Transitions involving conical magnetic phases in a model with bilinear and biquadratic interactions.

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I. INTRODUCTION

Localized magnetic moments in several compounds of the family RT_2X_2 (R = rare earth, T = transition metal, X = Ge, Si) are found to have "conical" ground state (GS) configurations (see eq.1) and to undergo transitions to configurations with higher symmetry (helical, canted, collinear) as temperature T increases \[1\]. Among the many works on this subject we can cite refs.2-6 (see also references therein and in ref.1). In this work we consider LaMn_2Ge_2 and LaMn_2Si_2 \[2\], \[3\], \[4\]. In such compounds only the Mn atoms bear a localized magnetic moment, so that we consider only the Mn sublattice. We take it as a base-centered tetragonal lattice, whose lattice constants are given, in terms of the crystallographic constants \(a\) and \(c\), by \(\alpha \equiv a\) and \(\cappa \equiv c/2\), and the lattice sites by \(\vec{R}_{hkl} \equiv \alpha_1\vec{e}_x + \kappa_1\vec{e}_y + \lambda_1\vec{e}_z\), where \(l\) is an integer and \(j, k\) are both integers or half-integers. In the following we will use subindices \(i\) and \(j\) as short notations for sets \((h, k, l)\). We work with the total atomic angular momentum (in units of \(\hbar\)), which we call spin and denote \(\vec{S}\), as in the spin Hamiltonian formalism. The corresponding magnetic moment is \(\vec{\mu} \equiv \mu_B\vec{S}\) \((\mu_B\) is Bohr’s magneton).

The magnetic structure can be characterized by a pair of polar angles \(\theta\) and \(\alpha\), such that at any site \(\vec{R}_{hkl} \equiv \alpha_1\vec{e}_x + \kappa_1\vec{e}_y + \lambda_1\vec{e}_z\) the average value of the local spin is given by

\[
\vec{S}_{hkl} = \vec{S}[(\sin(\theta)\cos(\alpha + \xi_{hkk})\vec{e}_x^* + \sin(\theta)\sin(\alpha + \xi_{hkk})\vec{e}_y^* + \cos(\theta)\vec{e}_z^*],
\]

where \(\xi_{hkk} = 0 \text{ or } \pi\) for \(h, k\) integers or half-integers respectively and the bars indicate the statistical averages (see also eqs. 22-24). We can also say that the polar angles \((\theta_i, \phi_i)\) characterizing the direction of the average spin at a site \(i \equiv (h, k, l)\) are given by \(\theta_i = \theta\) and \(\phi_i = (\alpha + \xi_{hkk})\). For general values of \(\theta\) and \(\alpha\) (i.e. values different from \(0, \frac{1}{2}\pi, \pi\) and \(\frac{3}{2}\pi\)) this gives what is customarily called a conical structure (even though perhaps the term "conical" tout court should be limited to the case \(\xi_{hkk} = 0 \forall h, k\)). Particular cases of the conical structure are the helical \((\theta = \frac{1}{2}\pi,\alpha = 0,\frac{1}{2}\pi,\pi,\text{or}\frac{3}{2}\pi)\) and the collinear (all the spins are parallel or anti-parallel to each other) structures. In the following we will call "general conical structure" a conical structure in which \(\theta\) and \(\alpha\) can take any values, including these particular values.

As \(T\) increases the spins get progressively disordered (so that their average values decrease), while angles \(\theta\) and \(\alpha\) vary, until eventually taking values corresponding to some of the above-mentioned "particular cases" so that a more symmetrical structure is obtained. In a previous work \[7\] a model was proposed assuming that the localized magnetic
moments are subjected to bilinear and biquadratic interactions up to fifth neighbours (and possibly to an anisotropy field). The interaction constants were obtained from \textit{ab initio} calculations of the energies of different structures. For any given \( T \) the structure was obtained by finding, in a mean-field approximation (MFA), the \( \theta \) and \( \alpha \) angles, the modulus of the average spins, and the average of \( m_i^2 \) (\( m = \) total angular momentum quantum number) for which the free energy was minimal. This model could qualitatively account in a reasonable way for the mentioned behaviours, but it gave exaggeratedly high values for the transition temperatures \( T_c \)'s between the different configurations (see sections IV, V). In this paper we modify the MFA in such a way that reasonable values of the \( T_c \)'s are obtained.

