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LETTER TO THE EDITOR

Ordering of microscopic Dzyaloshinskii–Moriya vectors and static properties of spin glasses

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Abstract. The mean-field theory of the Dzyaloshinskii–Moriya (DM) interaction in spin glasses which is based on the concept of the eigenmodes of the exchange integrals matrix is considered. There are additional degrees of freedom, sensitive to an external magnetic field, connected with ordering of microscopic DM vectors below the freezing temperature. They contribute to the magnetic susceptibility, and the macroscopical unidirectional anisotropy and modify the H–T phase diagram.

The Dzyaloshinskii–Moriya interaction [1,2] has been well studied in crystalline magnets. However, there are numerous experimental [3,4] and theoretical [5,6] papers (see also references in [7]) demonstrating the importance of this interaction in spin glasses (SG). This letter describes a new mean-field approach accounting for the Dzyaloshinskii–Moriya (DM) interaction and its influence on some static properties of SG.

Beginning from the first Anderson paper [8], a very fruitful approach for the description of the transition into the SG state was being developed by several authors [9–12]. This theory formulates the phase transition problem in terms of eigenmodes of the exchange integrals matrix J_{ik}. Such a representation is advantageous because it diagonalizes the main Heisenberg exchange interaction: $-\sum_{ik} J_{ik} \boldsymbol{S}_i \cdot \boldsymbol{S}_k \rightarrow -\sum_\alpha J_\alpha \boldsymbol{L}_\alpha^2$. Here $\boldsymbol{L}_\alpha = \sum_i u_{i\alpha} \boldsymbol{S}_i$ is the eigenmode corresponding to the eigenvalue J_α of the matrix J_{ik}, the spatial distribution of the wave function $u_{i\alpha}$ defines the character (localized or extended) of the eigenmode α, and $\boldsymbol{S}_i$ is the spin (classical vector) at site i. In the calculations of a susceptibility in [9, 12] the eigenmodes α are treated as magnetization vectors. However, due to the randomness of signs of the exchange matrix components J_{ik}, the signs of the values of $u_{i\alpha}$ are also random. Therefore, the mode $\boldsymbol{L}_\alpha$ is the vector of antiferromagnetism rather than the magnetization vector.

In this theory the very complicated non-linear problem of the intermode interactions has arisen—these may substantially change the initial spectrum of the non-interacting modes. We would like to mention here two different pictures of the phase transition into the SG state which are considered to be clearer upon the application of this approach.

Hertz *et al* [10] have shown, using the Hartree–Fock approximation, that for decreasing temperature T the intermode interactions suppress the condensation of the localized modes. Only one extended mode condenses when T reaches the mobility

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edge T_0 (see also [11] and section VI.F.2 in [7]), but marginal (logarithmically divergent) fluctuations can destroy such a phase transition. However, one cannot be sure that this result will not be changed beyond the scope of the Hartree–Fock approximation.

In this letter we shall examine the second picture of the cascade of the local phase transitions outlined firstly in [8] and described in detail by Hertz [9,12]. Here the suppression of the condensation is not so rigid and on reducing T the condensation starts from the localized modes with the largest eigenvalues J_α. The already condensed modes suppress the potentially soft modes and hence their renormalized critical temperatures of the condensation T_α are lower than J_α. This picture seems to be rather relevant to experiment (see [9,12]).

We consider the simplest pair DM interaction

$$w_{\mathrm{DM}} = (1/2)\sum_{ij} \boldsymbol{D}_{ij} \cdot [\boldsymbol{S}_i \times \boldsymbol{S}_j] \tag{1}$$

where $\boldsymbol{D}_{ij}$ is the DM vector (see microscopic calculations of $\boldsymbol{D}_{ij}$ in [2,5,13]). Here we assume that the vector $\boldsymbol{D}_{ij}$ is independent of spin variables.

In crystalline weak ferromagnets the directions of the microscopic vectors $\boldsymbol{D}_{ij}$ are dictated by the crystal symmetry. The macroscopic DM field forming the weak magnetic momentum is directed along some highly symmetrical crystalline axes. In contrast, in SG the orientation and length of the vector $\boldsymbol{D}_{ij}$ have random character.

There is a property which is principal for our consideration: *the microscopical DM vector is defined up to a sign*, i.e. $\boldsymbol{D}_{ij} = +\boldsymbol{D}^0_{ij}$ or $\boldsymbol{D}_{ij} = -\boldsymbol{D}^0_{ij}$, where $\boldsymbol{D}^0_{ij}$ is some fixed vector (see below). This property is well known in crystalline weak ferromagnets. For example, the sign of the DM field in the weak ferromagnet with two sublattices depends on the choice of the directions of ferro- and antiferromagnetic vectors [14].

