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Magnetic phase diagrams for three coupled magnetic moments

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Abstract

For an arbitrary system of three coupled magnetic moments (sublattices), the stable magnetic structures have been determined as a function of the applied field. At a certain field, only one structure (phase) appears to be stable. A classification of the possible collinear and non-collinear structures is worked out. The transitions, from a collinear structure to a non-collinear one, or vice versa, are smooth: no jump in the total magnetic moment does occur.

1. Introduction

In the past few years, enormous progress has been made in the study of the basic magnetic interactions in various RT-compounds (R: rare earth, or Y; T: transition metal) (see e.g. [1]). In many of these materials, the magnetic structures can be described in terms of a two-sublattice model with the following features: at low temperatures, all R-moments are parallel. The same is true for the T-moments. In many cases, the crystalline anisotropy is for the greater part due to the crystalline electric field acting on the R-moments. The ground state, at zero applied field, is collinear (ferromagnetic or antiferromagnetic, depending on the sign of the magnetic coupling between the two moments). In an applied field, however, a non-collinear structure (also called phase) can be formed (see e.g. [2]). In the absence of crystalline anisotropy, the magnetization curve has then a linear portion: $M = \mu_0 H/|n|$, of which the slope is determined by the (absolute value of the) coupling parameter n (here taken to be negative for anti-ferromagnetic coupling). In practice, the influence of the crystalline anisotropy can be minimized by performing the experiments on an assembly of small, freely (re)orientable, particles (see e.g. [3]): during the magnetization process, the particles are reoriented in such a way that the crystalline anisotropy energy is minimized. If the T-anisotropy can be neglected, the R-moments remain directed along the easy axis. In this way, from the slope of the linear portion of the magnetization curve, the coupling parameter n can be, and has been, determined.

One may ask, whether this model is not too simple. In ferritic materials it is well known, that a magnetic sublattice sometimes is split in two (antiferromagnetically coupled) subsystems. Moreover, in many of the RT-compounds under consideration, the R-atoms occupy different (but equivalent) crystallographic sites: a splitting in two subsystems might not be unreasonable. Verhoef [3] treats a case, in which, because of the tetragonal local symmetry, the rare-earth moments tend to point into two different (but equivalent) directions. Consequently, here, and in many more cases, a two-sublattice model is not adequate. For instance, even in the case that two different kinds of rare-earth moments are placed on one sublattice, a three-sublattice model is necessary, because the

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different R-atoms in general have different coupling parameters.

For the ferritic materials just mentioned, extensive studies have been performed of three-sublattice models, including spontaneous breaking up of one sublattice into two (see e.g. Ref. [4], and references therein). Clark et al. focus attention on the temperature dependence of the sublattice magnetizations [5]. The author, however, is not aware of a systematic study of the effect of an applied field. Recently, a simplified three-sublattice model was applied for the description of the system $RMn_{6-x}Cr_xSn_6$ [6]. Here, the Mn-atoms are imagined to occupy two equivalent sublattices (so with equal magnetic moments, and equal coupling parameters with respect to the R-sublattice).

In this article, a complete treatment is offered for the determination of the magnetization curves (i.e. the stable magnetic structures) in a three-sublattice model. Since the magnetization of a subsystem may depend on the effective field acting on that sublattice, general expressions for these effective fields are derived and presented. In this article, however, the moments are taken to be of constant magnitude. In Section 2, the stability criteria are explained, and applied to establish a classification of the possible stable structures. In Section 3, the class "three equal coupling parameters" is treated. In Section 4, the class "only two equal coupling parameters" is shown to yield the most interesting magnetization curves. For this interesting case, the stability criteria for the possible magnetic structures are gathered in Appendix A. The phase diagrams are presented in Appendix B (actually, the magnetization curves with an emphasis on the critical fields, where transitions from a collinear phase to a non-collinear one do occur). In Section 5, the feasibility of the occurrence of non-collinear structures is discussed for the most general class "three different coupling parameters". In Section 6, some concluding remarks are gathered and an outlook on further generalizations is offered.

2. Stability criteria, classification

We consider a system of magnetic moments $\boldsymbol{M}_1, \boldsymbol{M}_2, \boldsymbol{M}_3$, with fixed non-zero magnitudes. The moments are coupled, either ferromagnetically (positive molecular-field constants n_{ij}, labelled $i \neq j$) or antiferromagnetically ($n_{ij} < 0$). The influence of crystalline anisotropy is assumed to be negligible. The moments are placed in an external field $\mu_0 \boldsymbol{H}$. The (free) energy is given by

$$E = -(n_{12}\boldsymbol{M}_1\boldsymbol{M}_2 + n_{23}\boldsymbol{M}_2\boldsymbol{M}_3 + n_{13}\boldsymbol{M}_1\boldsymbol{M}_3) - \mu_0\boldsymbol{H}(\boldsymbol{M}_1 + \boldsymbol{M}_2 + \boldsymbol{M}_3). \quad (1)$$

Using the method of Lagrange multipliers, minimization of the energy with respect to the (directions of the) moments yields the equilibrium conditions below (see Eqs. (2)). These equations can be derived also, in a mathematically clear way, by direct differentiation to e.g. the x and y components only, or, in a physically clear way, by stating that, in order to have a stable configuration, the effective fields acting on each magnetic moment should be oriented along that moment, i.e. can be written as $a_j\boldsymbol{M}_j$ with positive a_j.

Mathematically, $a_j > 0$ is a necessary condition for a (local) minimum of the energy (indeterminate if $a_j = 0$). Moreover, from the following mathematical considerations (leading to e.g. Eqs. (7b) and (11)), or from an obvious physical argumentation, we conclude that the total magnetic moment, $\boldsymbol{M}_t = \boldsymbol{M}_1 + \boldsymbol{M}_2 + \boldsymbol{M}_3$, should be oriented along the applied non-zero field $\boldsymbol{H}$, i.e. $\boldsymbol{M}_t = Q\mu_0\boldsymbol{H}$ with, similarly, $Q \geqslant 0$. In order to facilitate later derivations, we add this requirement as a separate equation. The resulting expressions can be arranged as

$$a_1\boldsymbol{M}_1 - n_{12}\boldsymbol{M}_2 - n_{13}\boldsymbol{M}_3 = \mu_0\boldsymbol{H}, \quad (2a)$$

$$-n_{12}\boldsymbol{M}_1 + a_2\boldsymbol{M}_2 - n_{23}\boldsymbol{M}_3 = \mu_0\boldsymbol{H}, \quad (2b)$$

$$-n_{13}\boldsymbol{M}_1 - n_{23}\boldsymbol{M}_2 + a_3\boldsymbol{M}_3 = \mu_0\boldsymbol{H}, \quad (2c)$$

$$\boldsymbol{M}_1 + \boldsymbol{M}_2 + \boldsymbol{M}_3 = \boldsymbol{M}_t = Q\mu_0\boldsymbol{H} \quad (\boldsymbol{H} > 0). \quad (3)$$

The coefficients a_j and the discriminant (determinant of the coefficient matrix of the set of Eqs. (2a)–(2c)) should be non-negative in order to have an energy minimum. The determinant is

$$D = a_1a_2a_3 - \{a_1n_{23}^2 + \text{cycl.}\} - 2n_{12}n_{23}n_{13}. \quad (4)$$

Taking the components perpendicular to a non-zero field $\boldsymbol{H}$ separately, one finds immediately that in case the determinant D does not vanish, all these components vanish: we have a collinear structure or phase.

At zero field strength the determinant should vanish. A vanishing determinant is also a necessary condition for the occurrence of a non-collinear structure.

In the collinear cases Eq. (3) is fulfilled automatically. For non-vanishing D we can determine the parameter Q (the "susceptibility") by inversion of the matrix of the coefficients in Eqs. (2). Most of the following relations, however, are formulated in such a way that they are also valid and useful for the case that the determinant vanishes.

The elements Q_{ij} of the inverse matrix are determined by

$$DQ_{11} = m_{11} = a_2 a_3 - n_{23}^2, \tag{5a}$$

(cycl.)

$$DQ_{12} = m_{12} = a_3 n_{12} + n_{13} n_{23}. \tag{5b}$$

Here, DQ_{ij} is the "ordinary" product of the determinant D and the element Q_{ij}, and m_{ij} represents the appropriate minor (subdeterminant with appropriate sign). The definitions of m_{ij} presented here are of course valid also in case D vanishes.

The minor m_{11}, i.e. the expression at the right-hand side of Eq. (5a), is the discriminant of the energy expression (1) in which the moment $\boldsymbol{M}_1$ is fixed, describing the behaviour of the moments $\boldsymbol{M}_2$ and $\boldsymbol{M}_3$ (in different "external" fields $\mu_0 \boldsymbol{H} + n_{12}\boldsymbol{M}_1$, and $\mu_0 \boldsymbol{H} + n_{13}\boldsymbol{M}_1$, respectively). Consequently, the minors m_{ii} should also be non-negative (whether or not D vanishes).

For non-zero determinant D, the inverse matrix does exist. We find straightforwardly $\boldsymbol{M}_j = Q_j \mu_0 \boldsymbol{H}$, with $Q_j = Q_{1j} + Q_{2j} + Q_{3j}$. Evidently, this corresponds to a collinear state. We have

$$\begin{aligned} DQ_1 &= m_{11} + m_{12} + m_{13} \\ &= a_2 a_3 + a_3 n_{12} + a_2 n_{13} \\ &\quad + n_{23}(n_{12} + n_{13}) - n_{23}^2 \quad \text{[cycl.]}, \end{aligned} \tag{6a}$$

and

$$D\boldsymbol{M}_1 = (m_{11} + m_{12} + m_{13})\mu_0 \boldsymbol{H} \quad \text{[cycl.]}. \tag{6b}$$

Notice that Eq. (6b) can be derived quite generally in a form where both the left-hand side and the right-hand side are multiplied by a_2 or by a_3. Hence, Eq. (6b) holds unless, by incident, both a_2 and a_3 happen to vanish. For a non-collinear state, Eq. (6b) tells us only that the determinant D and the sums $\sum_j m_{ij}$ ($i = 1, 2, 3$) all vanish (see also Eq. (10a)). The "susceptibility" Q (see Eq. (3)) is given by $Q = Q_1 + Q_2 + Q_3$. Hence,

$$DQ = a_1 a_2 + 2a_3 n_{12} + 2n_{13} n_{23} - n_{12}^2 + \text{cycl.} \tag{7a}$$

or

$$D\boldsymbol{M}_t = \left(\sum_{ij} m_{ij}\right)\mu_0 \boldsymbol{H}. \tag{7b}$$

Other useful, and quite general, relationships are

$$m_{11}\boldsymbol{M}_2 = (a_3 + n_{23})\mu_0 \boldsymbol{H} + m_{12}\boldsymbol{M}_1, \tag{8a}$$

$$m_{11}\boldsymbol{M}_3 = (a_2 + n_{23})\mu_0 \boldsymbol{H} + m_{13}\boldsymbol{M}_1. \tag{8b}$$

Hence,

$$\begin{aligned} m_{11}\boldsymbol{M}_t &= (a_2 + a_3 + 2n_{23})\mu_0 \boldsymbol{H} \\ &\quad + (m_{11} + m_{12} + m_{13})\boldsymbol{M}_1. \end{aligned} \tag{8c}$$

Let us first discuss the cases in which the applied field H vanishes. Then, from Eqs. (8a) and (8b) we infer that either all minors have non-zero values (certainly a collinear structure) or all minors do vanish (then a non-collinear structure may occur). In the latter case we have

$$a_1 = -n_{12} n_{13}/n_{23}; \qquad a_2 = -n_{12} n_{23}/n_{13};$$
$$a_3 = -n_{13} n_{23}/n_{12}. \tag{9}$$

Consequently, since a_j should be positive (for all j), a non-collinear zero-field phase can only exist in case all molecular field constants are negative (antiferromagnetic) or in case only one such constant is negative, the other two being positive (ferromagnetic).

