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Dynamic susceptibility of a spin ice near the critical point

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We consider spin ice magnets (primarily, $Dy_2Ti_2O_7$) in the vicinity of their critical point on the (H,T) plane. We find that the longitudinal susceptibility diverges at the critical point, leading to the behaviour qualitatively similar to the one which would result from non-zero conductance of magnetic charges. We show that dynamics of critical fluctuations belongs to the universality class of easy-axis ferroelectric and calculate logarithmic corrections (within two-loop approximation) to the mean-field critical behavior.

Spin ice [1] is a kind of geometrically frustrated magnetic materials with a structure of low-energy states presenting deep similarity with usual water ice, those H-bond structure was analized long ago by Pauling [2]. The most studied examples of spin ice include pyrochlore oxides $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$; their structure is shown in Fig.1. Spin ice demonstrates a number of peculiar

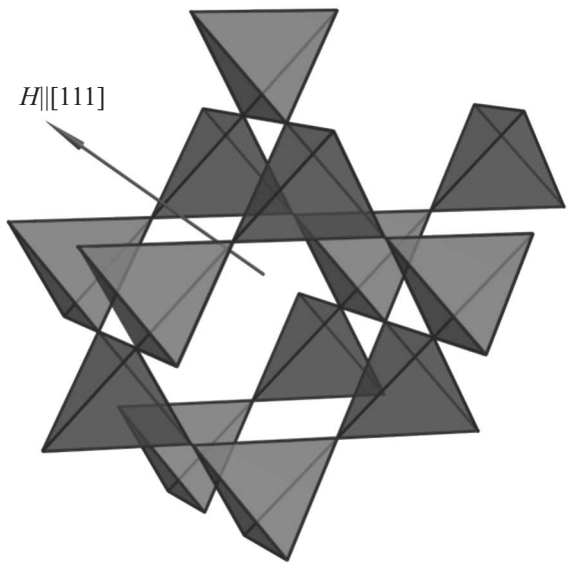

Fig.1. Pyrochlore lattice consists of corner-sharing tetrahedra; spins of dysprosium are siting at their vertices

low-temperature properties, the major of the are: i) an extensive residual entropy, and ii) elementary excitations resembling magnetic monopoles [3, 1]. Low temperature dynamics of spin ice is governed almost solely by these magnetic excitations, therefore one may expect spin ice to demonstrate phenomena similar to those known for the electrolytes. Indeed, it was recently shown that some aspects of spin ice behaviour can be described in terms of "magnetolyte" similar to the Onsager theory [4] of electolytes [5]. It is also known [1], that at low enough temperatures spin ice undergoes a first-order transition as function of applied magnetic field H. The first-order transition line in the (H,T) plane terminates at the critical point H_c, T_c. In particular, $H_c = 0.9\,\mathrm{T}$ and $T_c = 0.38\,\mathrm{K}$ in the case of $Dy_2Ti_2O_7$. In the present Letter we calculate dynamic longitudinal magnetic susceptibility of spin ice near this critical point; our results predict the behaviour formally similar to the one expected for the media with a nonzero "magnetoconductance". We will explain, however, that it would be incorrect to interpret these results in terms of "direct current" of monopoles. Specific numerical estimates will be done for $Dy_2Ti_2O_7$, whereas our general scheme is applicable to any pyrochlore spin ice.

The crucial difference between spin ice and electrolyte is the finite static magnetic susceptibility of the former as opposed to the diverging dielectric low-frequency response $\epsilon(\omega) \propto i\sigma/\omega$ of any electrolyte with a nonzero conductivity σ. The origin of this difference can traced to the fact that magnetic monopoles of spin ice are the sources of magnetization field $\mathbf{M}$ (or, equivalently, magnetic field $\mathbf{H}$), but they *do not generate* magnetic induction $\mathbf{B}$, which is the proper analog of electric field $\mathbf{E}$ for electrostatics [6].

Following [7], one can represent linear magnetic susceptibility of spin ice as

$$\chi(\omega, T) = \frac{1}{\chi_0^{-1}(T) - i\omega/\Gamma}, \qquad (1)$$

where $\chi_0(T) \sim \mu_B^2/(V_0 T)$ is static susceptibility (μ_B is the Bohr magneton and V_0 is the volume of an elementary cell). Therefore relaxation of magnetization after the sudden switching of magnetic field is given by

$$M(t) = \chi_0 H(1 - \exp[-\Gamma\chi_0^{-1} t]) \qquad (2)$$

At the small timescales $t \ll \chi_0/\Gamma$ the relaxation process (2) resembles direct conductivity with "magnetic cur-

rent" proportional to the external magnetic field: $j_m = dM/dt = \Gamma H$, thus the magnetic analog of conductivity is $\sigma_m = \Gamma$. However, finite value of static susceptibility $\chi_0(T)$ limits possible observation of magnetoconductivity to a transient regime only.