II. THE MODEL

A common approach to a phenomenological study of a crystal with localized magnetic moments is the employ of the bilinear Heisenberg Hamiltonian:

\[
\hat{H}_J = -\frac{1}{2} \sum_{i,j} J_{ij} \hat{S}_i \cdot \hat{S}_j
\]  

(2)

(the prime in the summation means \( i \neq j \)).

The ground state for such a Hamiltonian is a general type of helical structure \([8]\), so that in order to obtain a conical structure some other interaction must be added to it. The addition of a biquadratic term to the Heisenberg bilinear interaction was shown to explain another type of phase transitions in a \( RT_2X_2 \) compound, namely \( U\text{ Ni}_2\text{Si}_2 \) \([9]\). In ref.7 it was found that a Hamiltonian with bilinear and biquadratic interactions both up to the fifth neighbouring sites gives an acceptable fitting of \textit{ab initio} total energy values for several configurations in which the moduli of the spins are all equal to \( S \). The anisotropy field was not required, but we included the quadratic term in order to study its possible effects (a \( \cos^2(\theta) \) term does anyway appear in the bilinear interaction).

Accordingly, we assume the Hamiltonian

\[
\hat{H} = \hat{H}_J + \hat{H}_B + \hat{H}_D
\]  

(3)

with

\[
\hat{H}_B \equiv -\frac{1}{2} \sum_{i,j} B_{ij} [\hat{S}_i \cdot \hat{S}_j]^2
\]  

(4)

and

\[
\hat{H}_D \equiv D_2 \sum_i \hat{S}_{i,z}^2
\]  

(5)

where the \( J_{ij} \) and \( B_{ij} \) are non-zero for the five nearest sets of neighbours (which, for the reference site \((0,0,0)\), are: \((\pm \frac{1}{2}, \pm \frac{1}{2}, 0)\); \((\pm 1, 0, 0)\); \((0, \pm 1, 0)\); \((0, 0, \pm 1)\); \((\pm 1, \pm 1, 0)\); \((\pm \frac{1}{2}, \pm \frac{1}{2}, \pm 1)\) ).

Let us denote \(|m_i\rangle > \) the eigenstate of a spin at site \( i \) with eigenvalue \( m_i \) in the direction given by the polar angles \((\theta_i, \phi_i)\).

The interaction energy between the spins at sites \( i \) and \( j \) (i.e. \(<m_i| m_j| (\hat{H}_J + \hat{H}_B) |m_i\rangle > |m_j\rangle > \) is

\[
U_{J,B;ij}(m_i, m_j) \equiv U_{J;ij}(m_i, m_j) + U_{B;ij}(m_i, m_j),
\]  

(6)

with

\[
U_{J;ij}(m_i, m_j) = \frac{1}{2} J_{ij} m_i m_j p_{ij}
\]  

(7)

and

\[
U_{B;ij}(m_i, m_j) = -\frac{1}{2} B_{ij} \left[ \frac{1}{4}[S(S+1) - 3m_i^2][S(S+1) - 3m_j^2](p_{ij}^2 + 1) + S(S+1)(m_i^2 + m_j^2) - 3m_i^2 m_j^2 - \frac{1}{2} m_i m_j p_{ij} \right]
\]  

(8)
where $p_{ij}$ is the cosine of the angle between the spins at sites $i$ and $j$. The matrix element of $\hat{H}_B$ was calculated by rotating the spin operators $[10]$.

In a general conical structure all sites are energetically equivalent to each other, so that neglecting boundary effects the total energy is equal to $N U_s$, where $N$ is the total number of sites and $U_s$ is the energy per site. As reference site we take $(h = k = l = 0)$, also denoted $i = 0$. We have

$$U_s = \sum_{j=1}^{N-1} U_{ij}(m_0, m_j) + \frac{1}{2} D_2 \left[ (S(S + 1) - m_j^2) + [3m_j^2 - S(S + 1)] \cos^2(\theta) \right].$$

We shall denote respectively $J_{ij}$ and $B_{ij}$ the constants $J_{ij}$ and $B_{ij}$ for sites $i, j$ that are $\nu^{th}$ neighbours of each other.

