \exp\left[-i\sum_{\alpha,\beta} K_{\alpha\beta}\rho_{\alpha}l_{\beta}\right] - 1 \right\rangle = \left\langle l_{x}^{2} \right\rangle \left(\cos Q_{x} - 1\right) + \left\langle l_{y}^{2} \right\rangle \left(\cos Q_{y} - 1\right) + \left\langle l_{z}^{2} \right\rangle \left(\cos Q_{z} - 1\right) + i\left[\left\langle l_{x} \right\rangle \sin Q_{x} + \left\langle l_{y} \right\rangle \sin Q_{y} + \left\langle l_{z} \right\rangle \sin Q_{z}\right].$$
(14)

Here for brevity, we suppress index E in the thermal averages, *i.e.* put  $\langle ... \rangle_E \equiv \langle ... \rangle$ . The next step is to obtain the self-consistent equations for the components of the average dipole moment of the system  $L_{\alpha} \equiv \langle \overline{l_{\alpha}} \rangle$  and its average square  $M_{\alpha} \equiv \langle \overline{l_{\alpha}^2} \rangle$ . The first quantity is, indeed, an effective dipole representing the system as a whole (*i.e.* ferroelectric long-range order parameter or spontaneous polarization). Above equation (10) shows that to achieve self-consistency, we have to replace in Eq. (14) the single-particle averages  $\langle l_{\alpha} \rangle$  and  $\langle l_{\alpha}^2 \rangle$  by  $L_{\alpha}$  and  $M_{\alpha}$ , respectively. In this case, the distri-

bution function has the form

$$f^{(6)}(\boldsymbol{E}, \boldsymbol{L}, \boldsymbol{M}) = \frac{1}{(2\pi)^3} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \exp\left(i\sum_{\alpha} E_{\alpha}\rho_{\alpha}\right) \\ \times \exp\left[F_1^{(6)}(\rho) + iF_2^{(6)}(\rho)\right] d^3\rho, \\ F_1^{(6)}(\rho) = n \int_{V} \left\{M_x \left(\cos Q_x - 1\right) + M_y \left(\cos Q_y - 1\right) \right. \\ \left. + M_z \left(\cos Q_z - 1\right)\right\} d^3r, \\ F_2^{(6)}(\rho) = n \int_{V} \left\{L_x \sin Q_x + L_y \sin Q_y + L_z \sin Q_z\right\} d^3r,$$
(15)

where

$$L_{x,y,z} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \langle l_{x,y,z} \rangle f^{(6)}(\boldsymbol{E}, \boldsymbol{L}, \boldsymbol{M}) dE_x dE_y dE_z,$$
  
$$M_{x,y,z} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \langle l_{x,y,z}^2 \rangle f^{(6)}(\boldsymbol{E}, \boldsymbol{L}, \boldsymbol{M}) dE_x dE_y dE_z.$$
(16)

Here superscript denotes the number of dipole orientations. The expressions (16) are equivalent to a set of six quite cumbersome equations, determining the concentrational and temperature dependencies of both L and M.

The real part  $F_1^{(6)}(\rho)$  of the logarithm of the characteristic function defines the width of the distribution function, *i.e.* the dispersion of the random fields. The imaginary part  $F_2^{(6)}(\rho)$  determines the mean value of the random field, *i.e.* the first moment of the distribution function; it incorporates the components of the long-range order parameter. This means that function  $F_2^{(6)}(\rho)$  is "responsible" for possible long-range order in the system, while  $F_1^{(6)}(\rho)$  tends to destroy it. It can be shown that  $F_2(\rho) \neq 0$  for impurity dipoles only. This means that long-range order in the system (which occurs when mean value of the random field exceeds its dispersion) is due to the dipoles. Nondipole impurities can only destroy the long-range order, giving rise to the dipole glass state. The situation is qualitatively similar for spin systems.

The application of the above formalism for eight possible orientations (of [111] type) of the impurity dipole (this corresponds to Nb<sup>5+</sup> in KTaO<sub>3</sub>) yields very cumbersome set of equations for  $f^{(8)}(\boldsymbol{E}, \boldsymbol{L})$ , which we do not adduce here.