In the following, we find the zero-field structures without difficulty as the limits (for vanishing field) of the structures in non-zero field. Hence, in the following we assume non-zero values for H, unless stated otherwise.

Let us further discuss the occurrence of non-collinear structures (or phases), in a finite field.

As stated above, a non-collinear phase is only possible in case D vanishes. Moreover, by interchanging the first colon in the set of Eqs. (2a)–(2c) with the right-hand side, and taking into account that, again, the determinant should vanish in case a non-collinear solution exists (non-zero components perpendicular to $\boldsymbol{M}_1$ should exist), we find that the sum of the minors should vanish:

$$m_{11} + m_{12} + m_{13} = 0. \tag{10a}$$

Or, more generally,

$$\sum_j m_{ij} = 0 \quad (i = 1,2,3). \tag{10b}$$

Notice that in Eq. (6b) the left-hand side (D) and the right-hand side ($m_{11} + m_{12} + m_{13}$) both vanish. Of course, the same observation can be made with respect to Eq. (7b). Inserting the result of Eq. (10a) in Eq. (8c) we find

$$m_{11}\boldsymbol{M}_t = m_{11}(\boldsymbol{M}_1 + \boldsymbol{M}_2 + \boldsymbol{M}_3)$$
$$= (a_2 + a_3 + 2n_{23})\mu_0\boldsymbol{H} \quad \text{[cycl.]}. \tag{11}$$

For a non-vanishing m_{ii} (taken to be m_{11}) this equation proves the validity of Eq. (3) for the non-collinear case. The parameter Q can be calculated straightforwardly. More in detail, Q should be non-negative. This can be established by considering the expression

$$a_2(a_2 + a_3 + 2n_{23}) = (a_2 + n_{23})^2 + (a_2a_3 - n_{23}^2),$$

which is positive ($(a_2a_3 - n_{23}^2) = m_{11} > 0$), hence

$$Q = (a_2 + a_3 + 2n_{23})/m_{11} > 0. \tag{12}$$

Moreover, Eqs. (8a) and (8b) show that $\boldsymbol{M}_2$ and $\boldsymbol{M}_3$ are linear combinations of $\boldsymbol{H}$ and $\boldsymbol{M}_1$. Consequently, if a non-collinear structure exists, it is a coplanar one (in this case, $m_{11} > 0$, with non-collinear $\boldsymbol{H}$ and $\boldsymbol{M}_1$).

Conversely, assuming m_{11} to vanish (i.e. $a_2a_3 - n_{23}^2 = 0$), and inferring from Eq. (8c), or rather from Eq. (11), that then also $(a_2 + a_3 + 2n_{23})$ vanishes, we find $a_2 = a_3 = -n_{23}$. Thus, since the Eqs. (2b) and (2c) must not be false (which they would be in case $n_{12} \neq n_{13}$), they have to be identical (so, $n_{12} = n_{13}$). By cyclic permutation we conclude, that Eq. (3) (i.e. Eq. (11)) holds as soon as any m_{jj} does not vanish. In case all m_{jj} do vanish, however, Eqs. (2a)–(2c) are identical, and all molecular field constants are equal, i.e.

$$-a_j = n_{12} = n_{23} = n_{13}[= n] \quad (j = 1,2,3).$$

We classify this case as Case I. Obviously, in this case, we have $Q = -1/n$, thus again Eq. (3) appears to be valid. Moreover, since the coefficients a_j have to be non-negative, n is negative, ensuring a positive sign of Q. Furthermore, the resulting non-collinear structures in this case do not have to be coplanar, in contradistinction to the result obtained above for the other cases. In Section 3 we show in detail that a physically correct solution may exist.

For any non-collinear state, Eqs. (2a)–(2c) should be dependent. Consequently, we have either two determining equations (rank 2) or we are left with one independent equation (rank 1). Because Eqs. (2a)–(2c) have (non-zero) identical right-hand sides, we see immediately, that rank one (i.e. $m_{ij} = 0$ for all i and j) implies the equality of all molecular-field constants, thus again Case I, to be discussed further in Section 3. So, in the remaining part of the present section, we restrict the discussion to the case that the molecular-field constants are not all equal. Then, we have two determining equations (rank 2), which we choose to be, without loss of generality, the Eqs. (2b) and (2c). Above, we showed already that vanishing of m_{11} implies the identity of Eqs. (2b) and (2c), now in contradiction to our present hypothesis. Hence, we can rule out the possibility that m_{11} vanishes.

Let us examine the situation further by assuming that $\boldsymbol{M}_3$ happens to be collinear with $\boldsymbol{H}$. Then, from Eq. (8b) (with non-collinear $\boldsymbol{H}$ and $\boldsymbol{M}_1$), we find $m_{13} = 0$, i.e. $a_2 = -n_{12}n_{23}/n_{13}$, and, hence, $m_{11} = -(n_{23}/n_{13})m_{12}$. In combination with $m_{11} = -m_{12}$ (from Eq. (10a)), and recalling that m_{11} does not vanish, we find $n_{13} = n_{23}$, leading to $a_1 = a_2 = -n_{12}$. This situation, i.e. $n_{12} \neq n_{23} = n_{13}$, will be classified as Case II and will be discussed in Section 4.

Another particular situation is a non-collinear state in which for instance $\boldsymbol{M}_1$ and $\boldsymbol{M}_2$ would be collinear, but not collinear with $\boldsymbol{H}$. Later on, we refer to this stituation as to the pseudo-two-sublattice model. Then, in Eq. (8a), $(a_3 + n_{23})$ should vanish, i.e. $a_3 = -n_{23}$. Inserting this result in the explicit expressions for the minors in Eq. (10a), we can write

$$m_{11} + m_{12} + m_{13} = (a_2 + n_{23})(n_{13} - n_{23}) = 0.$$

Since we also have $m_{11} = -n_{23}(a_2 + n_{23}) > 0$ (i.e. $a_2 + n_{23}$ does not vanish), we must have $n_{13} = n_{23}$. The feasibility of this situation is also discussed in

Section 4 (Case II). The other way round, in case two molecular-field constants are equal, say $n_{13} = n_{23}$, we find, for a non-collinear state, $0 = m_{11} + m_{12} + m_{13} = (a_2 + n_{12})(a_3 + n_{13})$. Hence, the two non-collinear states discussed presently (with either $a_2 + n_{12} = 0$ or $a_3 + n_{13} = 0$) are the only ones which can exist in Case II.

Finally, we discuss briefly the situation in case $n_{12} \neq n_{23} \neq n_{13} \neq n_{12}$, classified as Case III and discussed in more detail in Section 5. From the discussion above, it is evident that (still in non-zero field) no minor m_{ii} does vanish, so the Eqs. (2a)–(2c) are pairwise independent. Neither of the particular non-collinear structures encountered in Case II can occur here. Still, we shall see in Section 5 that non-collinear phases do exist for certain field and parameter values. From the general discussion in this section, we conclude that these structures are coplanar, and that Eq. (3) holds, with a positive susceptibility Q (see Eq. (12)). For zero field, of course the general remark made above (Eq. (9)) holds here too: a non-collinear phase may exist in case all interactions are antiferromagnetic, or in case only one interaction is antiferromagnetic (see Eq. (9)).

3. Case I: all molecular-field constants are equal ($n_{12} = n_{23} = n_{13} [= n]$)

It is instructive to write the energy in the form

$$E = -n(\boldsymbol{M}_1\boldsymbol{M}_2 + \text{cycl.}) - \mu_0 \boldsymbol{H}\boldsymbol{M}_t$$

$$= -\tfrac{1}{2}nM_t^2 + \tfrac{1}{2}n\sum M_j^2 - \mu_0 HM_t \qquad (13a)$$

$$= -\tfrac{1}{2}nM_t^2 - \mu_0 HM_t + \text{const.} \qquad (13b)$$

We rederive quickly some results already mentioned in Section 2. By combining Eqs. (2a) and (3) (and so on) we find

$$(n + a_1)\boldsymbol{M}_1 = (n + a_2)\boldsymbol{M}_2$$

$$= (n + a_3)\boldsymbol{M}_3 = (nQ + 1)\mu_0 \boldsymbol{H}. \qquad (14)$$

Consequently, a non-collinear state is only possible if

$$a_j = -n > 0 \quad (\text{all } j), \qquad (15a)$$

$$Q = -1/n > 0. \qquad (15b)$$

This can only occur in case $n < 0$, i.e. all interactions are antiferromagnetic and equal. We refer to this possibility as "Case IA". The other possibility, $n > 0$, will be classified as "Case IF" (see below).

In this case we take (without loss of generality)

$$M_3 \geqslant M_2 \geqslant M_1. \qquad (16)$$

In order to indicate the collinear states, we define $\varepsilon_1, \varepsilon_2, \varepsilon_3$ with

$$M_c[\varepsilon_1, \varepsilon_2, \varepsilon_3] = \varepsilon_1 M_1 + \varepsilon_2 M_2 + \varepsilon_3 M_3 \quad (\varepsilon_j = \pm 1). \qquad (17)$$

We distinguish the following collinear states or phases, in an obvious notation:

[+ + +] $\quad M_t = M_1 + M_2 + M_3$;

[+ − +] $\quad M_t = M_1 - M_2 + M_3$;

[− + +] $\quad M_t = -M_1 + M_2 + M_3$;

[+ + −] $\quad M_t = M_1 + M_2 - M_3$;

or

[− − +] $\quad M_t = -M_1 - M_2 + M_3$.

Notice, that the states [− + −] and [+ − −] are omitted, since they have a negative total magnetization. It is physically evident that the inverted states ([+ − +] and [− + +], respectively) are favoured. This can also be seen directly by application of Eq. (18). More precisely, however, the states [− + −] and [+ − −] are not stable for any choice of the molecular-field parameter n. This statement is proved in the detailed discussion hereafter.

The energy difference between two collinear states can be written as

$$E_{c1} - E_{c2} = (M_{c1} - M_{c2})\left\{-\tfrac{1}{2}n(M_{c1} + M_{c2}) - \mu_0 H\right\}. \qquad (18)$$

Using the notation given above (Eq. (17)), we find for the collinear states:

$$(a_j + n)\varepsilon_j M_j = \mu_0 H + nM_c[\varepsilon_1, \varepsilon_2, \varepsilon_3] \qquad (19)$$

and, for instance

$$m_{33} = (\mu_0 H + n\varepsilon_3 M_3)(\mu_0 H + nM_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]) / \varepsilon_1\varepsilon_2 M_1 M_2; \tag{20a}$$

$$m_{13} = n\varepsilon_1 M_1(\mu_0 H + nM_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]) / \varepsilon_1\varepsilon_2 M_1 M_2; \tag{20b}$$

$$m_{23} = n\varepsilon_2 M_2(\mu_0 H + nM_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]) / \varepsilon_1\varepsilon_2 M_1 M_2; \tag{20c}$$

$$DQ_3 = m_{13} + m_{23} + m_{33} = \varepsilon_3 M_3(\mu_0 H + nM_c[\varepsilon_1, \varepsilon_2, \varepsilon_3])^2 / \varepsilon_1\varepsilon_2\varepsilon_3 M_1 M_2 M_3; \tag{21a}$$

$$DQ = DQ_1 + DQ_2 + DQ_3 = M_c[\varepsilon_1, \varepsilon_2, \varepsilon_3](\mu_0 H + nM_c[\varepsilon_1, \varepsilon_2, \varepsilon_3])^2 / \varepsilon_1\varepsilon_2\varepsilon_3 M_1 M_2 M_3 \tag{21b}$$

$$D = \mu_0 H(\mu_0 H + nM_c[\varepsilon_1, \varepsilon_2, \varepsilon_3])^2 / \varepsilon_1\varepsilon_2\varepsilon_3 M_1 M_2 M_3. \tag{22}$$

From Eqs. (21b) and (22), we find $Q = (M_c[\varepsilon_1, \varepsilon_2, \varepsilon_3] / \mu_0 H)$, as expected.