In ref.7 eq.(9) with $m_0 = m_j = 0$ was used to determine the parameters $(J_{ij}, B_{ij}, D_2)$ of the Hamiltonian. The energy dependence on $\theta$ and $\alpha$ was written as

$$U_T(\theta, \alpha) = X_1 + X_2 \cos^2 \theta + X_3 \sin^2 \theta \cos \alpha + X_4 \cos^4 \theta + X_5 \cos^2 \theta \sin^2 \theta \cos \alpha + X_6 \cos^2 \theta - \sin^2 \theta \cos \alpha \right)^2.$$

where the $X_n$'s are constants.

In principle the $X_n$'s can be obtained by fitting the results of $ab$ initio total energy calculations for several configurations with $m_0 = m_j = S$. Such energies were obtained by using the FLEUR code, as described in ref.7. If this fitting is done for six configurations, i.e. for six pairs $(\theta, \alpha)$, a linear system is obtained, which can be solved for the $X_n$. We did this for several sets of six configurations, so as to check the consistency of the formalism (i.e. that in each case the same set of $X_n$ is obtained with a reasonable accuracy). We found that this consistency is achieved if these configurations are neither too close to each other nor too far from the GS. In the former case the determinants appearing in the numerical solution of the system are nearly zero (they are sums of terms which nearly cancel out each other); in the latter case it is possible that if one forces the electrons associated with the magnetism to occupy high-energy states, one cannot neglect the additional energy due to changes of the other electronic states. In both cases the error is large.

The $X_n$'s to be adopted in this work for LaMn$_2$Ge$_2$ and LaMn$_2$Si$_2$ are the averages of the $X_n$'s of the selfconsistent sets.

Having obtained the $X_n$'s, we must face the problem of determining the eleven parameters appearing in the Hamiltonian.

The part of the magnetic energy not depending on $\theta$ and $\alpha$ (i.e. $X_1$) is not the same as the analogous part of the total energy (as a matter of fact, it is several orders of magnitude smaller), so that it cannot be singled out in the $ab$ initio values. Therefore, $X_1$ is of no use for the determination of the parameters. As a consequence, $J_2, J_4, B_2$ and $B_4$ cannot be determined because they multiply terms with $p_{02}$ or $p_{04}$, which in a conical structure are constant, being equal to 1. In order to express the other seven parameters in terms of the five constants $X_n$ ($n=2, \ldots, 6$) we compare eq.(10) with the energy per site (eq.9 with (6-8)) for $m_i = m_j = S \forall i, j$. In doing so we must take into account that any site has four first, second and third neighbours, two fourth neighbours and eight fifth neighbours. We obtain the system

$$X_2 = -\frac{1}{2} S^2 [4(2J_1 - B_1) + (2J_3 - B_3) + 4(2J_5 - B_5)] + 8S^2(S - \frac{1}{2})^2 B_1 + S(S - \frac{1}{2}) D_2$$

$$X_3 = -\frac{1}{2} S^2 (2J_3 - B_3 - 8J_5 + 4B_5)$$

$$X_4 = -8S^2(S - \frac{1}{2})^2 B_1$$

$$X_5 = -S^2(S - \frac{1}{2})^2 B_3$$

$$X_6 = -4S^2(S - \frac{1}{2})^2 B_5$$

From here we obtain directly $B_1, B_3$ and $B_5$, after which we are left with two equations for the unknowns $J_1, J_3, J_5$ and $D_2$, two of which must be chosen arbitrarily (as well as $J_2, J_4, B_2$ and $B_4$). Concerning $D_2$, we shall give it a small value, while for the other three unknowns we think that the less arbitrary procedure is that of choosing $J_5$ in such a way that it and the values of $J_1$ and $J_3$ calculated in terms of it and of $D_2$ have a smooth variation with the interatomic distance, and are, in principle, about an order of magnitude greater than the corresponding $B_j$. An analogous criterion shall be followed for $J_2, J_4, B_2$ and $B_4$. 