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The simplest case is the situation when impurity dipole has two permissible orientations in a host lattice. This corresponds also to disordered magnetic system (*e.g.* diluted magnetic semiconductor) with spin S = 1/2, see [14, 15]. The self-consistent equation for order parameter  $L \equiv L_z$  reads (see, *e.g.* [21])

$$f^{(2)}(\boldsymbol{E}, \boldsymbol{L}) \equiv f(E, L) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp(i\rho E) \exp[F_1(\rho) - iF_2(\rho)] d\rho,$$
  

$$F_1(\rho) \equiv F_1^{(2)}(\rho) = n \int_{V} \{\cos(\mathcal{K}_{zz}\rho) - 1\} d^3r,$$
  

$$F_2(\rho) \equiv F_2^{(2)}(\rho) = nL \int_{V} \sin(\mathcal{K}_{zz}\rho) d^3r \equiv \rho E_0(\rho)L,$$
  

$$L = \int_{-\infty}^{\infty} \tanh(\beta E) f(E, L) dE.$$
(17)

Note, that the suggested procedure of calculation of the random field distribution function can be also applied for classical spin. In this case, the auxiliary single particle thermal averages can be expressed through Langevin function and its derivatives. The self-consistent procedure in this case can be done only implicitly and we do not even know what kind of order parameters (and how many) appear as a result of this procedure.

The form of  $F_i(\rho)$  and thus  $f(\mathbf{E})$  depends strongly on the form of  $\mathcal{K}_{\alpha\beta}(\mathbf{r})$ or  $J(\mathbf{r})$ . Consequently, the general form of  $f(\mathbf{E})$  will not be Gaussian, this limit is realized at high impurities concentration only, corresponding to  $\rho \to 0$ . For instance,  $f(\mathbf{E})$  is a Lorentzian when  $r_c \to 0$  in (2), *i.e.* for the case of the pure dipole–dipole interaction.

### 2.1. Asymptotics of distribution functions

As high impurity concentration corresponds to  $\rho \to 0$ , we can expand the corresponding characteristic function in power series and obtain corresponding distribution functions explicitly. The easiest way of doing so is to consider the case (17), corresponding to spin 1/2 in magnetic systems.

We observe that the lowest order in  $\rho$  is first order so that up to this order in (17)  $F_1^{(2)}(\rho) \equiv 0$  and

$$F_2^{(2)}(\rho) = L\rho E_0, \qquad E_0 = n \int_V \mathcal{K}_{zz}(r) d^3 r.$$

Substitution of this equation back into f(E, L) gives for it the delta-function

$$f_{\rm MF}(E,L) = \delta(E - E_0 L) \,,$$

corresponding to well-known mean field approximation for Ising model. Substitution of this delta-function into the equation for order parameter L also gives well-known mean filed equation for Ising model

$$L = \tanh\left(\frac{LE_0}{k_{\rm B}T}\right)\,,\tag{18}$$

which at  $L \to 0$  generates the equation for mean field transition temperature  $k_{\rm B}T_{\rm cMF} = E_0 = n \int_V \mathcal{K}_{zz}(\mathbf{r}) d^3 r.$ 

Next approximation in  $\rho$  generates the finite width of distribution function

$$F_1^{(2)}(\rho) = -\Delta E \frac{\rho^2}{2}, \qquad \Delta E = n \int\limits_V \mathcal{K}_{zz}^2(\boldsymbol{r}) d^3 r$$

so that f(E, L) in this approximation is Gaussian

$$f_{\rm G}(E,L) = \frac{1}{\sqrt{2\pi\Delta E}} \exp\left[-\frac{(E-E_0L)^2}{2\Delta E}\right].$$
 (19)

It is seen that function (19) is normalized  $\int_{-\infty}^{\infty} f_{\rm G}(E,L)dE = 1$ .

Next terms in  $\rho$  expansions generate already non-Gaussian distribution functions. The physical meaning of above asymptotic solution is that the less disorder (*i.e.* more dipoles or spins occupying the free host lattice cites) has a system, the narrower is the distribution function of random fields. So, if there is almost no disorder, the distribution function is delta-function, signifying the validity of a mean field approximation, for a little stronger disorder we have Gaussian distribution function and for stronger fluctuations the function becomes non-Gaussian.