From the condition $D \geqslant 0$, we infer from Eq. (22) that the only stable collinear states in Case I can be the states $[+++]$ and $[--+]$.

We apply these equations in the more detailed description of the Cases IF ($n > 0$) and IA ($n < 0$).

Case IF: $n_{12} = n_{23} = n_{13}[= n] > 0$.

Stable non-collinear states do not exist (see Eq. (15a)): $n > 0$, so a_j would become negative. Indeed, it is easy to show that in any collinear state other than $[+++]$, an effective field oriented along $\boldsymbol{H}$ is acting on an oppositely oriented moment. As an example, we consider the effective field acting on $\boldsymbol{M}_2$, in the state $[--+]$: $a_2 M_2 = \mu_0 \boldsymbol{H} + n(\boldsymbol{M}_3 + \boldsymbol{M}_1)$, or, using the parameters ε_j, $a_2\varepsilon_2 M_2 = \mu_0 H + n\varepsilon_3 M_3 + n\varepsilon_1 M_1$ with $\varepsilon_1 = \varepsilon_2 = -1$ and $\varepsilon_3 = 1$, yields $a_2 M_2 = -\mu_0 H - n(M_3 - M_1) < 0$, evidently having the wrong sign.

So, the only stable state is $[+++]$. This is corroborated by the observation, that, at a certain field H, the collinear state with the largest total magnetic moment (i.e. the state $[+++]$ with $M_t = M_1 + M_2 + M_3$) is always the preferred one (see Eq. (18)).

Finally, it is necessary to show that in the state $[+++]$ the effective fields acting on the moments are correctly oriented, i.e. that the coefficients a_j (and Q) are positive. Indeed

$$a_j M_j = \{\mu_0 H + n(M_c[+++] - M_j)\} > 0;$$

$$Q = M_c[+++]/\mu_0 H > 0 \quad (j = 1, 2, 3). \tag{23}$$

For the sake of completeness, we remark that also the determinant D and the minors m_{jj} are properly positive (see Eqs. (20)–(22)).

The resulting magnetization curve is very simple indeed:

$M_t = M_c[+++]$ (constant: type C, see Table 1).

Case IA: $n_{12} = n_{23} = n_{13}[= n] < 0$.

Using Eq. (13b), with $M_t = -\mu_0 H/n$ (from Eqs. (15b) and (3)), we write the energy of a non-collinear state as

$$E_{nc} = -\left(\tfrac{1}{2}|n|\right)(\mu_0 H)^2 + \text{const.} \tag{24}$$

Many (coplanar and non-coplanar) non-collinear states exist. In fact, any combination of the vectors $\boldsymbol{M}_1, \boldsymbol{M}_2$ and $\boldsymbol{M}_3$ yielding $\boldsymbol{M}_t = \mu_0 \boldsymbol{H}/|n|$ forms a stable non-collinear state. The coefficients a_j are given by Eq. (15a): $a_j = |n|$, and $Q = 1/|n|$ (Eq. (15b)). Hence, the combination mentioned of the vectors $\boldsymbol{M}_1, \boldsymbol{M}_2$ and $\boldsymbol{M}_3$ yielding $\boldsymbol{M}_t = \mu_0 \boldsymbol{H}/|n|$ can be regarded as representing also the sum of the vectors $-n\boldsymbol{M}_1$, $-n\boldsymbol{M}_2$, and $-n\boldsymbol{M}_3$, yielding $\mu_0 \boldsymbol{H}$ in accordance with Eqs. (2a)–(2c). The effective fields are thus correctly oriented. With these values for the coefficients a_j (and Q) in the set of Eqs. (2a)–(2c) (and Eq. (3)), we see that the determinant D and all minors m_{ij} do vanish, in accordance with the results of the discussion in Section 2 (coefficient matrix of rank one).

From Eq. (24), it is clear that all these states have the same energy (at the same field). Since it is neither possible to construct a vector $\boldsymbol{M}_t = \boldsymbol{M}_1 + \boldsymbol{M}_2 + \boldsymbol{M}_3$ with a magnitude larger than $M_1 + M_2 + M_3$ (that of the "stretched" vectors $\boldsymbol{M}_1, \boldsymbol{M}_2$ and $\boldsymbol{M}_3$), nor with a magnitude smaller than zero or, in case $M_3 \geqslant M_1 +$

Table 1
Type of magnetization curve and possible configurations (Case II: $n_{12} \neq n_{13} = n_{23}$. The cases that can occur in case $M_2 = M_1$ are marked with an asterisk)

Type	Case	Consecutive phases	Extra conditions
C	*FF	[+ + +]	
	*AFa	[+ + +]	$B_c[+ + +] \leqslant 0$
LC	*FA	[ncF] [+ + +]	$M_3 = M_2 + M_1$ (compensation)
	*AA>	[ncF] [+ + +]	
NC	*AFb	[nc3++] [+ + +]	$B_c[- + +] \leqslant 0 < B_c[+ + +]$
	*AA<b	[nc3+−] [nc3++] [+ + +]	$M_2 = M_1$; $B_c[- - +] \leqslant 0$
CLC	*FA	[− − +] [ncF] [+ + +]	$M_3 > M_2 + M_1$
	*AA>	[− − +] [ncF] [+ + +]	
	*FA	[+ + −] [ncF] [+ + +]	$M_3 < M_2 + M_1$
	*AA>a	[+ + −] [ncF] [+ + +]	$M_3 < M_2 + M_1$; $B_c[+ + -] > 0$ (also if $M_3 < M_2 - M_1$)
CNC	AFc	[− + +] [nc3++] [+ + +]	$0 < B_c[- + +]$; ($M_2 > M_1$)
	*AA<c	[− − +] [nc3+−] [nc3++] [+ + +]	$M_2 = M_1$; $M_3 > 2M_1$; $0 < B_c[- - +]$
N^-CLC	*AA>b	[nc3−] [+ + −] [ncF] [+ + +]	$M_3 < M_2 + M_1$; $B_c[- + -] \leqslant 0 < B_c[+ + -]$
CN^-CLC	AA>c	[− + −] [nc3−] [+ + −] [ncF] [+ + +]	$M_3 < M_2 - M_1$; $0 < B_c[- + -]$
$CLCN^+C$	AA<a	[+ − +] [ncA] [− + +] [nc3++] [+ + +]	$M_3 > M_2 - M_1$; $B_c[+ - +] \leqslant 0$
	AA<	[− + −] [ncA] [− + +] [nc3++] [+ + +]	$M_3 < M_2 - M_1$
LCN^+C	AA<	[ncA] [− + +] [nc3++] [+ + +]	$M_3 = M_2 - M_1$
N^+CLCN^+C	AA<b	[nc3+−] [+ − +] [ncA] ... ...[− + +] [nc3++] [+ + +]	$M_3 > M_2 - M_1$; $B_c[- - +] \leqslant 0 < B_c[+ - +]$
CN^+CLCN^+C	AA<c	[− − +] [nc3+−] [+ − +] [ncA] ... ...[− + +] [nc3++] [+ + +]	$M_3 > M_2 + M_1$; $0 < B_c[- - +]$

Note: Type: C constant; L linear $M_1 = \mu_0 H/|n_{13}|$; N, N^+, N^- slope $1/|n_{12}|$. Case: indicates allowed range of molecular-field values. Collinear phases: [+++] and so on; non-collinear [ncF] and so on. Critical fields: $B_c[+ + +]$ and so on; see text for details.

M_2, smaller than the length of the smallest combination, $M_3 - M_1 - M_2$, we obtain straightforwardly that the non-collinear states can be constructed, and are stable, in case

$$\max[0, (M_3 - M_1 - M_2)] \leqslant M_t = \mu_0 H/|n| \leqslant M_3 + M_2 + M_1. \quad (25)$$

We may compare the energy of the non-collinear states with that of any collinear state. At a certain field, the energy difference between a collinear state (with $M_t = M_c$) and a non-collinear state is

$$E_c - E_{nc} = \tfrac{1}{2}|n|\{M_c - \mu_0 H/|n|\}^2. \quad (26)$$

Obviously, the non-collinear states have the lower energy, i.e. are preferred provided they exist. Hence, Eq. (25) defines a lower critical field, H_1 (possibly zero) and an upper critical field H_2, indicating the field values between which the non-collinear state is the preferred one:

$$\mu_0 H_1 = \max[0, |n|(M_3 - M_1 - M_2)], \quad (27a)$$

$$\mu_0 H_2 = |n|(M_1 + M_2 + M_3). \quad (27b)$$

From Eq. (27a) we infer, that we have to distinguish two possibilities, according to the sign of $(M_3 - M_1 - M_2)$. We identify these cases by indicating the ground state at zero field, i.e. we distinguish Case IA [$- - +$] and Case IA [nc], respectively.

Case IA[$- - +$]: $M_3 \geqslant (M_1 + M_2)$.

$$\mu_0 H_1 = |n|(M_3 - M_1 - M_2); \tag{28a}$$

$$\mu_0 H_2 = |n|(M_1 + M_2 + M_3). \tag{28b}$$

From the condition $m_{33} \geqslant 0$ we infer (using Eq. (20a)), that the collinear state [$+ + +$] is only stable at fields H exceeding H_2, and, analogously, that the state [$- - +$] is only stable at fields lower than H_1. In the intermediate region

$$\mu_0 H_1 = |n|(M_3 - M_2 - M_1) < \mu_0 H < |n|(M_3 + M_2 + M_1) = \mu_0 H_2, \tag{29}$$

only the non-collinear states are stable.

At lower fields, $\mu_0 H \leqslant \mu_0 H_1$, in the collinear structure [$- - +$], the effective field acting on $\boldsymbol{M}_1$, $a_1 M_1$, starts as $|n|(M_3 - M_2) - \mu_0 H$, decreases down to $|n|M_1$ at $H = H_1$, remains constant in the non-collinear phase up to $H = H_2$, and increases in the collinear phase (state [$+ + +$]) as $a_1 M_1 = \mu_0 H - |n|(M_3 + M_2)$. An analogous behaviour is found for $a_2 M_2$. $a_3 M_3$ increases as $\mu_0 H + |n|(M_1 + M_2)$ up to $a_3 M_3 = |n| M_3$ at $H = H_1$, remains constant up to $H = H_2$, and increases as $a_3 M_3 = \mu_0 H - |n|(M_1 + M_2)$ for larger fields.

The magnetization curve is shown in Fig. 1 (type CLC, see also Table 1). In the next section, we show that the critical fields, and the concurrent magnetic phases, in this particular case, do not vary in a large region of n_{12} values (keeping $n_{13} = n$ constant).

Case IA[nc]: $M_3 < (M_1 + M_2)$.

The non-collinear states exist for the field region

$$0 \leqslant \mu_0 H < |n|(M_3 + M_2 + M_1) = \mu_0 H_2, \tag{30}$$

i.e. up to the field where the fully oriented state [$+ + +$] becomes stable. In the same way as in the previous case, we can show, that no other collinear structure is stable, for any field. The field dependence of the effective fields in the non-collinear phase and in the high-field collinear phase is as given in the previous case.