Setting $\frac{1}{2}S^2(S - \frac{1}{2})^2 \equiv C_2$ we have

$$B_1 = -\frac{1}{16}X_4/C_2$$  (12)
$$B_3 = \frac{1}{2}X_5/C_2$$  (13)
$$B_5 = \frac{1}{8}X_6/C_2$$  (14)
$$J_3 = -\frac{1}{2}X_3/S^2 + \frac{1}{2}B_3 + J_5 - 2B_5$$  (15)
$$J_1 = \frac{1}{4}X_2/S^2 + [8(S - \frac{1}{2})^2 + 2B_1 - J_3 + \frac{1}{2}B_3 - 4J_5 + 2B_5 + [(S - \frac{1}{2})]/S]D_2$$  (16)

### III. CALCULATION OF THE FREE ENERGY

In this work we must find the magnetic structure of the LaMn$_2$X$_2$ compounds (which we know from experiment to be a general conical structure) at any temperature $T$. Therefore we must determine which is for the different sets of values of the interaction constants the conical structure that minimizes the free energy. In a general state of the system the spin at each site $i$ is in a state $|m_i>$, so the system is characterized by the $3N$ quantities $m_i$, $\theta_i$, $\phi_i$ ($i = 1, \ldots, N$).

For $T > 0$ the state of the system will be that superposition of the above-defined states which minimizes the free energy $F = -k_B T \log Z$, i.e. which maximizes the partition function. The latter is given by

$$Z = \sum_{m_0, \ldots, m_{N-1} = -S}^{S} \int d\Omega_0 \ldots \int d\Omega_{N-1} \exp[-\beta U(m_i, \theta_i, \phi_i)]$$  (17)

where $\beta \equiv 1/kT$, $d\Omega_i \equiv \sin \theta_i d\theta_i d\phi_i$ and each integration is carried out over all the space directions.

Taking advantage of the fact that all the sites are physically equivalent to each other we define the partition function per site $Z_s$ through $Z \equiv Z_s^N$.

We calculate $Z_s$ for the reference site $i = 0$. To do this we must make a MFA, i.e. we must approximate the energies of the states at that site in the field of the spins at the other sites (which will be referred to as ”source sites”) by replacing each of the latter by some average state. This is done as follows. The $m_j$, $\theta_j$, and $\alpha_j$ at each source site $j$ are taken according to eq.(1), where $\bar{S}$, $\theta$ and $\alpha$ are unknowns. Furthermore, as the contribution of a $j$ site to the energy of the reference spin depends also (due to the biquadratic interaction) on $m_j^2$ we must assign to each source site a fourth variable, $m_j^2$. We have

$$Z_{s,MFA}(\theta, \alpha, \bar{S}, \bar{m}^2) = \sum_{m_0 = -S}^{S} \int d\Omega_0 \exp[-\beta U(m_0, \theta_0, \phi_0; \theta, \alpha, \bar{S}, \bar{m}^2)].$$  (18)

The energy in the integrand contains the contributions of all the source sites, and consists of an interaction term and an anisotropy term (eqs.7,8,9). The latter, being a local term, requires no approximation.

For the interaction energy associated with the reference site, whose exact value is

$$\sum_{j=1}^{N-1} U_{JB,0j}(m_0, m_j; p_{0j})$$  (19)

(where $U_{JB,0j}(m_0, m_j; p_{0j})$ is given by eq.(6) with (7,8) taking into account that the spin at the $i = 0$ site has the general orientation $(\theta_0, \phi_0)$), there are several possible MFA expressions, and it is not clear which one is the best in any particular case (see section VI).