With the basis of the eigenmodes $\boldsymbol{L}_\alpha$ the formula (1) has the form

$$w_{\mathrm{DM}} = (1/2)\sum_{\alpha,\beta} \boldsymbol{D}_{\alpha\beta} \cdot [\boldsymbol{L}_\alpha \times \boldsymbol{L}_\beta]\,. \tag{2}$$

Here $\boldsymbol{D}_{\alpha\beta} = \sum_{ij} \boldsymbol{D}_{ij} u_{i\alpha} u_{j\beta}$. Let us make the following assumption which leads to essential simplification but does not restrict the generality of the consideration: the magnetization vector $\boldsymbol{M} \equiv (1/\sqrt{N})\sum_i \boldsymbol{S}_i$ is one of the extended eigenmodes, i.e. $\boldsymbol{M} \equiv \boldsymbol{L}_\mu$ and $u_{i\mu} = 1/\sqrt{N}$, where N is the total number of spins. This is possible if the sum $\tilde{J} = \sum_i J_{ij}$ is independent of the index j. Such a condition is quite reasonable for the homogeneous SG. Then in (2) we may select the terms containing the magnetization vector $\boldsymbol{M}$

$$w'_{\mathrm{DM}} = -\boldsymbol{M} \cdot \sum_\alpha [\boldsymbol{L}_\alpha \times \boldsymbol{D}_\alpha] \tag{3}$$

where $\boldsymbol{D}_\alpha = (1/\sqrt{N})\sum_{ij} \boldsymbol{D}_{ij} u_{i\alpha}$. The vector $\boldsymbol{H}_{\mathrm{DM}} = \sum_\alpha [\boldsymbol{L}_\alpha \times \boldsymbol{D}_\alpha]$ has the meaning of the macroscopic DM field or the field of the unidirectional magnetic anisotropy [5–7]. According to our model, the DM field $\boldsymbol{H}_{\mathrm{DM}}$ arises as a result of a summation of the microscopic random vectors $\boldsymbol{D}_{ij}$ ordered below the freezing temperature.

Let us consider the isotropic SG and fix the directions of the already condensed modes $\boldsymbol{L}_\alpha$ below T_f. We can always take such directions of the vectors $\boldsymbol{D}_{ij}$ when the

value $H^z_{DM} = \boldsymbol{H}_{DM} \cdot \boldsymbol{z}$ reaches a maximum; here $\boldsymbol{z}$ is a unit vector along the external field $\boldsymbol{H} = H\boldsymbol{z}$. We shall label these vectors with the index '0': $\boldsymbol{D}_{ij} \equiv \boldsymbol{D}^0_{ij}$. There is thermal orientational disorder at finite temperature T, and we have to substitute in (3) the average vector $\langle \boldsymbol{D}_{ij} \rangle = p_{ij}\boldsymbol{D}^0_{ij} - (1-p_{ij})\boldsymbol{D}^0_{ij}$ instead of $\boldsymbol{D}_{ij}$. Here p_{ij} is the probability that the vector $\boldsymbol{D}_{ij} = +\boldsymbol{D}^0_{ij}$ and $(1-p_{ij})$ is the probability that $\boldsymbol{D}_{ij} = -\boldsymbol{D}^0_{ij}$. We do not assume here a special interaction between the different vectors $\boldsymbol{D}_{ij}$. Due to this the free energy, connected with the DM interaction (3), may be written in the general form $f_{DM}(\boldsymbol{L}_\alpha, \sigma_{ij}, \boldsymbol{M}, T) = w'_{DM} - TS$, where S is the configurational entropy which is calculated in the standard way:

$$S = -(1/2)\sum_{ij}\left[p_{ij}\ln p_{ij} + (1-p_{ij})\ln(1-p_{ij})\right].$$

Introducing the new variables $\sigma_{ij} = 1 - 2p_{ij}$ we obtain

$$f_{DM} = -(1/\sqrt{N})\boldsymbol{M}\cdot\sum_{ij}\sigma_{ij}\sum_{\alpha}u_{i\alpha}\left[\boldsymbol{L}_\alpha \times \boldsymbol{D}^0_{ij}\right] + (1/4)T\sum_{ij}\left\{\ln\left[(1-\sigma^2_{ij})\right] + \sigma_{ij}\ln\left[(1+\sigma_{ij})/(1-\sigma_{ij})\right]\right\}. \tag{4}$$

The variables σ_{ij} characterize degrees of freedom related to the ordering of the microscopic DM vectors. We would like to emphasize that these variables can be governed by the external magnetic field because of the direct interaction with the magnetization.