The magnetization curve can be derived directly from Fig. 1 by shifting the origin upwards along the dashed line (type LC, see Table 1).

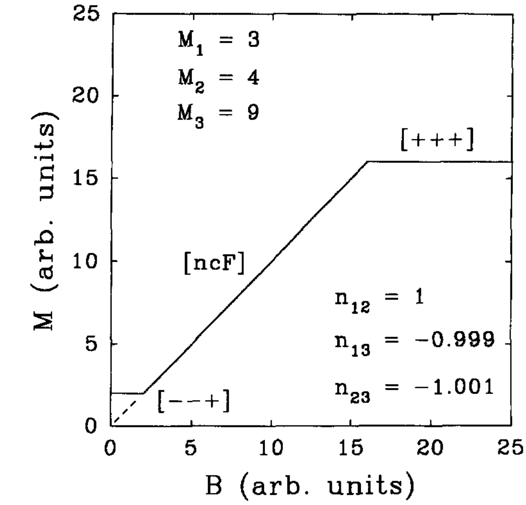

Fig. 1. Type CLC in Case IIFA, with $M_3 > M_1 + M_2$. Since $\boldsymbol{M}_1$ and $\boldsymbol{M}_2$ point always in the same direction, this case is referred to as the pseudo-two-sublattice model. The small difference between n_{13} and n_{23} is introduced by the computer program (Section 6), but can be ignored here. Critical fields: $\mu_0 H_1 = |n_{13}| M_c[- - +] = |n_{13}|\{M_3 - (M_2 + M_1)\}$ and $\mu_0 H_2 = |n_{13}| M_c[+ + +] = |n_{13}|\{M_3 + (M_2 + M_1)\}$. The same critical fields occur in Case IA[$- - +$] (Eq. (29)). Type LC can be obtained by decreasing M_3 until $M_3 = M_1 + M_2$, i.e. by shifting the origin diagonally upwards, until [ncF] is reached.

4. Case II: only two equal molecular-field constants ($n_{12} \neq n_{23} = n_{13}$)

Without loss of generality we assume in this case: $M_2 \geqslant M_1$.

Notice, that now M_3 can adopt any value larger or smaller than M_2 or M_1, or than $M_2 + M_1$ or $M_2 - M_1$.

In Section 2, we derived that only two (main) types of non-collinear structures may exist, both coplanar:

- $\boldsymbol{M}_1$ and $\boldsymbol{M}_2$ collinear ($a_3 = -n_{13}$; pseudo-two-sublattice model), i.e. with $\boldsymbol{M}_{12} = \boldsymbol{M}_1 + \boldsymbol{M}_2$, we have either $M_{12} = M_1 + M_2$, or $M_{12} = M_2 - M_1$. We refer to these states as [ncF] and [ncA], respectively.
- $\boldsymbol{M}_3$ and $\boldsymbol{H}$ are collinear ($a_1 = a_2 = -n_{12}$), i.e. the component of $\boldsymbol{M}_3$ parallel to $\boldsymbol{H}$ is either M_3 or $-M_3$. We refer to these states as [nc3+] and [nc3−], respectively. In fact, [nc3+] can be subdivided in the alternatives [nc3++] and [nc3+−], with $M_t = M_3 + M_{12}$ and $M_t = M_3 - M_{12} \geqslant 0$, respectively. A subdivision of the

state [nc3−] is not necessary, because the state [nc3−−], with $M_t = -M_{12} - M_3$, would have a total moment directed opposite to $\boldsymbol{H}$. Consequently, only the state [nc3−] ≡ [nc3−+] has to be considered.

In this section, we start by determining the regions (for M_t, and for $\mu_0 H$) where the non-collinear states are stable, together with the subsidiary conditions on the molecular field constants, necessary for the stability of the non-collinear state. Rather than comparing energy expressions, we use the criteria mentioned in Section 2, i.e. $a_j \geqslant 0$ and $m_{jj} = a_{j-1}a_{j+1} - n^2_{j-1,j+1} \geqslant 0$. Moreover, it must be possible to construct $\boldsymbol{M}_t$ (with the appropriate length at the chosen field value) as the vector sum of $\boldsymbol{M}_3$ and the appropriate $\boldsymbol{M}_{12} = \boldsymbol{M}_1 + \boldsymbol{M}_2$. We treated all non-collinear states in this way. In Appendix A we list the stability ranges for all these non-collinear states, together with the effective field parameters a_j. Subsequently, we derive the stability ranges for the collinear states, very much in the same way. The stability conditions, the field regions and the effective fields for the collinear states are listed also in Appendix A.

Finally, the way to construct the possible magnetization curves is treated. The results are given in Appendix B.

Although all our actual derivations rest on stability criteria (not on comparing energies), it is instructive to write the energy in the forms

$$E = -n_{12}\boldsymbol{M}_1\boldsymbol{M}_2 - (n_{13}\boldsymbol{M}_3 + \mu_0\boldsymbol{H})\boldsymbol{M}_{12} - \mu_0\boldsymbol{H}\boldsymbol{M}_3 \tag{31a}$$

$$= -\tfrac{1}{2}n_{12}\{\boldsymbol{M}_{12} + (\mu_0\boldsymbol{H} + n_{13}\boldsymbol{M}_3)/n_{12}\}^2 + \tfrac{1}{2}n_{12}(M_1^2 + M_2^2) + \tfrac{1}{2}n_{13}M_3^2 - \tfrac{1}{2}\{(n_{12} - n_{13})/n_{12}n_{13}\}\{\mu_0\boldsymbol{H} + n_{13}\boldsymbol{M}_3\}^2 + \tfrac{1}{2}(\mu_0 H)^2/n_{13} \tag{31b}$$

$$= -\tfrac{1}{2}(n_{12} - n_{13})M_{12}^2 + \tfrac{1}{2}n_{12}(M_1^2 + M_2^2) + \tfrac{1}{2}n_{13}M_3^2 - \tfrac{1}{2}n_{13}M_t^2 - \mu_0\boldsymbol{H}\boldsymbol{M}_t \tag{31c}$$

$$= -\tfrac{1}{2}(n_{12} - n_{13})M_{12}^2 + \text{const.} - \tfrac{1}{2}n_{13}M_t^2 - \mu_0 H M_t. \tag{31d}$$

In our discussion, we apply the notation established in Section 3 (Eq. (17)). Apart from $M_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]$ (Eq. (17)), we introduce "critical fields" $B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]$, defined as

$$B_c = B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3] = -n_{12}(\varepsilon_1 M_1 + \varepsilon_2 M_2) - n_{13}\varepsilon_3 M_3. \tag{32}$$

For the derivation of the stability ranges, we have to apply a number of simple mathematical tricks. We restrict ourselves to demonstrate the application of the procedure in some detail to one non-collinear state (chosen to be the state [ncF]) and, for the collinear states, after some general derivations, also to one collinear state in particular (chosen to be the state [− + +]).

4.1. Stability ranges for the non-collinear state [ncF]

[ncF] is the non-collinear state in which $\boldsymbol{M}_1$ and $\boldsymbol{M}_2$ are oriented "Ferromagnetically", i.e. in the same direction ($M_{12} = M_1 + M_2$).

As stated above, this state may occur in case $a_3 = -n_{13}$. Since a_3 should be non-negative, we have $n_{13} < 0$ (we exclude vanishing coupling constants), hence, also $a_3 > 0$. Vectors $\boldsymbol{M}_t$ can be constructed only with a length M_t in the range from $|M_3 - (M_1 + M_2)|$ up to $(M_3 + M_1 + M_2)$, i.e. between $M_c[- - +]$ {or $M_c[+ + -]$} and $M_c[+ + +]$. From Eq. (2c) (with $a_3 = -n_{13}$ and $n_{23} = n_{13}$) we find $M_t = \mu_0 H/|n_{13}|$, or (in Eq. (3)) $Q = 1/|n_{13}|$ (hence, $Q > 0$). In this way, the range of allowed magnetization values can be established. After multiplication by $|n_{13}|$ the corresponding range of allowed fields is found.

We proceed by checking the remaining stability criteria. Substracting Eqs. (2a) and (2c) we find $a_1 M_1 = |n_{13}|(M_1 + M_2) + n_{12}M_2$. From the condition $m_{22} = |n_{13}|(a_1 - |n_{13}|) \geqslant 0$, we find $a_1 \geqslant |n_{13}|$, implying also $a_1 > 0$. The calculated value of a_1 satisfies this inequality only in case $n_{12} + |n_{13}| \geqslant 0$. Since we treat Case II, with $n_{12} \neq n_{13}$, we should demand $n_{12} + |n_{13}| > 0$ (hence, $m_{22} > 0$). The analogous reasoning with respect to a_2 yields no other limitations. Neither do the conditions $m_{11} \geqslant 0$ and $m_{33} \geqslant 0$ (actually, also $m_{11} > 0$ and $m_{33} > 0$). We omit the detailed calculations.

We treated all non-collinear states in this way. In Appendix A we list the stability ranges for all these non-collinear states, together with the effective field parameters a_j.

4.2. Stability ranges for the collinear states

We start by expressing a_j, and so on, in the parameters ε_j defined in Eq. (17):

$$a_1 = (\mu_0 H + \varepsilon_2 n_{12} M_2 + \varepsilon_3 n_{13} M_3)/\varepsilon_1 M_1; \qquad (33a)$$

$$a_2 = (\mu_0 H + \varepsilon_1 n_{12} M_1 + \varepsilon_3 n_{13} M_3)/\varepsilon_2 M_2; \qquad (33b)$$

$$a_3 = (\mu_0 H + \varepsilon_1 n_{13} M_1 + \varepsilon_2 n_{13} M_2)/\varepsilon_3 M_3. \qquad (33c)$$

Using the notation established before (Eq. (27a, b)), we find

$$a_1 + n_{12} = (\mu_0 H - B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3])/\varepsilon_1 M_1, \qquad (34)$$

$$m_{11} = \{(\mu_0 H)^2 + \mu_0 H (n_{13} M_c[\varepsilon_1, \varepsilon_2, \varepsilon_3] + \varepsilon_1 n_{12} M_1) - \varepsilon_1 n_{13} M_1 B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]\}/\varepsilon_2 M_2 \varepsilon_3 M_3; \qquad (35a)$$

$$m_{12} = \{n_{13}(\mu_0 H - B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]) + (n_{12} - n_{13})\mu_0 H\}/\varepsilon_3 M_3; \qquad (35b)$$

$$m_{13} = n_{13}(\mu_0 H - B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3])/\varepsilon_2 M_2; \qquad (35c)$$

$$m_{33} = (\mu_0 H + \varepsilon_3 n_{13} M_3) \times (\mu_0 H - B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3])/\varepsilon_1 M_1 \varepsilon_2 M_2; \qquad (35d)$$

$$DQ_3 = m_{13} + m_{23} + m_{33} = (\mu_0 H - B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]) \times (\mu_0 H + n_{13} M_c)/\varepsilon_1 M_1 \varepsilon_2 M_2; \qquad (36)$$

$$D = \mu_0 H (\mu_0 H - B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]) \times (\mu_0 H + n_{13} M_c)/\varepsilon_1 M_1 \varepsilon_2 M_2 \varepsilon_3 M_3. \qquad (37)$$

The stability criteria to be checked are, again, $D > 0$, $a_j > 0$ and $m_{jj} > 0$. A consequence of our choice $M_2 \geqslant M_1$ is, that, in the collinear states under consideration, $\boldsymbol{M}_2$ should point in the direction of $\boldsymbol{M}_{12}$.