We emphasize that Eq.(1) for dynamic susceptibility $\chi_0(H,T)$ with some effective relaxation rate $\Gamma(H,T)$ should be valid in a broad range of temperatures and fields, when monopoles are present. Spin ice in the absence of external field, and spin ice in sufficiently strong field when kagome ice structure develops, represent the two well-defined limiting cases. Below we consider the vicinity of the critical point (H_c, T_c) where static susceptibility is expected to diverge, leading to a broad timescale for the observation of magnetoconductivity. We will use virial expansion in order to estimate the parameters of the Ginzburg-Landau free energy $F\{m\}$ which describes fluctuations in the critical region. Then we use this $F\{m\}$ functional to construct an effective Martin-Siggia-Rose functional [8 – 10] which describes critical dynamics of spin ice, and study it up to the two-loop logarithmic approximation.

The Hamiltonian of the spin ice is of the form [1]

$$H = -\frac{1}{2}\sum_{i,j} J_{ij}(\mathbf{e}_i \cdot \mathbf{e}_j) S_i S_j - g\mu_B \mathbf{H} \sum_i \mathbf{e}_i S_i + \\ + Da^3 \sum_{(ij)} \left[\frac{\mathbf{e}_i \cdot \mathbf{e}_j}{|\mathbf{r}_{ij}|^3} - \frac{3(\mathbf{e}_i \cdot \mathbf{r}_{ij})(\mathbf{e}_j \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5} \right] S_i S_j. \quad (3)$$

Here $S_i = \pm 1$ is an effective Ising variable which describes sign of magnetic moment $\mu_i = S_i \mathbf{e_i}$ of the i-th Dysprosium ion; the magnitudes $|\mu_\mathbf{i}| = \mu \approx 10\mu_B$, whereas $\mathbf{e}_i$ are the unit vectors along easy axis, see Fig.2, the direction of axes being chosen with respect

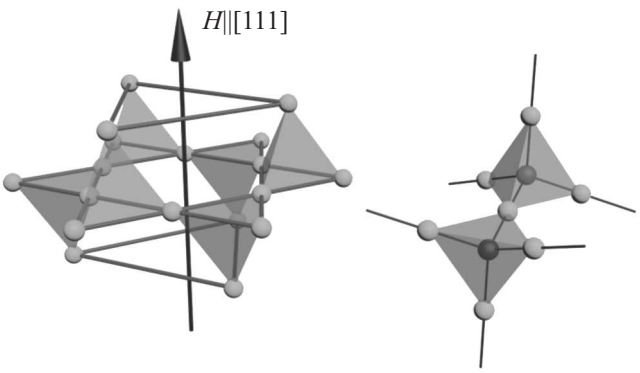

Fig.2. Pyrochlore lattice can be represented as a stack of triangular and kagome lattices, with easy axis directions being aligned with the bonds of diamond lattice, which is dual to the pyrochlore lattice. Magnetic monopoles are situated in the centers of tetrahedra

to one of the tetrahedral sublattices, see Fig.1. Parameters J_{ij} describe exchange interactions between first-, second- and third-order neighbours, their magnitudes were determined [11] as $J_1 = -3.72\,\mathrm{K}$, $J_2 = 0.1\,\mathrm{K}$ and $J_3 = -0.03\,\mathrm{K}$ respectively. External magnetic field $\mathbf{H}$ is directed along the [111] axis, and the last term in Eq.(3) stands for the magnetic dipole-dipole interaction, the dipole interaction constant $D = \mu^2/a^3 = 1.41\,\mathrm{K}$, where $a = 3.54\,\text{Å}$ is the pyrochlore lattice constant (it is equal to the nearest-neighbour distance, see Fig.2).