Now we must find, for any given $T$, the values of $\theta$, $\alpha$, $\bar{S}$, and $\bar{m}^2$ characterizing the state of minimal free energy, i.e. of maximal $Z$. For a conical structure the extremum conditions are
\[ \frac{\partial Z}{\partial \theta} = 0 \]
\[ \frac{\partial Z}{\partial \alpha} = 0. \]

On the other hand, both \( \bar{S} \) and \( \vec{m} \) must satisfy a self-consistence condition, i.e. they must be equal to the respective average values they give rise to at the reference site. \( \bar{S} \) is the modulus of

\[ \bar{S} = Z^{-1}\sum_{m_0=-S}^S m_0 \int d\Omega_0 [\sin \theta_0 \cos \phi_0 \vec{e}_x + \sin \theta_0 \sin \phi_0 \vec{e}_y + \cos \theta_0 \vec{e}_z] \exp[-\beta U(m_0, \theta_0, \phi_0; \theta, \alpha, \bar{S}, \vec{m}^2)], \]

i.e. is given by

\[ \bar{S} = \sqrt{\bar{s}_x^2 + \bar{s}_y^2} \]

(due to the symmetry of the structure, the \( y \) component vanishes), while

\[ \vec{m}^2 = Z^{-1}\sum_{m_0=-S}^S \int d\Omega_0 m_0^2 \exp[-\beta U(m_0, \theta_0, \phi_0; \theta, \alpha, \bar{S}, \vec{m}^2)]. \]

So, we must solve the system of equations (20), (21), (23) and (24)

For the more symmetrical structures the number of equations is reduced: for the helical structure we set \( \theta = \frac{1}{2}\pi \) and drop eq.(16a); similarly, for the canted structure, we set \( \alpha = \pi \), and instead of (16a) we drop (16b). Finally, in the collinear structure we have \( \theta = \frac{1}{2}\pi \), \( \alpha = \pi \), and the equation system is reduced to (14) and (15).

IV. NUMERICAL CALCULATIONS. LAM\(_{2}\)GE\(_{2}\)

The interaction constants \( J_{ij}, B_{ij} \) and \( D_2 \) are obtained from the \( X_n \)'s used in ref.7, taking into account that the latter were evaluated for a cell with four sites, i.e. must be divided by 4. According to the \textit{ab initio} calculations of ref.7 and to the experimental results of ref.2 (where the helical and FM components of the Mn magnetic moment are found to be approximately 2.6\( \mu_B \) and 1.6\( \mu_B \) respectively), and assuming \( L = 0 \) because of quenching, we take \( S = \frac{1}{2} \).

As we said above, only \( B_1, B_3 \) and \( B_5 \) can be derived from the \textit{ab initio} energies. It turns out that these \( B_n \)'s are negative, and their absolute values decrease smoothly for increasing interatomic distance.

For the other constants we proceed as explained in section 2: we choose first \( J_3 \) and \( D_2 \), thereby determining \( J_1 \) and \( J_3 \), and then, without affecting the other constants, we choose \( J_2, J_4, B_2 \) and \( B_4 \). We can get a set of \( J_n \)'s about an order of magnitude greater than the \( B_n \)'s only if \( J_1 \) and \( J_3 \) have different signs. This is a quite reasonable condition, being a property of the RKKY interaction

Among the sets of parameters that we used in our calculations, one of those yielding the closer agreement with the experimental results is: \( J_1 = -0.002137, J_2 = -0.0005, J_3 = 0.002792, J_4 = 0.0010, J_5 = 0.000755, B_1 = -0.000482, B_2 = -0.00023, B_3 = -0.000155, B_4 = -0.00009, B_5 = -0.000053, D_2 = 0.0005 \).

With these parameters we have at low-temperatures (LT) a conical structure with \( \theta \sim 60^\circ \) and \( \alpha \sim 114^\circ \).

The experimental values are \( \theta \sim 58^\circ \) and \( \alpha \sim 133^\circ \) (see ref.2, taking into account that therein \( \alpha \) refers to the rotation of the spin at site (\( \frac{1}{2}, \frac{1}{2}, 1 \)) (instead of (0,0,1), as in this work) with respect to that at (0,0,0), and that their lattice constant in the \( z \) direction is \( c \) while ours is \( \frac{1}{2}c \).