After multiplying Eq. (2a) by a_2 and Eq. (2b) by n_{12}, and adding, we find Eq. (38a) below (using $n_{13} = n_{23}$; see also Eq. (8)). Proceeding in an analogous way, we find

$$\boldsymbol{M}_1 = (a_2 + n_{12})(\mu_0 \boldsymbol{H} + n_{13} \boldsymbol{M}_3)/m_{33}; \qquad (38a)$$

$$\boldsymbol{M}_2 = (a_1 + n_{12})(\mu_0 \boldsymbol{H} + n_{13} \boldsymbol{M}_3)/m_{33}; \qquad (38b)$$

$$\boldsymbol{M}_{12} = (a_1 + a_2 + 2n_{12})(\mu_0 \boldsymbol{H} + n_{13} \boldsymbol{M}_3)/m_{33}. \qquad (38c)$$

Eq. (38c), the addition of Eqs. (38a) and (38b), reveals that $\boldsymbol{M}_{12}$ points in the direction of the effective field $(\mu_0 \boldsymbol{H} + n_{13} \boldsymbol{M}_3)$, since $(a_1 + a_2 + 2n_{12}) > 0$ (see Eq. (11), and discussion there, in Section 2). In order to have $\boldsymbol{M}_2$ pointing in the same direction, we have to demand $a_1 + n_{12} > 0$ in Eq. (38b). This relation is useful in case $n_{12} < 0$. According to Eq. (34), we have then

$$\text{sign}(\mu_0 H - B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]) = \varepsilon_1.$$

In the same spirit, we have $\text{sign}(a_2 + n_{12}) = \varepsilon_1 \varepsilon_2$. Hence,

$$\text{sign}(\mu_0 H - B_c[\varepsilon_1, \varepsilon_2, \varepsilon_3]) = \varepsilon_1; \qquad (39a)$$

$$\text{sign}(\mu_0 H + n_{13} \varepsilon_3 M_3) = \varepsilon_2; \qquad (39b)$$

$$\text{sign}(\mu_0 H + n_{13} M_c) = \varepsilon_2 \varepsilon_3. \qquad (39c)$$

These relations (Eqs. (39a)–(39c)) should not be considered as "extra" conditions, but only as being useful in a quick derivation.

As an example of the detailed analysis of the collinear states, we show here the application of the inequalities in the analysis of the collinear state $[-++]$:

$$M_t = M_c[-++] = -M_1 + M_2 + M_3,$$

$$\varepsilon_1 = -1, \quad \varepsilon_2 = \varepsilon_3 = +1.$$

Since $\boldsymbol{M}_1$ is opposite to $\boldsymbol{M}_{12}$, we have $a_2 + n_{12} < 0$ (from Eq. (38a)), so necessarily $n_{12} < 0$. From Eq. (39a), we infer $(\mu_0 H - B_c[-++]) < 0$, hence, $\mu_0 H < B_c[-++]$.

Consequently, the state can only be stable in case $B_c[-++] > 0$, i.e. $n_{13} M_3 < |n_{12}|(M_2 - M_1)$. This limitation is only important in case $n_{13} > 0$, and then, for instance excludes the case $M_2 = M_1$. We treat the cases $n_{12} < 0$, $n_{13} > 0$ (Case AF) and $n_{12} < 0$, $n_{13} < 0$ (Case AA) separately.

Case AF: $n_{12} < 0$, $n_{13} > 0$.

The field range (and stability conditions) can be written as

$$0 < \mu_0 H < -n_{13}M_3 + |n_{12}|(M_2 - M_1) = B_c[- + +] > 0.$$

It is easy to show, that, in the field range indicated, the conditions $a_j > 0$ ($j = 1,2,3$) and $m_{33} > 0$ are satisfied.

As a last step we investigate the sign of m_{22} and m_{33}. From Eq. (35a) (exchanging the indices 1 and 2) we find, irrespective of the sign of n_{13}:

$$M_1 M_3 m_{22} = -(\mu_0 H)^2 + \mu_0 H\{|n_{12}|M_2 - n_{13}M_c\} + n_{13}M_2 B_c[-,+,+].$$

At the limiting values $\mu_0 H = 0$ and $\mu_0 H = B_c[-,+,+]$, we find

$$M_1 M_3 m_{22} = n_{13} M_2 B_c[-,+,+] > 0$$

and

$$M_1 M_3 m_{22} = (|n_{12}| + n_{13}) M_2 B_c[-,+,+] > 0,$$

respectively. Hence, since m_{22} has only one contingent positive region (being a maximum parabola as a function of H), it must be positive in the complete interval. m_{11} is equally positive at $H = 0$, with positive slope $n_{13}M_c + |n_{12}|M_1$. So, we are at the right-hand side of the minimum, where m_{11} is a monotonously increasing function of H. Hence, it must be positive for all positive field values.

Case AA: $n_{12} < 0$, $n_{13} < 0$.

For negative n_{13} and $\varepsilon_2 = \varepsilon_3 = +1$, the inequality Eq. (39c) is stronger than (implies) inequality Eq. (39b). Consequently, $\mu_0 H > |n_{13}|M_c[-,+,+]$. This leads to

$$|n_{13}|M_3 + |n_{13}|(M_2 - M_1) = |n_{13}|M_c[-,+,+] < \mu_0 H < |n_{13}|M_3 + |n_{12}|(M_2 - M_1) = B_c[-,+,+].$$

We see, that this field range exists only in case $M_2 - M_1 > 0$ (excluding $M_2 = M_1$), and $|n_{13}| < |n_{12}|$, implying $n_{12} < n_{13} < 0$ (to be referred to as Case AA<).

It is, again, easy to show, that, in the field range indicated, the conditions $a_j > 0$ ($j = 1,2,3$) and $m_{33} > 0$ are satisfied.

In order to check $m_{22} > 0$, we consider again the end-points (see above). At the left end-point, $\mu_0 H = |n_{13}| M_c[- + +]$, we find $m_{22} = |n_{13}|(|n_{12}| - |n_{13}|) > 0$, and at the right end-point, again, $M_1 M_3 m_{22} = (|n_{12}| - |n_{13}|) M_2 B_c[-,+,+] > 0$, apparently correct under the same extra subsidiary condition $n_{12} < n_{13}$ (< 0). In the same way as above, we conclude that m_{22} is positive in the complete interval.

m_{11} equals m_{22} at the left end-point (exchange indices 1 and 2), and possesses a positive slope $|n_{13}|M_c + |n_{12}|M_1$, so must be positive in the complete field interval.

The stability conditions, the field regions and the effective fields for the collinear states are listed also in Appendix A.

We are now in a position to construct the magnetic phase diagrams and the magnetization curves, for any set of parameters satisfying $n_{12} \neq n_{23} = n_{13}$.

In Appendix B we give the complete list of all possible magnetization curves. These curves appear to consist of a sequence of constant and linear portions. This property is the basis of a shorthand type definition, explained in Appendix B.

As an example we treat Case IIAA<, i.e. $n_{12} < n_{13} < 0$. More precisely, we only discuss the situation in case $M_3 \geqslant M_1 + M_2; M_2 > M_1$.

We start by listing the critical fields, in decreasing order. These fields are read from the detailed description of the different cases in Appendix A.

$$B_c[+ + +] = |n_{12}|(M_2 + M_1) + |n_{13}|M_3 \quad (> 0);$$

$$B_c[- + +] = |n_{12}|(M_2 - M_1) + |n_{13}|M_3 \quad (> 0);$$

$$|n_{13}|M_c[- + +] = |n_{13}|(M_2 - M_1) + |n_{13}|M_3 \quad (> 0);$$

$$|n_{13}|M_c[+ - +] = |n_{13}|M_3 - |n_{13}|(M_2 - M_1) \quad (> 0);$$

$$B_c[+ - +] = |n_{13}|M_3 - |n_{12}|(M_2 - M_1);$$

$$B_c[- - +] = |n_{13}|M_3 - |n_{12}|(M_2 + M_1).$$

Here, we indicated that, in the present situation, only the fields $B_c[+ - +]$ and $B_c[- - +]$ may adopt negative values. That means, that we have to distinguish the following possibilities. In each case, the stable structure is given, in an obvious notation, between the appropriate "critical" field values.

Case (a): $B_c[+-+]\leqslant 0$.

$$\mu_0 H: \quad 0 < [+-+] < |n_{13}|M_c[+-+] < [\text{ncA}] < |n_{13}|M_c[-++] < [-++] < B_c[-++] < [\text{nc3}++] < B_c[+++] < [+++];$$

$$M_t: \quad M_3-(M_2-M_1) < M_t = \mu_0 H/|n_{13}| < M_3+(M_2-M_1) < M_t = M_3(1-|n_{13}|/|n_{12}|)+\mu_0 H/|n_{12}| < M_3+M_2+M_1;$$

Type: CLCN$^+$C.

Case (b): $B_c[--+]\leqslant 0 < B_c[+-+]$.

$$\mu_0 H: \quad 0 < [\text{nc3}+-] < B_c[+-+] < [+-+]$$

(and so on, see Case (a));

$$M_t: \quad M_3(1-|n_{13}|/|n_{12}|)+\mu_0 H/|n_{12}| < M_3-(M_2-M_1)$$

(and so on, see Case (a));

Type: N$^+$CLCN$^+$C; N$^+$ parts extrapolated form one straight line.

Case (c): $0 < B_c[--+] < B_c[+-+]$.

We infer from the condition $B_c[--+] > 0$, that $M_3 > (|n_{12}|/|n_{13}|)(M_2+M_1)$. Since $(|n_{12}|/|n_{13}|) > 1$ (see above), we have now also the restriction on the moments:

$$M_3 > (M_2+M_1), \quad \text{excluding } M_3 = (M_2+M_1).$$

$$\mu_0 H: \quad 0 < [--+] < B_c[--+] < [\text{nc3}+-]$$

(and so on, see Case (b));

$$M_t: \quad M_3-(M_2+M_1) < M_3(1-|n_{13}|/|n_{12}|)+\mu_0 H/|n_{12}|$$

(and so on, see Case (b));

Type: CN$^+$CLCN$^+$C; N$^+$ parts extrapolated form one straight line.

An example of this very interesting type of magnetization curve (Case (c)) is given in Fig. 2. The dashed line shows that the extrapolated N$^+$ parts do form one straight line, indeed. The other types (results from Case (b) and Case (a)) can be imagined by shifting the origin diagonally upwards (until the [ncA] phase is reached).

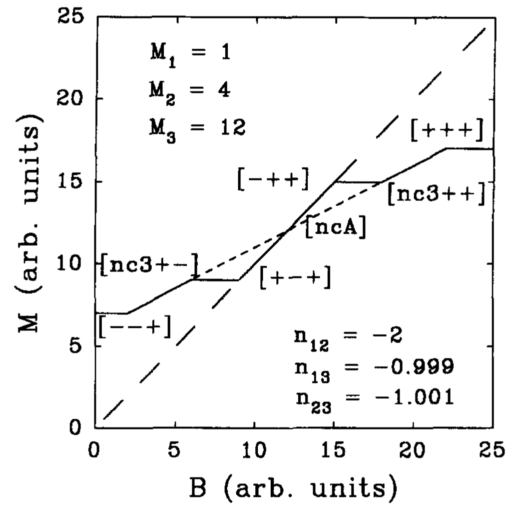

Fig. 2. Type CN$^+$CLCN$^+$C in Case IIAA<c, with $M_3 > M_1+M_2$; $M_1 \neq M_2$. The small difference betweeen n_{13} and n_{23} can be ignored here. Types N$^+$CLCN$^+$C, CLCN$^+$C and LCN$^+$C can be obtained by decreasing M_3 until $M_3 = M_2 - M_1$, i.e. by shifting the origin diagonally upwards, until [ncA] is reached.