We will study the system described by the Hamiltonian (3) near the critical point, where large average magnetization $\bar{M}_\parallel$ along the field direction is induced. Deviation m of the actual magnetization $M_\parallel$ from its average value at the critical point $\bar{M}_\parallel(H_c, T_c)$ can be considered as an order parameter which describes the state of the system in the vicinity of the critical point. The corresponding Ginzburg-Landau free energy functional is of the form

$$\mathcal{F} = \int dV \left[\frac{1}{2} m(a + \hat{b}) m + \frac{\lambda}{4!} m^4 - mh \right], \quad (4)$$

where $a = \alpha(T - T_c)$ and $h = H - \gamma(T - T_c)$. In a system with local interactions one would find $\hat{b} = bk^2 = -b\Delta$, but our case is different due to the presence of strong dipole-dipole interactions. Similar problem of second-order phase transition in uniaxial ferroelectric was considered in the seminal paper [12], where it was shown that in presence of dipole-dipole interaction operator $\hat{b}$ should be modified as follows:

$$\hat{b} = bk^2 + 4\pi x^2, \quad (5)$$

where $x = k_z/k$ and the z is [111] axis. We are interested here in the frequency-dependent susceptibility of spin ice, and thus we need to extend the renormalization group analysis developed in Ref. [12] for the critical dynamics. Due to absence of any locally conserved quantities in the problem, the system can be described by purely relaxational dynamics governed by the free energy (4):

$$\partial_t m = -\Gamma \frac{\delta \mathcal{F}}{\delta m}. \quad (6)$$

To obtain estimates for coefficients in (4) we use virial expansion as described below; the estimate for the kinetic coefficient Γ will be provided at the end of this Letter. At low temperatures $T \ll T_c$, in the high-H phase almost all spins are aligned with the magnetic field, while in the low-H phase the same is valid for the spins of triangular sublattices only; configurations of other spins is governed by the kagome ice rules, see Fig.2

and Ref.[1]. One possible way to estimate the parameters of $\mathcal{F}$ near the critical point would be to consider gas of interacting monopoles taking into account their direct as well as entropic interactions [13]; similar problem of lattice ion systems was considered in [14]. However, we prefer to start from the high-H phase and to consider an exponentially dilute (far from the critical point) gas of flipped spins as a lattice gas of interacting particles. Such an approach will be approved *a posteriori* by the numerically small concentration of flipped spins even at the critical point. Using the Hamiltonian (3) we obtain the energy of such a particle (siting in the site 0) in the form

$$E_0 = \frac{2}{3}\mu H + 2\sum_i J_{0i}(\mathbf{e}_0\cdot\mathbf{e}_i)S_i - \\ -2Da^3\sum_i\left[\frac{\mathbf{e}_0\cdot\mathbf{e}_i}{|\mathbf{r}_{0i}|^3} - \frac{3(\mathbf{e}_0\cdot\mathbf{r}_{0i})(\mathbf{e}_i\cdot\mathbf{r}_{0i})}{|\mathbf{r}_{0i}|^5}\right]S_i, \qquad (7)$$

where $S_i = 1$ on triangular sublattices and $S_i = -1$ on kagome sublattices. The interaction of two particles siting in the sites i and j is equal to

$$U_{ij} = \frac{4}{3}J_{ij} + 4Da^3\left[\frac{\mathbf{e}_i\cdot\mathbf{e}_j}{|\mathbf{r}_{ij}|^3} - \frac{3(\mathbf{e}_i\cdot\mathbf{r}_{ij})(\mathbf{e}_j\cdot\mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5}\right]. \qquad (8)$$

Now we can employ virial expantion [15] in terms of small density of particles n, to obtain free energy

$$F(n) = TN_0[-1 - n - b_2n^2 - \\ - (b_3 - 2b_2^2)n^3 + \frac{E_0}{T}n + n\ln n], \qquad (9)$$

where T-dependent functions

$$b_2 = \frac{1}{2!}\sum_i f_{0i}, \quad \text{with} \quad f_{ij} = \exp\left[-\frac{U_{ij}}{T}\right] - 1,$$

$$b_3 = \frac{1}{3!}\sum_i\sum_j[f_{0i}f_{ij}f_{j0} + f_{0i}f_{0j} + f_{i0}f_{ij} + f_{0i}f_{0j}],$$

are "cluster integrals" given by the summation over the lattice. We calculate these sums numerically and find the position of the critical point $H_c^{th} = 1.37\,\mathrm{T}$, $T_c^{th} = 0.85\,\mathrm{K}$ and critical concentration of "particles" $n_c = 0.14$ from the conditions

$$\frac{dF}{dn} = 0, \quad \frac{d^2F}{dn^2} = 0, \quad \frac{d^3F}{dn^3} = 0. \qquad (10)$$

Smallness of n_c supports qualitative validity of our virial expansion; however, experimental values of H_c and T_c are below our estimates by factors 1.5-2. With the position of the critical point being determined, we can estimate values of the parameters entering free energy functional $\mathcal{F}$:

$$\alpha = \frac{da}{dT} = 50\,\mathrm{K}^{-1},\ \lambda = 1300\,\text{Å}^3\cdot\mathrm{K}^{-1},\ \gamma = 0.2\,\mathrm{T}\cdot\mathrm{K}^{-1} \qquad (11)$$