As \( T \) increases, both \( \theta \) and \( \alpha \) increase, until the helical structure is attained (with \( \alpha \sim 144^\circ \)). This happens at \( T \sim 439 \) K. Meanwhile, \( \bar{S} \) and \( \vec{m}^2 \) decrease from their LT values 1.5 and 2.25. Upon a further increase of \( T \) (keeping \( \theta = 90^\circ \)) \( \alpha \) keeps increasing, reaching 180\( ^\circ \) (collinear structure) for \( T \sim 448 \) K.

Finally, both \( \theta \) and \( \alpha \) are kept constant at 90\( ^\circ \) and 180\( ^\circ \) respectively. As \( T \) increases, \( \bar{S} \) and \( \vec{m}^2 \) decrease until, for \( T = T_{c2} \sim 1041 \) K, the former gets equal to zero (paramagnetic structure). Another set of parameters obtained from the same \( X_n \)'s is

\[ J_1 = -0.00223, J_2 = -0.0005, J_3 = 0.00297, J_4 = 0.0010, J_5 = 0.0008, B_1 = -0.000482, B_2 = -0.00034, B_3 = -0.000155, B_4 = -0.0001, B_5 = -0.000053, D_2 = 0.0005. \]

The results are qualitatively the same as for the previous set, and the \( T_c \)'s are 442.5, 455.3 and 1116 K. Notice that while \( T_{c1} \) and \( T_{c2} \) decrease with respect to the previous ones, \( T_{c3} \) increases (a behaviour we found in many other cases).
Experimentally, the transition "conical ↔ helical" is indeed observed (\( T_{c1} \approx 320 \text{ K} \)), but from the helical the system goes to the PM structure (\( T_{c2} \approx 420 \text{ K} \)) skipping the collinear structure\([4]\). However, the complete sequence of transitions obtained in this work is observed in a slightly different compound, namely \( \text{La}_{1-x} \text{Y}_{x} \text{Mn}_{2} \text{Ge}_{2} \), for \( 0.1 < x < 0.2 \) (see fig.13 of ref.4). There, as in our results, the temperature range of the helical structure is relatively small.

It is worthwhile to notice that in the PM phase the spins are not totally disordered (i.e. the states with \( m = \pm \frac{1}{2} \) are less populated than those with \( m = \pm \frac{3}{2} \), so that \( m^2 \) is not equal to 1.25) until a much higher \( T \) is reached.

V. NUMERICAL CALCULATIONS. \( \text{LaMn}_{2} \text{Si}_{2} \)

As for \( \text{LaMn}_{2} \text{Ge}_{2} \), the interaction constants are obtained from the \( X_i \)'s used in ref.7. Concerning the \( S \) of Mn, now both \textit{ab initio} calculations\([7,12]\) and experimental results\([2,5]\) give a magnetic moment near to \( \frac{3}{2} \mu_{\text{B}} \), which by Landé formula and for a quenched orbital momentum gives a total angular momentum (denoted \( S \) in this work) equal to 1. In this case no conical structure is obtained for our Hamiltonian. However, \textit{ab initio} studies\([12]\) indicate that this lowering of the magnetic moment is due to Si-Mn hybridization, so that, as for \( X = \text{Ge} \), we have \( S = 1.5 \).

Another possibility, leading to the same \( S \), is that of having an incomplete quenching, \( L = 1 \), with the total spin momentum and the total angular momentum both equal to \( \frac{3}{2} \). This would give a Landé factor \( g = 26/15 \) i.e. a magnetic moment \( \mu = 2.6 \mu_{\text{B}} \), in good agreement with the above value.

Now the set of parameters which we found to give the closest agreement with experiment is:

\[
\begin{align*}
J_1 &= -0.00045406, \quad J_2 = -0.0003, \quad J_3 = 0.00177942, \quad J_4 = 0.0010, \quad J_5 = 0.0006, \\
B_1 &= -0.00045278, \quad B_2 = -0.0003, \quad B_3 = -0.00024431, \quad B_4 = -0.00017, \quad B_5 = -0.00010941, \quad D_2 = 0.000.
\end{align*}
\]

The behaviour of the \( J_{\nu} \), \( B_{\nu} \) in terms of \( \nu \) is very similar to that of the Ge compound.