For Case II, all magnetic phase diagrams have been investigated (see Appendix A and B). The results are listed in Table 1. i.e. for all possible types of magnetization curve, the possible concurrent configurations are given. Some examples of magnetization curves are given in Fig. 1 (already discussed in Section 3), Fig. 2 (discussed above), Fig. 3 (see Remark 2), and Fig. 4.

We conclude this section by making two remarks:

Remark 1. In the end-points of the stability regions, the criteria for a local minimum are no longer fulfilled. This means, in general, that a second derivative of the energy vanishes, or rather, changes sign, signalling the transition to another stability region. In the above example, and indeed quite generally (see Appendix B), we find, that only one structure is stable given a certain set of n_{ij}-parameters and magnetic-moment values, and that all transitions to another regime are of second order. Indeed, jumps in the calculated magnetization curves do not occur. Apart from that, we never have to compare the energies of two competing

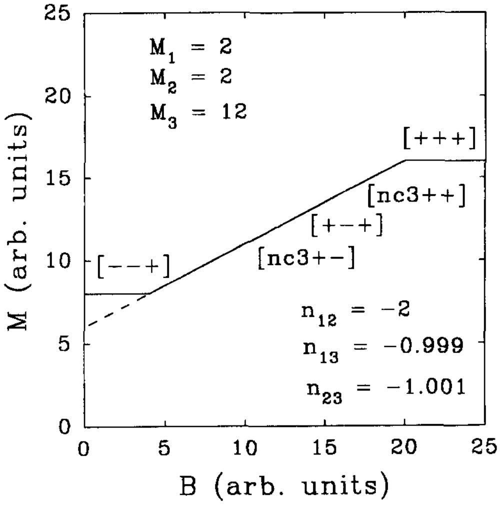

Fig. 3. Type CNC in Case IIAA<c, with $M_1 = M_2$; $M_3 > 2M_1$. The small difference between n_{13} and n_{23} allows the computer program to identify the transition from [nc3 + −] and [nc3 + +] as (a remnant of) the collinear structure [+ − +] (see Remark 2, end of Section 4).

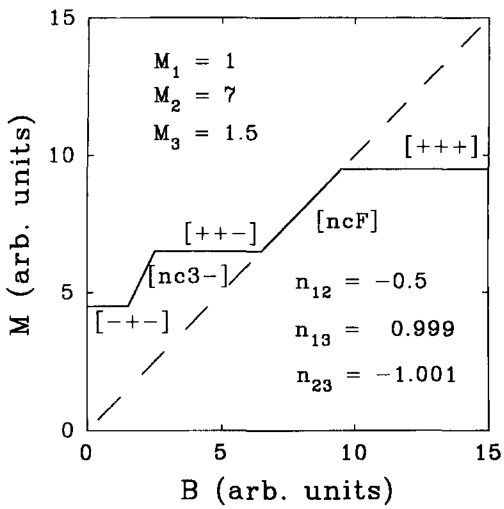

Fig. 4. Type CN⁻CLC in Case IIAA>c, with $M_3 < M_2 - M_1$. The small difference between n_{13} and n_{23} can be ignored here.

structures: the investigation of the stability appears to be sufficient.

Remark 2. In the case that $M_1 = M_2$, and $n_{12} < n_{13} < 0$ (Case IIAA<), in our formalism, strictly speaking, the collinear states [− + +] or [+ − +] cannot be stable, whereas, in general, the non-collinear states [nc3 + +] or [nc3 + −] can be stable. Nevertheless, in case, incidentally, $\mu_0 \boldsymbol{H} + n_{13}\boldsymbol{M}_3$ vanishes, so $\mu_0 H = |n_{13}|M_3$, we have rotational freedom (keeping $M_{12} = 0$), and we have a choice to consider this state as a non-collinear [nc3+] state or a collinear one ([− + +] or [+ − +]). We prefer to consider this situation as the transition point (field value) between the region where [nc3 + +] is stable and the region where [nc3 + −] is. Comparing this situation with the example given in Fig. 2, we see that the fields B_c[− + +] down to B_c[+ − +] coincide. The result is that the N^+ parts are joined, i.e. at the transition point, $\mu_0 H = |n_{13}|M_3$, the part [nc3 + −] is joined to the part [nc3 + +]. An example is given in Fig. 3. The applied computer program (see Section 5) introduces a small difference between n_{13} and n_{23}, strictly speaking not quite according to Case II under discussion here. Nevertheless, we see that the small difference can be ignored in Fig. 2, but here forces the programme to identify the transition, in this case, as a remnant of the collinear phase [+ − +].

5. Case III: three different molecular-field constants ($n_{12} \neq n_{23} \neq n_{13} \neq n_{12}$)

In this case, there is no reason to impose any subsidiary condition on M_1, M_2 and M_3. We start by investigating the stability of the non-collinear structures, then that of the collinear states. Finally, the way to construct the magnetization curves is discussed.

5.1. Non-collinear structures

In Section 2, we derived already that in the non-collinear phases, the Eqs. (2a)–(2c) are pair-wise independent, whereas the determinant does vanish. Taking into account also the right-hand side of these equations, we conclude that there must be, for instance, a real, non-zero, number α, such that Eq. (2b) plus α times Eq. (2a) yields $(1 + \alpha)$ times Eq. (2c). Hence, the coefficients a_j and the minors m_{ij} can be expressed as functions of the single parameter α:

$$a_1 = -n_{13} + (n_{12} - n_{13})/\alpha; \tag{40a}$$

$$a_2 = -n_{23} + (n_{12} - n_{23})\alpha; \tag{40b}$$

$$a_3 = -(\alpha n_{13} + n_{23})/(1 + \alpha); \tag{40c}$$

$$m_{13} = n_{23}(n_{12} - n_{13}) + n_{13}(n_{12} - n_{23})\alpha; \tag{41a}$$

$$m_{23} = m_{13}/\alpha; \tag{41b}$$

$$m_{33} = -(m_{13} + m_{23}) = -(1 + 1/\alpha)m_{13}. \tag{41c}$$

For any suitable α value, the non-collinear state can be constructed in the following way. By subtracting Eq. (2b) from Eq. (2a) we obtain

$$(a_1 + n_{12})\boldsymbol{M}_1 - (a_2 + n_{12})\boldsymbol{M}_2 + (n_{23} - n_{13})\boldsymbol{M}_3 = 0. \tag{42}$$

Inserting the functions of α given above, we find

$$(n_{12} - n_{13})(1 + 1/\alpha)\boldsymbol{M}_1 - (n_{12} - n_{23})(1 + \alpha)\boldsymbol{M}_2 + (n_{23} - n_{13})\boldsymbol{M}_3 = 0. \tag{43}$$

In other words, a necessary condition for the existence of the non-collinear state (corresponding to the chosen α value) is, that a triangle can be constructed with sides of length $|(n_{12} - n_{13})(1 + 1/\alpha)M_1|$, $|(n_{12} - n_{23})(1 + \alpha)M_2|$ and $|(n_{23} - n_{13})\boldsymbol{M}_3|$, respectively. Having constructed the triangle, we can derive the relative orientations of the magnetic moment vectors, as well as that of the total moment. The appropriate field value can now be calculated by applying any one of the Eqs. (2a)–(2c). One may imagine that the constructed triangle is rotated in such a way that the total magnetic moment vector points in the direction of the applied field.

We remark here, that for zero applied field the parameter α must have the value

$$\alpha = (n_{23}/n_{13})(n_{12} - n_{13})/(n_{23} - n_{12}). \tag{44}$$

For this α value, the relations for a_j given in Section 2 (Eq. (9)) are easily verified. Of course, the choice of α should be restricted in such a way that the stability criteria are fulfilled ($a_j \geqslant 0$, $m_{jj} \geqslant 0$). The quantities a_j happen to be monotonous functions of α (see Eqs. (40a)–(40c)). Consequently, for any contingent region of α values, only the end-points have to be investigated. With regard to the minors m_{jj}, we infer from Eq. (11), and the related discussion in Section 2, that, for finite fields H, m_{jj} cannot change sign as long as M_t remains finite. Since the M_t value is limited ($M_t \leqslant M_1 + M_2 + M_3$), and since the quantities m_{jj} are continuous functions of α, also for the minors an investigation of the end-points suffices. This statement remains true in case one of the end-points happens to be the zero-field value given in Eq. (44) (where m_{jj} vanishes). For finite fields, it is obvious, from a physical point of view, that the end-points must correspond to collinear structures, being the limiting triangles in the construction discussed above. Moreover, just as we found in Case II (preceding section), the corresponding field value must be a "critical" field for the collinear structure (for that field value, the determinant vanishes). We refrained from searching for a concise, mathematically sound, proof for these physically plausible properties. We return to this question after a discussion of the stability of the collinear structures.

5.2. Collinear structures

In this general case, we used a computer program to find the stability regions for the collinear structures. For a given set of molecular-field constants n_{ij} and magnetic moments M_1, M_2 and M_3, the program does calculate the field values at which the determinant D, or rather $DQ_1 = m_{11} + m_{12} + m_{13}$ vanishes (see Eq. (6b)). In a comparison with Case II (Section 4), one should consider the relations given by Eqs. (36) and (37). Notice, that in a collinear structure, Eqs. (2a)–(2c) just define the quantities a_j (and so on) as functions of H.

As a next step, the stability criteria are checked at these field values. Non-stable situations are rejected, the stable ones yield the actual "critical" fields. In view of the discussion of the non-collinear phases above, the α values in these end-points are calculated. The program does order the critical fields according to their magnitude. In all cases investigated, we find that the higher critical fields do correspond to collinear structures with higher total magnetic-moment values. Moreover, at any field value we find at most one stable collinear structure (Case II, preceding section, exhibits the same property). Again, whereas for Case II we did investigate all possibilities, in this more general Case III, we did not bother to look for a general, mathematically sound, proof.

5.3. Magnetization curves

From a physical point of view, we expect that (in the non-collinear structures) the total magnetic moment will increase with increasing applied field. So, we expect that, in this general case, the magnetization curves do resemble those found in Case II: constant parts (collinear structures), connected by – in the general case possibly curved – parts corresponding to the non-collinear phases. In fact, the program does calculate the full curves. The magnetization curves for the non-collinear phases are calculated by taking a large number of α values, interpolated between the values found for the end-points, i.e. at the "critical fields". For each α value, the triangle construction (Eq. (43)) is applied in a calculation of the total magnetic moment and the corresponding field value, in the manner discussed above. The resulting curves do resemble those found in Case II, indeed. Actually, all examples in this article were treated as if they belonged to Case III, just by introducing small deviations between n_{13} and n_{23}, if necessary (see Figs. 1–4). In general, strong curvatures can occur, especially in cases where, in a comparison to Case II, an intermediate collinear structure cannot be reached completely. An example is presented in Fig. 5.

In case, at vanishing applied field, a non-collinear phase does exist (see Fig. 6), the program takes the α value from Eq. (44), and calculates the total moment, again using the triangle construction discussed above. In the example of Fig. 6, we have $\alpha = 0.5$, yielding a total moment $M_t(0) = 7.75$. These zero-field values are taken to be the starting point for the interpolation procedure, up to the first "critical field" for a collinear structure.