In the final formula for the total free energy of the SG in the magnetic field we write terms containing the magnetization $\boldsymbol{M}$ in the explicit form

$$f = f_0(\boldsymbol{L}_\alpha, T) + (M^2/2\chi) + (1/2)\sum_{\alpha}\left(\zeta^{(1)}_\alpha M^2 L^2_\alpha + \zeta^{(2)}_\alpha(\boldsymbol{M}\cdot\boldsymbol{L}_\alpha)^2\right) + f_{DM}(\boldsymbol{L}_\alpha, \sigma_{ij}, \boldsymbol{M}, T) - \boldsymbol{M}\cdot\boldsymbol{H}. \tag{5}$$

Here χ is a susceptibility, $\chi^{-1} \propto (T + \bar{J})$ ($\bar{J}$ has the sense of a paramagnetic Curie temperature), and $\zeta^{(1)}_\alpha$ and $\zeta^{(2)}_\alpha$ are positive constants determining the intermode interactions: $\zeta^{(1)}_\alpha, \zeta^{(2)}_\alpha \propto \sum_i u^2_{i\alpha}$. Due to the randomness of signs of $u_{i\alpha}$ and small overlaps of localized modes we have neglected in (5) the terms proportional to the factors $\sum_i u_{i\alpha}$, $\sum_i u_{i\alpha}u_{i\beta}$, $\sum_i u_{i\alpha}u_{i\beta}u_{i\gamma}$ ($\alpha \neq \beta \neq \gamma$) etc (for details see [12]). Such an expansion (in $\boldsymbol{M}$ and $\boldsymbol{L}_\alpha$) is similar to the expansion of the free energy in the vectors of ferro- and antiferromagnetism for usual antiferromagnets [14,15].

The first term f_0 includes: (i) the largest exchange interaction, (ii) the DM interaction between modes $\boldsymbol{L}_\alpha$ which was excluded from (3) (it is much less than the exchange interaction), and (iii) intermode interactions (see [10,12]). The above-mentioned problem of the phase transition theory is connected with this term which describes the condensation of eigenmodes. However, we may avoid this problem and there is no need to go into the details of this term or to examine it thoroughly, since we are interested only in that part of the DM interaction which is sensitive to $\boldsymbol{H}$.

Let us analyse the process of magnetization. For zero-field cooling (ZFC) the equilibrium values $\sigma^0_{ij} = 0$. This follows from the equation $\partial f/\partial\sigma_{ij} = 0$ at $M = 0$ and means that below T_f the vectors $\boldsymbol{D}_{ij}$ are frozen chaotically and the resulting macroscopic DM field is absent.

There is an important question of the relaxation of the DM field which we omit here since it needs special consideration. In this letter we calculate the ZFC linear (initial) susceptibility χ_{ZFC} under the assumption that the values σ_{ij} are really frozen on switching on the magnetic field and that there is no relaxation of the DM field. From the equality $\partial f/\partial M = 0$ at $\sigma_{ij}^0 = 0$ it is easy to obtain

$$\chi_{ZFC}^{-1} = \chi^{-1} + \sum_\alpha \zeta_\alpha (L_\alpha^0)^2. \tag{6}$$

Here $\zeta_\alpha = \zeta_\alpha^{(1)} + \zeta_\alpha^{(2)} (l_\alpha^0 \cdot z)^2$, $l_\alpha^0 = L_\alpha^0/|L_\alpha^0|$, and L_α^0 are the equilibrium eigenmodes obtained from the equations $\partial f_0/\partial L_\alpha = 0$ (in a calculation of linear susceptibility the terms of second order in H may be neglected).

For slow quasistatic field cooling (FC) magnetization, the variables σ_{ij} reach their equilibrium values

$$\sigma_{ij}^0 = \tanh\left(2M \cdot \sum_\alpha \sum_{ij} u_{i\alpha} [L_\alpha \times D_{ij}]/\sqrt{N}T\right). \tag{7}$$

In the weak field $\tanh x \approx x$, and thus $\sigma_{ij}^0 \propto H$. Using (5) with the equilibrium value σ_{ij}^0 we may find the FC susceptibility

$$\chi_{FC}^{-1} = \chi_{ZFC}^{-1} - (2/NT) \sum_{\alpha,\beta} \sum_{ij} (u_{i\alpha} [L_\alpha^0 \times D_{ij}^0]_z)^2. \tag{8}$$

There are some consequences of equations (6) and (8). The deviation from the Curie law '$\chi^{-1} \propto T$+constant' and the difference between χ_{ZFC} and χ_{FC} on lowering T start at the temperature at which the first eigenmodes are condensed. This deviation increases due to the growth of the already condensed modes and the condensation of the new modes. This picture gives a natural explanation of the slightly rounded cusp in the temperature dependence of χ, which appears in some region of T around the mobility edge T_0 [9], where the true long-range order exists. The formula (8) shows that χ_{ZFC} is always less than χ_{FC}. This inequality clearly displays the fact that the state of SG essentially depends on the thermodynamic path of the SG sample ('magnetic history'). It can be seen that there are no thermodynamic equilibria in the ZFC state on switching on the field, unlike what is found in the FC state. This verifies the suggestion that under field cooling the SG reaches the true equilibrium state (see [7]).