Proceeding as for the latter we have at LT a conical structure with \( \theta \sim 53^\circ \) and \( \alpha \sim 130^\circ \) The agreement of these angles with experiment is not as good as for the Ge compound\([15]\).

As \( T \) increases, both \( \theta \) and \( \alpha \) increase, but now the former does it more slowly, so that \( \alpha \) reaches \( 180^\circ \) (i.e. the canted structure is attained) while \( \theta \) is still nearly \( 54^\circ \). This happens at \( T_{c1} \approx 269 \text{ K} \).

When \( T \) (keeps increasing (with \( \alpha = 180^\circ \)) \( \theta \) increases more rapidly, reaching \( 90^\circ \) for \( T = T_{c2} \sim 393 \text{ K} \). This sequence of transitions is the one observed experimentally\([5]\).

Finally, in the collinear structure \( \bar{S} \) and \( m^2 \) decrease with increasing \( T \), until, for \( T = T_{c3} \sim 1061 \text{ K} \), the former gets equal to zero (paramagnetic structure). The experimental values reported in ref.5 are \( T_{c1} = 45 \text{ K} \) and \( T_{c2} = 305 \text{ K} \) (the \( T > 305 \text{ K} \) region is not studied therein).

VI. ABOUT THE MFA’S.

There are many ways of making a MFA in systems like those we study here. One can approximate the exact expression of the energy of the spin at the reference site \((i = 0)\) in the field of the spins at the other sites \((j)\) in different ways, and thereafter replace \( m_j \) and \( m_j^2 \) by their average values \( \bar{S} \) and \( \bar{m}^2 \) respectively. Some of such approximations are the following:

- since in the expression for the total energy the \( i \) and \( j \) indices are dummy, one can replace the terms with \( (m_i^2 + m_j^2) \) in the summation over \( i, j \) by \( 2m_i^2 \) or \( 2m_j^2 \) before of choosing site \( i = 0 \) as reference;

- one can replace in \( H_J \) each spin operator \( \hat{S}_i \) by \( \bar{S}_i + \hat{\Delta}_i \) with \( \hat{\Delta}_i \equiv \hat{S}_i - \bar{S}_i \), and neglect higher-order terms in the \( \Delta \) operators\([3]\);

- one can do the same thing for the \( m_i m_j \) term in (8);

- for the \( m_i^2 m_j^2 \) one can proceed analogously to the case of bilinear terms introducing the operators \( \hat{\Delta}_{2,i} \equiv \bar{S}_i^2 - \bar{m}_i^2 \).

Having chosen the approximate expression of the energy, one can make different choices for the excited states available to the spin (see below).

In this work we found that one can obtain completely different results with different MFA’s. The most interesting example of this (and the only one for which we have an explanation!) is the spectacular decrease of the calculated critical temperatures when the spin at the reference site is allowed to have a set of excited states with a continuous energy spectrum, instead of a discontinuous one.

In ref.7, with a certain set of parameters and a MFA using a discontinuous spectrum, we obtained for the \( T_c \)'s of \( \text{LaMn}_{2} \text{Ge}_{2} \) the values 1943 K, 2318 K and 4211 K. With the same parameters and one of the MFA’s used in this
work (continuous spectrum) the corresponding values are 468, 473 and 977. A similar situation occurs for the Ge compound. This difference in the $T_c$'s can be explained as follows.

In ref.7 the spin at site $i$ could be in any of the eigenstates $|m>$ with $m = \pm \frac{1}{2}, \pm \frac{3}{2}$ with respect to the $(\theta, \alpha)$ direction corresponding to the directions of the other spins. As for any given $T \theta$ and $\alpha$, as well as $S$ and $m^2$, are fixed, for any set of values of these four quantities the spin at the reference site needs a finite energy in order to jump to an excited state.

In the present work, as described in section III, that spin can have (in addition to the different $m$ values) any orientation $(\theta_i, \phi_i)$, i.e. it can vary its orientation by an infinitesimal angle, so that it can acquire a small degree of disorder at a very low $T$. This disordering causes a decrease in $S$, which in its turn reduces the field at the reference site, making easier for the spin to jump to an excited state. In addition to this, at higher $T$'s the $m = S - 1$ state begins to be populated, so that also $m^2$ starts to decrease from its LT value $S^2$. This feedback process must be what causes the great decrease of the calculated $T_c$'s.