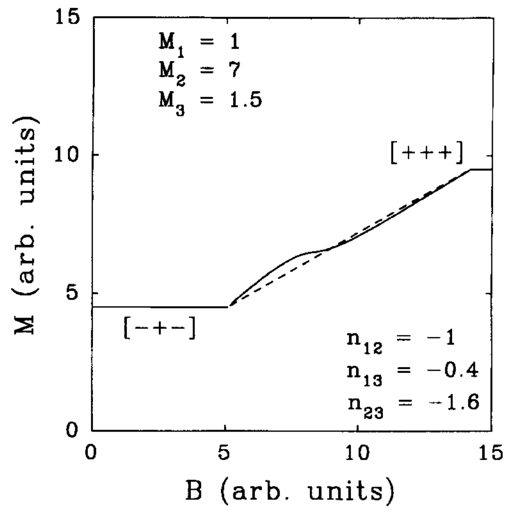

Fig. 5. An example of a magnetization curve in Case III, with three different negative (antiferromagnetic) molecular-field constants. The strong variation of the curvature indicates that a collinear structure (i.e. [+ + −]) is approached, but not formed. The dashed curve is a guide to the eye.

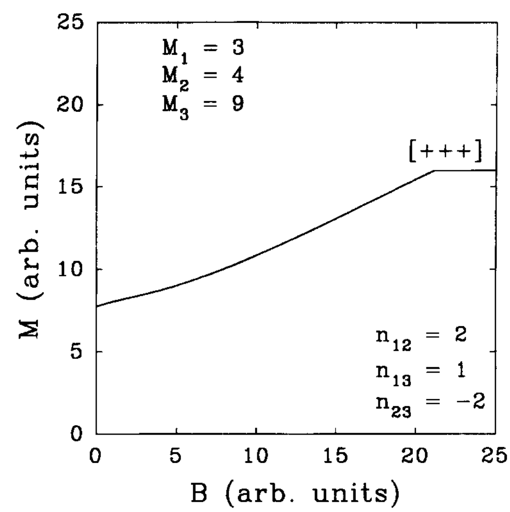

Fig. 6. An example of a magnetization curve in Case III, with two positive (ferromagnetic) molecular-field constants and a negative one. Noticie that at vanishing field a non-collinear phase does exist.

6. Concluding remarks

Let us start by remarking that the present "three-sublattice model without anisotropy" does contain all the results of the "two-sublattice model without anisotropy" (as a part of Case II) and also of the "symmetric three-sublattice model without anisotropy" (considering $M_1 = M_2$ in Case II). In this sense, the present work does extend the work of Colpa et al. [6], in particular by discussing more precisely the stability criteria. It is perhaps worthwhile to mention that the computer program (discussed in Section 5) can be applied to all cases, inclusive of the (pseudo) two-sublattice model. Moreover, we want to stress

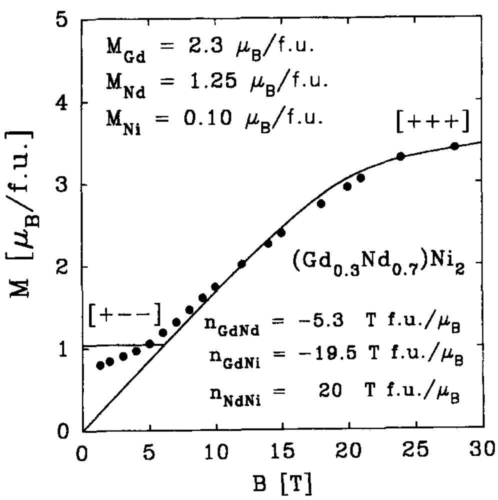

Fig. 7. A trial to explain the curvature in the magnetization curve as observed in free powder measurements on (Nd,Gd)Ni_2; see Ref. [8] for more details.

the fact that, in both models, no first order transitions (no jumps) do occur, and that, at any applied field, only one stable structure does exist. Comparison of energies of two stable configurations is not necessary.

With respect to actual applications, we may stress that the type of the magnetization curve does not change by scaling up all the magnetic moments by the same factor, neither does scaling up of the molecular-field constants. In both cases, the (critical) fields are scaled up with the same factor.

We have shown that in the "three-sublattice model without anisotropy" rather complicated magnetization curves may occur. One should bear in mind, however, that in the "two-sublattice model" similar curves can be expected in case the crystalline anisotropy cannot be neglected (see Zhao Zhi-gang [7]). Nevertheless, we expect the model to be useful in the determination of the molecular-field constants, i.e. the relevant exchange interactions, in a number of applications. Indeed, one of the motivations to undertake this analysis was to investigate the possibility to determine the (ferromagnetic) RT-interaction between a transition metal T and a *light* rare earth R, just by placing on one sublattice a mixture of light and heavy rare-earth atoms (preferably Gd, in order to minimize anisotropy complications). Some preliminary experiments do confirm the feasibility of this approach [8]. As an example, we show the expected kind of results for e.g. (Nd, Gd)Ni_2 in Fig. 7. Moreover, it should be possible to apply and test the model by preparing suitable multilayer samples.

An obvious, and desirable, extension of the model is to include the field dependence of the magnetic moments. In fact, in the preliminary experiments mentioned, the magnetic moment in the collinear phase(s) often appears to be field dependent, even at temperatures well below the ordering temperature. Including (free) energy terms $F_j(M_j)$ in Eq. (1), we see that the equilibrium conditions as represented by Eqs. (2a)–(2c), remain valid, the quantities a_j now being functions of (the variable) M_j. These functions, in principle, can be determined experimentally. So, the extension of the model seems to be straightforward. At the moment we restrict ourselves to two remarks. Whereas in the "two-sublattice model", in a non-collinear phase, the effective field acting on a sublattice magnetization is strictly constant ($a_j \boldsymbol{M}_j = |n|\boldsymbol{M}_j$), in the "three-sublattice model" the effective field can vary. Consequently, in the "two-sublattice model" the sublattice magnetization remains constant, whereas in the "three-sublattice model" the magnetic moments can change, possibly given rise to even more complicated magnetization curves. Nevertheless, as a second remark, it is straightforward to show that in case the sublattices possess linear magnetization curves (field-independent high-field susceptibility, for instance), the system is equivalent to a system with constant magnetic moments, possibly even to a two-sublattice system. That means, that the present model can be applied to a large number of actual systems, provided the influence of crystalline anisotropy can be circumvented or ignored.

Acknowledgements

The author wants to thank Dr. J.H.P. Colpa for critically reading a first version of this article. In case any mathematical inconsistency did survive, it is the author who has to bear the blame.

Appendix A: Stability ranges and effective fields in Case II ($n_{12} \neq n_{13} = n_{23}$)

A.1. Notation

The sign of n_{12} is indicated by $F(>0)$ or $A(<0)$; consecutively, the sign of n_{13} is indicated in the same way. The condition $n_{12} > [<] n_{13}$ is indicated by a trailing $>$ [$<$]. In this way, we distinguish Case AA> and Case AA<. Moreover, in order to stress the importance of the signs, we write $|n_{12}|$ and $|n_{13}|$ rather than $-n_{12}$ and $-n_{13}$, respectively, in case non-negative values of the coupling parameter are excluded. The notation of the states themselves is explained in the beginning of Section 4.

A.2. Non-collinear states

[ncF]: Only in case $n_{13} < 0$ and $n_{12} - n_{13} > 0$ (Cases IIFA and IIAA>).

$$|M_3 - (M_2 + M_1)| < M_t = \mu_0 H/|n_{13}| < M_3 + (M_2 + M_1);$$

$$|n_{13}||M_3 - (M_2 + M_1)| < \mu_0 H < |n_{13}|(M_3 + (M_2 + M_1));$$

or, equivalently,

$$|n_{13}|M_c[--+] < \mu_0 H < |n_{13}|M_c[+++] \quad M_3 > M_2 + M_1;$$

$$|n_{13}|M_c[++-] < \mu_0 H < |n_{13}|M_c[+++] \quad M_3 < M_2 + M_1;$$

$$0 < \mu_0 H < |n_{13}|M_c[+++] \quad M_3 = M_2 + M_1;$$

$$a_1 M_1 = |n_{13}|(M_1 + M_2) + n_{12} M_2;$$

$a_2 M_2$ analogous;

$$a_3 M_3 = |n_{13}| M_3.$$

Remark. Notice that the effective fields do not depend on the applied field, a pseudo-two-sublattice model feature.

[ncA]: Only in case $n_{12} < n_{13} < 0$, implying $n_{12} - n_{13} < 0$ (Case IIAA<).

$$|M_3 - (M_2 - M_1)| < M_t = \mu_0 H/|n_{13}| < M_3 + (M_2 - M_1) = M_c[-++];$$

$$|n_{13}||M_3 - (M_2 - M_1)| < \mu_0 H < |n_{13}|\{M_3 + (M_2 - M_1)\};$$

or

$$|n_{13}|M_c[+-+] < \mu_0 H < |n_{13}|M_c[-++] \quad M_3 > M_2 - M_1;$$

$$|n_{13}|M_c[-+-] < \mu_0 H < |n_{13}|M_c[-++] \quad M_3 < M_2 - M_1;$$

$$a_1 M_1 = -|n_{13}|(M_2 - M_1) + |n_{12}| M_2 = |n_{13}| M_1 + (|n_{12}| - |n_{13}|) M_2;$$

$a_2 M_2$ analogous;

$$a_3 M_3 = |n_{13}| M_3.$$

Remarks. (1) This state does not exist in case $M_1 = M_2$ (see also [nc3+−]).

(2) Again, the effective fields do not depend on the applied field.

[nc3++]: Only in case $n_{12} < 0$ and $n_{12} - n_{13} < 0$ (Cases IIAF and IIAA<). Moreover $B_c[+++] = |n_{12}|(M_2 + M_1) - n_{13} M_3 > 0$ (otherwise already "stretched" at zero field).

$$\max\{M_3(1 + n_{13}/|n_{12}|), M_3 + M_2 - M_1\} < M_t = M_3(1 + n_{13}/|n_{12}|) + \mu_0 H/|n_{12}| < M_3 + M_2 + M_1;$$

$$\max\{0, -n_{13} M_3 + |n_{12}|(M_2 - M_1)\} < \mu_0 H < -n_{13} M_3 + |n_{12}|(M_2 + M_1);$$

or

$$\max\{0, B_c[-++]\} < \mu_0 H < B_c[+++];$$

$$a_1 = a_2 = |n_{12}|;$$

$$a_3 M_3 = [\mu_0 H(|n_{12}| + n_{13}) + n_{13}^2 M_3]/|n_{12}|.$$

[nc3+−]: Only in case $n_{12} < n_{13} < 0$, implying $n_{12} - n_{13} < 0$ (Case IIAA<). Moreover

$B_c[+-+] = |n_{13}|M_3 - |n_{12}|(M_2 - M_1) > 0.$

$$\max\{M_3(1 - |n_{13}|/|n_{12}|), M_3 - M_2 - M_1\} < M_t = M_3(1 - |n_{13}|/|n_{12}|) + \mu_0 H/|n_{12}| < M_3 - M_2 + M_1;$$

$$\max\{0, |n_{13}|M_3 - |n_{12}|(M_2 + M_1)\} < \mu_0 H < |n_{13}|M_3 - |n_{12}|(M_2 - M_1),$$

or

$$\max\{0, B_c[--+]\} < \mu_0 H < B_c[+-+],$$

a_1, a_2 and a_3: see [nc3++]:

$$a_1 = a_2 = |n_{12}|;$$

$$a_3 M_3 = [\mu_0 H(|n_{12}| - |n_{13}|) + n_{13}^2 M_3]/|n_{12}|.$$

Remark. In case $M_1 = M_2$, the regions [nc3++] and [nc3+−] can be joined.