# 3. The correspondence between interaction potentials and distribution functions

We have shown above that for weak disorder the system has Gaussian distribution function of random field (19). In the opposite limiting case of strong disorder, the distribution function of random fields is the heavy-tailed non-Gaussian distribution. To model the effect of strong disorder, we assume that our system has small numbers of dipoles or spins, interacting via long-range (like dipole–dipole) interaction

$$\mathcal{K}^{dd}_{\alpha\beta}(\boldsymbol{r}) = \frac{d^2}{\varepsilon_0 r^3} \left( \delta_{\alpha\beta} - 3m_\alpha m_\beta \right) \,. \tag{20}$$

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We show that such interaction generates the Lorentzian distribution of random field. To be more specific, we consider the system of electric dipoles (or Ising spins) at zero temperature having two possible orientations along z-axis. In this case  $\mathcal{K}^{dd}_{\alpha\beta} = \mathcal{K}^{dd}_{zz}$  and we have

$$f_L(E) = \lim_{\substack{N \to \infty \\ V \to \infty}} \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{iE\rho} [\Psi(\rho)]^N d\rho ,$$
$$\Psi(\rho) = \frac{1}{2V} \left( \int_V e^{-i\rho \mathcal{K}_{zz}^{dd}(\boldsymbol{r})} d^3r + \int_V e^{i\rho \mathcal{K}_{zz}^{dd}(\boldsymbol{r})} d^3r \right) .$$
(21)

With respect to explicit form of  $\mathcal{K}_{zz}^{dd}$ , we obtain

$$\Psi(\rho) = 1 - \frac{\alpha}{N} |\rho|, \qquad \alpha = \frac{8\pi^2}{9\sqrt{3}} \frac{nd}{\varepsilon_0}, \qquad (22)$$
$$f_L(E) = \frac{1}{2\pi} \lim_{N \to \infty} \int_{-\infty}^{\infty} e^{iE\rho} \left(1 - \frac{\alpha}{N} |\rho|\right)^N d\rho$$
$$= \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{iE\rho - \alpha |\rho|} d\rho = \frac{\alpha}{\pi (E^2 + \alpha^2)}. \qquad (23)$$

Function (23) is a Lorentzian and, as it is easy to see, normalizable. Although any distribution function of random fields (not only Lorentzian) behaves similarly at the impurity concentration variation, the Lorentzian has its own peculiarities. Namely, the function (23) at any z does not admit any moments. This shows its physical meaning, namely that this distribution (regardless of value of z) occurs in solids with strong disorder. Also, the Lorentzian (23) is centered at E = 0, while Gaussian (19) at the point  $E_0L$ . This is because for Lorentzian case the disorder is so strong that it suppresses long-range order parameter L.

The interaction (20) for classical spins generates the 3D Lorentz distribution (see also [22]). Denoting for a moment  $E \equiv |\mathbf{E}|$ , we obtain

$$f_L^{3D}(E) = \frac{1}{(2\pi)^3} \int e^{i\boldsymbol{E}\rho - \delta\rho} d^3\rho = \frac{\delta}{\pi^2 (E^2 + \delta^2)^2},$$
  
$$4\pi \int_0^\infty E^2 f_L^{3D}(E) dE = 1, \qquad \delta = \frac{\alpha}{2},$$
 (24)

where parameter  $\alpha$  is defined by Eq. (22).

For a six-orientable dipole, the interaction (20) generates the distribution function, which is a product of partial functions in x, y and z directions

$$f(\mathbf{E}) = \frac{1}{\pi^3} \frac{\delta_x \delta_y \delta_z}{(\delta_x^2 + E_x^2)(\delta_y^2 + E_y^2)(\delta_z^2 + E_z^2)}, \\ \delta_x = \frac{nd}{\varepsilon_0} 2.462051, \qquad \delta_y = \frac{nd}{\varepsilon_0} 1.69643, \qquad \delta_z = \frac{nd}{\varepsilon_0} 1.493629.(25)$$

The intermediate case between Gaussian function (19) and the above Lorentzians is achieved at lowering of the dipoles or spin concentration along with corresponding temperature variations. Generally, at concentration lowering and fixed temperature, the distribution function varies from Gaussian to Lorentzian "through" Holtzmarkian with characteristic function logarithm  $\sim \rho^{3/2}$ . The easiest way to analyze this is to consider the case of Ising spins 1/2 (or two-orientable dipoles) (17). In Fig. 1 we plot schematically the general behavior of distribution function of random fields on the example of latter model. This behavior is qualitatively similar to the random field model for arbitrary impurity spin or the number of orientations of impurity dipole. Namely, the curves 1–3 of Fig. 1 correspond to long-range order existence with  $L \neq 0$ . Curve 1 corresponds to the largest impurity concentration and largest L (which occurs at low enough temperatures), being symmetric Gaussian. Curve 2 is asymmetric because at L > 0 all significant random field configuration lay at positive fields. Curve 3 still corresponds to the long-