However, virial expansion is not useful to determine the coefficient b entering the gradient term in (4); thus we estimate it within nearest-neighbours approximation, neglecting long-distance tail of the dipole-dipole interaction:

$$b \approx \frac{5\sqrt{2}}{24}\frac{J_1 + 5D}{D}a^2 \approx 26\,\text{Å}^2. \qquad (12)$$

Coming back to the problem of longitudinal susceptibility and considering it in the mean-field approximation, one obtains (1) with $\chi_0^{-1} = \alpha(T - T_c)$, so exactly in the critical point $\chi \propto i\Gamma/\omega$. We will see below that account of fluctuations do not change this result considerably, leading to logarithmic corrections only.

With the parameters of the free energy (4) specified, we move to the analysis of critical fluctuations and their role in the dynamics. Thus we construct, using Eqs.(4), (6), the Martin-Siggia-Rose [8–10] dynamic action

$$\mathcal{I} = \int dtdV[\Gamma^{-1}p\partial_t m + apm + p\hat{b}m + \\ + \tfrac{\lambda}{6}pm^3 - ph + iT\Gamma^{-1}p^2], \qquad (13)$$

where $p = p(t,\mathbf{r})$ is the dynamic field conjugated to the magnetization field $m = m(t,\mathbf{r})$, and the last term in the action describes thermal noise. The estimate for the kinetic coefficient Γ will be provided later. Summation of leading logarithmic corrections with the action (13) up to one-loop approximation can be done in complete analogy with the paper of Larkin and Khmelnitsky [12], as it refers to the static quantities only:

$$\frac{d\lambda}{d\xi} = -g\lambda, \quad \frac{da}{d\xi} = -\frac{1}{3}ga, \qquad (14)$$

$$g = \frac{3T}{16\pi\sqrt{4\pi b^3}}\lambda, \quad \xi = \frac{1}{2}\ln\frac{b\Lambda^2}{bk^2 + 4\pi x^2}, \qquad (15)$$

where $\Lambda \sim a^{-1}$ is an ultraviolet cutoff. Renormalization of the gradient term b and the kinetic coefficient Γ appears in the second-loop approximation only. The corresponding diagrams are represented in Fig.3. Calculation of these diagrams leads to the renormalization-group equations

$$-\frac{d\Gamma}{d\xi} = \frac{2C}{3^3\pi^6}g^2\Gamma,\ \frac{db}{d\xi} = \frac{2^2}{3^5}g^2b,\ C = \int_0^\infty\frac{dpds}{p^3s^2}f^3(s,p), \qquad (16)$$

$$f(s,p) = \int_{q>0}\frac{q^2dqd\xi d\varphi}{q^2+\xi^2}\exp[-\frac{q^2+\xi^2}{s} + i\frac{q}{p}\cos\varphi + iq\xi],$$

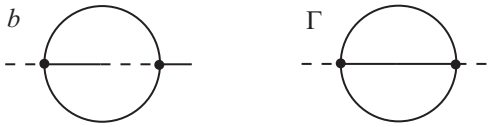

Fig.3. Diagrams responsible for the renormalization of coefficients b and Γ. Solid lines represent correlation function $\langle m_1 m_2 \rangle$, and mixed lines correspond to response function $\langle m_1 p_2 \rangle$

where the constant $C \approx 400$. The solution of equations (16) is as follows:

$$g = \frac{g_0}{1+g_0\xi}, \quad a = \frac{a_0}{(1+g_0\xi)^{\frac{1}{3}}},$$
$$b = b_0 \exp\left[\frac{4}{3^5}\frac{g_0^2}{1+g_0\xi}\right], \tag{17}$$
$$\Gamma = \Gamma_0 \exp\left[-\frac{2C}{3^3\pi^6}\frac{g_0^2}{1+g_0\xi}\right],$$
$$\xi = \frac{1}{2}\ln\frac{b\Lambda^2}{|a|+bk^2+4\pi x^2+|\omega|/\Gamma}.$$

Using Eqs.(11), (15) we find that initially value of coupling constant is $g_0 = 0.15 \ll 1$. Then renormalization of b and Γ is very small numerically and we neglect it, so the susceptibility takes the form

$$\chi = \frac{1}{a(k,x,\omega)+bk^2+4\pi x^2 - i\omega/\Gamma} \tag{18}$$

and in the uniform field

$$\chi = \frac{1}{a - i\omega/\Gamma}, \quad a(T) = \frac{\alpha(T-T_c)}{(1+\frac{1}{2}g_0\ln\frac{b\Lambda^2}{\alpha|T-T_c|+|\omega|/\Gamma})^{\frac{1}{3}}}. \tag{19}$$