It is reasonable to assume that there are no correlations between the directions of the different eigenmodes as well as between the directions of L_α and D_{ij}. The averaging over orientations gives

$$\delta(\chi^{-1}) \equiv \chi_{ZFC}^{-1} - \chi_{FC}^{-1} = \sum_\alpha \nu_\alpha (L_\alpha^0)^2 \tag{9}$$

where $\nu_\alpha = (4/9NT) \sum_{ij} (D_{ij}^0 u_{i\alpha})^2$. The formulae (6) and (8) may also be used when the picture of the phase transition described in [10] applies, i.e. when at $T = T_f = T_0$ only one extended mode L_e condenses. In rough estimates we may drop summation over α and replace L_α by L_e.

After the FC process there arises a macroscopic field of unidirectional anisotropy (the DM field) directed along z

$$H^z_{DM} = (1/\sqrt{N}) \sum_{ij} u_{i\alpha} [L_\alpha \times D^0_{ij}]_z \sigma^0_{ij}. \tag{10}$$

At low T and for strong H the variables $\sigma^0_{ij} \to 1$ and hence the field H^z_{DM} reaches a maximum (we do not make more detailed numerical estimates of H^z_{DM} here; they will be presented elsewhere).

There is an important question of how the DM interaction could modify the H–T phase diagram of the SG. For a weak field H that part of the free energy that depends on L_α can be expressed in the following form

$$f(L_\alpha, H, T) = f_0(L_\alpha, T) + (1/2)(\chi_{ZFC})^2 \sum_\alpha (\zeta_\alpha - \nu_\alpha) L^2_\alpha H^2 \tag{11}$$

where we replaced M by its equilibrium value $\chi_{FC}H$. To analyse of the H–T diagram we must detail the term f_0 in (11). Being aware of all the difficulties described above, it is nevertheless reasonable to examine the simplest mean-field approach. The first part in the usual Landau-type expansion of the f_0 near T_f [8,10], quadratic in L_α, is: $a \sum_\alpha (T - T_\alpha) L^2_\alpha$, where the constant $a > 0$. Then from (11) one can see that the shift of the effective temperature of the αth mode condensation in the magnetic field: $T_\alpha - T_\alpha(H)$ is equal to $(\chi^2_{FC}/2a)(\zeta_\alpha - \nu_\alpha)H^2$. This means that the line of the SG transition on the H–T phase diagram at small values of the field follows the law $T_f - T_f(H) \propto KH^2$, as well as the Gabay–Toulouse line [16]. In our approach the nature of the T_f shift and of the phase transition line is the same as the nature of the Néel temperature shift and of the corresponding line in antiferromagnets [14]. Usually, $T_f(H)$ falls with increasing H. This corresponds to $K > 0$ in our equations. But when the DM interaction is not small and $\zeta_\alpha < \nu_\alpha$, the constant K may be negative. This leads to the anomalous behaviour of the phase transition line, i.e. $T_f(H)$ grows with increasing H [17].

In conclusion, some remarks are in order. It is to be noted that apart from the effects of non-ergodicity and irreversibility in SG, there exists a close similarity between the static behaviour of SG and antiferromagnets. For example, in both cases an interpretation of the cusp in the $\chi(T)$-dependence is the same. Like in SG, in antiferromagnets the magnetization is not a soft mode. Soft antiferromagnetic modes which arise below the Néel temperature and grow with a decrease of T are not thermodynamically conjugate to H, but they essentially change the response to the magnetic field and this causes the cusp [14]. The similarity has a deeper reason dictated by the proper nature of the SG state which needs a predominance of antiferromagnetic couplings over ferromagnetic ones. It looks as if the SG is the antiferromagnet with an infinite number of magnetic sublattices with different spatial distributions. Our approach was based on a strong assumption. We considered the microscopic DM vectors D_{ij} to be independent. But it is impossible to exclude some correlation between the neighbouring vectors. Anyway, only microscopic consideration (separately for metallic and non-metallic SG) should clarify this point.

If this model is valid, there is an attractive possibility of implementing the Hopfield model of associative memory [18] in the real SG. The appearance of the memory in our case will be connected not with an arrangement of the exchange integrals, but with ordering of the microscopic DM vectors.

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