Fig. 1. Schematic plot of random fields distribution function at different degree of disorder (figures near curves). Curve 1 corresponds to Gaussian function  $(L \sim 1 \text{ and large } z = nr_c^3)$ , curve 2 — intermediate  $L \sim 0.5$  and z, curve 3 — small L and lower (then in curve 2) z, curves 4 and 5 comprise Lorentz curves, corresponding to L = 0. Parameter z for curve 5 is lower then that for curve 4.

range order (more precisely, to the mixed ferro-glass phase) with smaller L and impurities concentrations. Curves 4 and 5 correspond to purely glassy phases with absence of long-range order (L = 0) and wide distributions of random fields. Curve 5 has the lowest impurity concentration z. At further diminishing of z the width of distribution function goes to infinity while its amplitude to zero.

### 4. Discussion and conclusions

Our random field treatment is valid (within approximations made, see above) for arbitrary law of interaction between impurity dipoles or spins as well as for arbitrary number of the dipole orientations or spin projection. Both classical and quantum treatment of spins is possible. The distribution functions of random field, which appear in our treatment of disordered solids, are essentially non-Gaussian, the Lorentzian functions being their limiting case for strongest possible disorder in a system. The fact that distribution functions are non-Gaussian, plays an important role in the glassy (dipole and spin glass) phases formation in the above disordered solids.

Our random field theory deliver pretty good description of experiments about concentrational dependence of ferroelectric phase transition temperature in disordered dielectrics [23] as well as the temperature and concentrational dependences of their equilibrium thermodynamic characteristics like dielectric permittivity and/or specific heat [8,24]. The same had been done for polymeric disordered ferroelectics [25], where the dynamical properties had also been considered. The equilibrium thermodynamics of diluted magnetic semiconductors, predicting the ferromagnetic phase transition in them, had been put forward in Refs. [14,15]. For the system with localized spins 1/2 our method permits to construct the free energy function, which, in turn, makes possible to consider many problems of these substances within well developed phenomenological approach. Among them is the theory of domain structure in disordered ferroelectrics [26] and magnetic semiconductors [16] as well as the dynamical properties within Landau–Khalatnikov approach [22].

Let us finally say more about different physical systems, where heavytailed non-Gaussian distribution functions appear. The first example, where non-Gaussian distributions are employed, is the dynamics of amorphous materials like conventional glasses (see, *e.g.*, [28]), the long-time relaxation in spin [5, 28] and orientational glasses [4]. It had also been shown (see [27]) that random electric fields with the above Lorentzian distribution function, while acting between dipole impurities in different kinds of disordered dielectrics, generate long-time logarithmic relaxation in them. This relaxation is a source of many experimentally observed anomalies. One more impor-

tant application of non-Gaussian PDFs is the theory of inhomogeneously broadened resonant lines [12]. Such broadening occurs in condensed matter and/or biological species, in a number of spectroscopic manifestations like the electron paramagnetic resonance (including the microwave spectroscopy of so-called Josephson media in granular high  $-T_c$  superconductors [7]), nuclear magnetic resonance, optical and neutron scattering methods. The broadening arises due to random electric and magnetic fields, strains and other perturbations from defects in a substance containing the centers whose resonant transitions between energy levels are studied. The usual technique of calculation of shapes of such resonant lines is the so-called statistical method [12]. Our expressions (23), (24) and (25) have actually been obtained in the framework of this method. This method determines the line shape as an averaged (over spatial disorder and over defects intrinsic degrees of freedom) delta-function, stemming from the resonance of a single center. The outcomes depend strongly on the distribution of the centers, their concentration, and interactions between them. In the above statistical method, the Gaussian PDF is a reliable outcome in the case of large center concentration and strong short-range interaction (it may be even a contact interaction) between them, being itself a suitable limiting case. The opposite limiting case refers to small center concentrations and long-range (like dipole–dipole) interaction between them. It gives rise to the Lorentz distribution of random fields, see above. The intermediate regimes generate various non-Gaussian PDFs, with the Holtzmark function among them [12]. Long-time experimental research on resonant techniques confirms its efficiency in determination of the resonant line shapes, which generally are by no means Gaussian. The above discussion shows that many physical systems are characterized intrinsically by non-Gaussian (especially Lorentzian) distributions. In many cases, they can be obtained (through intermediate PDFs like the Holtzmark one) from the Gaussian PDF by varying the system parameters, as for example the temperature and concentration of defects in the case of above disordered dielectrics and magnets.

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