Concerning the way of calculating the energy of the state at the reference site, in this work we tried several formulae, i.e. several MFA's, and ended up using three of them (that we call MFA0, MFA2 and MFA4). Let us consider $\sum_{i,j} U_{ij} B_{ij}$ (in which, however, as what we need is the energy per site the summation over $i$ is not carried out).

In MFA0 we simply replace $(m_i$ and $m_j^2)$ by $\bar{S}$ and $\bar{m}^2$ respectively.

In MFA2 we replace $\sum_{i,j} U_{ij} B_{ij}(m_i^2 + m_j^2)(p_{ij}^2 + 1)$ by $2 \sum_{i,j} U_{ij} m_i^2(p_{ij}^2 + 1)$, after which we take $i$ as the reference site ($i = 0$). This is a usual procedure in order to have a more exact value of the total energy before making a MFA. In this case, in which we do not carry out the summation over $i$, it is not clear this to be an improvement.

In MFA4 we use the $\hat{\Delta}$ and $\Delta_2$ operators for the terms bilinear and biquadratic in $m_i$ and $m_j$, after which we proceed as for the MFA2.

In all cases the spin at site $i$ is given as described in section III.

In the calculations we carried out for the Ge compound the sequence "conical $\longleftrightarrow$ helical $\longleftrightarrow$ collinear $\longleftrightarrow$ PM" was obtained for several sets of parameters with the MFA2, but not with the MFA4 (where in most cases the system of equations to be solved (section IV) ceases to have a solution above a certain temperature).

Surprisingly, for the Si compound something nearly opposite is true: the sequence "conical $\longleftrightarrow$ canted $\longleftrightarrow$ collinear $\longleftrightarrow$ PM" was obtained for three sets of parameters with the MFA4 and only for one set with the MFA2 (in the latter case the free energy is discontinuous at $T = T_{c2}$).

With the MFA0 we obtained the mentioned sequences for only one set of parameters for each of the compounds (in the case of Si, with the discontinuity at $T_{c2}$).

VII. CONCLUSION.

This work is concerned with the hypothesis that the great variety of magnetic structures observed in different RT$_2$X$_2$ compounds at different temperatures can be explained by assuming that the localized magnetic moments are subjected to bilinear and biquadratic interactions and that the interaction constants can be derived from ab initio calculations of the energies of different unstable structures. The following results of this work give support to this hypothesis:

- starting from ab initio energies we find a set of interaction constants for which the observed structures and the transitions between them are predicted for LaMn$_2$Si$_2$;
- the interaction constants we obtained by the same procedure for LaMn$_2$Ge$_2$ yield the observed structures and the transitions between them of a very similar compound, i.e. La$_{0.8}$Y$_{0.2}$Mn$_2$Ge$_2$;
- for both compounds the dependence of the constants on the interatomic distance is perfectly sound for both the bilinear and biquadratic interactions;
- due to the many competing interactions, the behaviour of the system is very sensitive to small changes of the constants, so that it can be expected that the same model can be applied to other compounds of this family, for which different magnetic structures have been observed.

If our hypothesis is valid, this paper is only a first step towards a complete theory. Among the additional studies that should be carried out we can list the following.

Further work is required to improve the calculation of the interaction constants.
It is important to understand the pros and cons of each MFA, in order to adopt the most adequate one.
It is likely that more neighbours must be included in the calculation.
One should also take into account a possible variation of the interaction constants with temperature due to the change of the lattice constants, which implies a variation of the interatomic distances.
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[13] To speed up the calculation and improve its accuracy, the integrations over $\phi_0$ coming from eq.(12) are carried out analytically, the results being series of the modified Bessel functions $I_n(x)$ \[11\].
[14] Of course, some $J_n$'s can be near to zero, in which case it can be smaller in modulus than the corresponding $B_n$.
[15] This comes from the \textit{ab initio} results of ref.12, as pointed out therein.