For $T < T_c$ static susceptibility is modified by a factor of two: α should be replaced by $\alpha/2$. This is the result in the absence of effective field h which is defined after Eq.(4). If the temperature $T = T_c$, then the renormalization cutoff will be determined by h, i.e. by the deviation $H - H_c$

$$\chi = \frac{1}{a^* - i\omega/\Gamma}, \quad a^*(h) = \frac{\left(\frac{9}{2}\lambda_0 h^2\right)^{\frac{1}{3}}}{(1+\frac{1}{2}g_0\ln\frac{b\Lambda^2}{(\lambda_0 h^2)^{\frac{1}{3}}+|\omega|/\Gamma})^{\frac{1}{3}}}. \tag{20}$$

Exactly at the critical point $a = 0$ and $\chi \sim i\omega^{-1}$. However, in this system there is no conductivity in the usual sense. The point is that our lattice consists of a stack of interchanging kagome and triangular layers. In moderate magnetic fields (in particular, in the vicinity of the critical point), orientation of spins on the triangular layers are almost fixed. This results in the situation when monopoles are bound to theirs kagome layers and thus there is no direct motion of monopoles in the direction of the field. In fact, monopoles of one kagome layer reside in centers of tetrahedra, i. e. on two sublattices that are separated by a finite distance $h = a_d/3$ in the direction $(1,1,1)$ of the applied field. Thus positive-charge and negative-charge monopoles will be siting, preferably, on the "upper" and "lower" sublattices correspondingly, with the energy difference $\mu H/3$ between them. As a result, magnetization is primarily determined by the concentration of the monopoles.

Finally, we need to estimate relaxation rate Γ. Dynamical processes in spin ice at low temperatures and in low-field phase are governed almost solely by monopoles. Ground state doublet of each spin is separated by large energy gap $\Delta \sim 300\,\mathrm{K}$ from higher-energy states [1]. Thus, at temperatures $T \leq 1\,\mathrm{K}$, processes of spins flips are solely quantum. Then, the dependence of magnetoconductivity on temperature follows Arrhenius law with activation energy $E_m \approx 1\,\mathrm{K}$ corresponding to the creation of one monopole. Reminding the relation between Γ and σ, we find

$$\Gamma(T, H=0) = \Gamma_0 e^{-E_m/T}, \tag{21}$$

where Γ_0 is temperature-independent. Low-field dynamical magnetic susceptibility of $Dy_2Ti_2O_7$ was measured in Ref. [16]. Using the results [16] for the imaginary part of susceptibility together with Eq.(1), we obtain

$$\Gamma(T = 1.8\,\mathrm{K}, H = 0) \sim 100\,\mathrm{s}^{-1}.$$

Comparing it with Eq.(21) and the estimate for E_m. we conclude that $\Gamma_0 \sim 100\,\mathrm{s}^{-1}$ as well.

The nature of spin relaxation in spin ice near critical point is not quite clear. As we already mentioned above, in presence of magnetic field $\sim 1\,\mathrm{T}$, positive and negative magnetic charges reside on different sublattices and the tunneling processes become essentially modified, as the positions of monopoles on the kagome lattices are not equivalent. There are two possible scenarios for tunneling processes: sequential and simultaneous flips of two spins. Below we estimate the rate of sequential process, which will provide the lower bound for the relaxation rate near critical point. Energy difference for the monopoles of the same sign siting in two kagome sublattices leads to the exponential ratio of their concentration: $N_2 = N_1 \exp[-2\mu H/3T]$. On the other hand, detailed balance condition reads $N_1\Gamma_1 = N_2\Gamma_2$ where subscript "1" refers to monopoles situated on "native" sublattice, and subscript "2" to "foreign" ones. Assuming $\Gamma_2 \sim \Gamma_0$ since no additional barrier exists for the

hopping of monopole from a "foreign" site to a "native one", we come to the estimate

$$\Gamma^{\text{upper}}(H,T) \sim \Gamma_0(0,T)\exp[-2\mu H/3T] \qquad (22)$$

and eventually $\Gamma_c \sim (10^{-2} \div 100)\,\text{s}^{-1}$.

In conclusions, we calculated dynamic spin susceptibility of spin ice material $Dy_2Ti_2O_7$ near the critical point. Logarithmic corrections due to critical fluctuations are found to be small. The response of the form $\chi(\omega) \sim (i\omega)^{-1}$ is predicted in the broad range of low frequencies. Measurement of the prefactor in this dependence would allow to determine the nature of elementary spin flip processes near critical point.

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