[nc3−]: Only in case $n_{13} < n_{12} < 0$, implying $n_{12} - n_{13} > 0$ (Case IIAA>). Moreover $B_c[++-] = |n_{12}|(M_2 + M_1) - |n_{13}|M_3 > 0$.

$$\max\{M_3(|n_{13}|/|n_{12}| - 1), M_2 - M_1 - M_3\} < M_t = M_3(|n_{13}|/|n_{12}| - 1) + \mu_0 H/|n_{12}| < M_2 + M_1 - M_3,$$

$$\max\{0, -|n_{13}|M_3 + |n_{12}|(M_2 - M_1)\} < \mu_0 H < -|n_{13}|M_3 + |n_{12}|(M_2 + M_1),$$

or

$$\max\{0, B_c[-+-]\} < \mu_0 H < B_c[++-],$$

a_1 and a_2: see [nc3++]:

$$a_1 = a_2 = |n_{12}|;$$

$$a_3 M_3 = \{\mu_0 H(|n_{13}| - |n_{12}|) + n_{13}^2 M_3\}/|n_{12}|.$$

A.3. Collinear states

[+++]: no extra conditions, always stable at high enough field.

$$M_t = M_c[+++] = M_1 + M_2 + M_3$$

Case IIFF: $n_{12} > 0; n_{13} = n_{23} > 0.$

$\mu_0 H > 0;$

$a_1 M_1 = \mu_0 H + n_{12} M_2 + n_{13} M_3;$

a_2 analogous;

$a_3 M_3 = \mu_0 H + n_{13}(M_1 + M_2).$

Case IIAF: $n_{12} < 0; n_{13} = n_{23} > 0.$

$\mu_0 H > \max\{|n_{12}|(M_1 + M_2) - n_{13} M_3, 0\} = \max\{B_c[+++], 0\};$

$a_1 M_1 = \mu_0 H - |n_{12}|M_2 + n_{13} M_3;$

a_2 analogous;

a_3: see IIFF.

Case IIFA: $n_{12} > 0; n_{13} = n_{23} < 0.$

$\mu_0 H > |n_{13}|(M_1 + M_2 + M_3) = |n_{13}|M_c[+++];$

$a_1 M_1 = \mu_0 H + n_{12} M_2 - |n_{13}|M_3;$

a_2 analogous;

$a_3 M_3 = \mu_0 H - |n_{13}|(M_1 + M_2).$

Case IIAA>: $n_{12} < 0; n_{13} = n_{23} < 0; n_{12} > n_{13}.$

$\mu_0 H > |n_{13}|(M_1 + M_2 + M_3) = |n_{13}|M_c[+++];$

$a_1 M_1 = \mu_0 H - |n_{12}|M_2 - |n_{13}|M_3;$

a_2 analogous;

a_3: see IIFA.

Case IIAA<: $n_{12} < 0; n_{13} = n_{23} < 0; n_{12} < n_{13}.$

$\mu_0 H > |n_{12}|(M_1 + M_2) + |n_{13}|M_3) = B_c[+++];$

a_1, a_2 and a_3: see IIAA>.

[−++]: Only in case $n_{12} < 0$, and $|n_{12}| + n_{13} > 0$. Moreover $M_2 > M_1$ (excluding $M_2 = M_1$).

$$M_t = M_c[-++] = -M_1 + M_2 + M_3.$$

Case IIAF: $n_{12} < 0$; $n_{13} > 0$. Only in case $B_c[-++] = -n_{13}M_3 + |n_{12}|(M_2 - M_1) > 0$.

$$0 < \mu_0 H < -n_{13}M_3 + |n_{12}|(M_2 - M_1)$$
$$= B_c[-++];$$
$$a_1 M_1 = -\mu_0 H + |n_{12}|M_2 - n_{13}M_3;$$
$$a_2 M_2 = \mu_0 H + |n_{12}|M_1 + n_{13}M_3;$$
$$a_3 M_3 = \mu_0 H + n_{13}(M_2 - M_1).$$

Case IIAA<: $n_{12} < 0; n_{13} = n_{23} < 0; n_{12} < n_{13}$.

$$|n_{13}|M_c[-++] = |n_{13}|M_3 + |n_{13}|(M_2 - M_1) < \mu_0 H$$
$$< |n_{13}|M_3 + |n_{12}|(M_2 - M_1)$$
$$= B_c[-,+,+];$$
$$a_1 M_1 = -\mu_0 H + |n_{12}|M_2 + |n_{13}|M_3;$$
$$a_2 M_2 = \mu_0 H + |n_{12}|M_1 - |n_{13}|M_3;$$
$$a_3 M_3 = \mu_0 H - |n_{13}|(M_2 - M_1).$$

[+−+]: Only in case $n_{12} < n_{13} < 0$ (Case IIAA< only). Moreover: $M_3 > (M_2 - M_1) > 0$ (excluding $M_2 = M_1$).

$$M_t = M_c[+-+] = M_1 - M_2 + M_3.$$

Case IIAA<: $n_{12} < 0; n_{13} = n_{23} < 0; n_{12} < n_{13}$.

$$\max\{0, B_c[+-+]\} = |n_{13}|M_3 - |n_{12}|(M_2 - M_1)\}$$
$$< \mu_0 H < |n_{13}|M_3 - |n_{13}|(M_2 - M_1);$$
$$a_1 M_1 = \mu_0 H + |n_{12}|M_2 - |n_{13}|M_3;$$
$$a_2 M_2 = -\mu_0 H + |n_{12}|M_1 + |n_{13}|M_3;$$
$$a_3 M_3 = \mu_0 H + |n_{13}|(M_2 - M_1).$$

[++−]: Only in case $n_{13} < 0$, and $|n_{13}| + n_{12} > 0$ (Cases IIFA and IIAA>). Moreover $M_1 + M_2 > M_3$.

$$M_t = M_c[++-] = M_1 + M_2 - M_3.$$

Case IIFA: $n_{12} > 0; n_{13} = n_{23} < 0$.

$$0 < \mu_0 H < |n_{13}|(M_1 + M_2 - M_3)$$
$$= |n_{13}|M_c[++-];$$
$$a_1 M_1 = \mu_0 H + n_{12}M_2 + |n_{13}|M_3;$$

a_2 analogous;

$$a_3 M_3 = -\mu_0 H + |n_{13}|(M_1 + M_2).$$

Case IIAA>: $n_{12} < 0; n_{13} = n_{23} < 0; n_{12} > n_{13}$. If $B_c[++-](= |n_{12}|(M_2 + M_1) - |n_{13}|M_3) < 0$, then see Case IIFA (with $n_{12} = -|n_{12}|$), else:

$$B_c[++-] < \mu_0 H < |n_{13}|(M_1 + M_2) - |n_{13}|M_3)$$
$$= |n_{13}|M_c[++-];$$
$$a_1 M_1 = \mu_0 H - |n_{12}|M_2 + |n_{13}|M_3;$$

a_2 analogous;

a_3: see IIFA.

[−−+]: Only in case $n_{13} < 0$ (Cases IIFA, IIAA>, IIAA<). Moreover $M_3 > (M_1 + M_2)$.

$$M_t = M_c[--+] = -M_1 - M_2 + M_3$$

Case IIFA: $n_{12} > 0; n_{13} = n_{23} < 0$.

$$0 < \mu_0 H < |n_{13}|(M_3 - M_1 - M_2)$$
$$= |n_{13}|M_c[--+];$$
$$a_1 M_1 = -\mu_0 H + n_{12}M_2 + |n_{13}|M_3;$$

a_2 analogous;

$$a_3 M_3 = \mu_0 H + |n_{13}|(M_1 + M_2).$$

Case IIAA>: $n_{12} < 0; n_{13} = n_{23} < 0; n_{12} > n_{13}$.

$\mu_0 H$: see IIFA;

$$a_1 M_1 = -\mu_0 H - |n_{12}|M_2 + |n_{13}|M_3;$$

a_2 analogous;

a_3: see IIFA.

Case IIAA<: $n_{12} < 0; n_{13} = n_{23} < 0; n_{12} < n_{13}$. Only in case $B_c[--+] = |n_{13}|M_3 - |n_{12}|(M_1 + M_2) > 0$

$$0 < \mu_0 H < B_c[--+];$$

a_1, a_2 and a_3: see IIAA>.

[−+−]: Only in case $n_{13} < 0$ and $n_{12} < 0$ (Cases IIAA> and IIAA<). Moreover $M_2 - M_1 > M_3$ (excluding $M_2 = M_1$).

$$M_t = M_c[-+-] = -M_1 + M_2 - M_3.$$
$$a_1 M_1 = -\mu_0 H + |n_{12}| M_2 - |n_{13}| M_3;$$
$$a_2 M_2 = \mu_0 H + |n_{12}| M_1 + |n_{13}| M_3;$$
$$a_3 M_3 = -\mu_0 H + |n_{13}|(M_2 - M_1).$$

Case IIAA<: $n_{12} < 0; n_{13} = n_{23} < 0; n_{12} < n_{13}$.

$$0 < \mu_0 H < |n_{13}|(M_2 - M_1) - |n_{13}| M_3 = |n_{13}| M_c[-+-].$$

Case IIAA>: $n_{12} < 0; n_{13} = n_{23} < 0; n_{12} > n_{13}$. Only in case $B_c[-+-] = |n_{12}|(M_2 - M_1) - |n_{13}| M_3 > 0$.

$$0 < \mu_0 H < |n_{12}|(M_2 - M_1) - |n_{13}| M_3) = B_c[-+-].$$

Appendix B: The magnetic phase diagrams (magnetization curves) in Case II ($n_{12} \neq n_{13} = n_{23}$)

We use the same notation as before to indicate regions of parameter values. For each range of parameters, we start by giving expressions for the relevant "critical fields", in decreasing order.

The field ranges and the field dependence of the magnetization in each field range are given in an obvious notation.

We classify the magnetization curve by its type. The magnetization curves appear to consist of a sequence of constant and linear portions. The type is a sequence of characters C, L and N, or N^+ or N^-, where C indicates a constant total magnetic moment, L a linear portion (i.e. $M_t = \mu_0 H / |n_{13}|$, in practice checked by extrapolation: the extrapolation should pass through the origin; the slope yields $1/|n_{13}|$). N, N^+ and N^- represent also linear portions (by extrapolation cutting off a (positive) magnetic moment at vanishing field; the slope now yields $1/|n_{12}|$), the difference being that N^+ and N^- occur in a magnetization curve with a L part, so one can determine the differences in the slopes. N^+ has a slope $1/|n_{12}| < 1/|n_{13}|$:

$$M_t = M_3(1 - |n_{13}|/|n_{12}|) + \mu_0 H/|n_{12}|,$$

whereas N^- has a slope $1/|n_{12}| > 1/|n_{13}|$:

$$M_t = M_3(|n_{13}|/|n_{12}| - 1) + \mu_0 H/|n_{12}|.$$

Case IIFF: $n_{12} > 0; n_{13} = n_{23} > 0$.
No critical field;
No non-collinear states; only the collinear state [+ + +] is stable.

$\mu_0 H$: $0 < [+++]$;
$M_t = M_c[+++] = M_1 + M_2 + M_3$, for $\mu_0 H > 0$;

Type: C.

Case IIAF: $n_{12} < 0; n_{13} > 0$.
Critical fields:

$$B_c[+++] = |n_{12}|(M_2 + M_1) - n_{13} M_3;$$
$$B_c[-++] = |n_{12}|(M_2 - M_1) - n_{13} M_3$$
(only in case $M_2 > M_1$).

Case (a): $B_c[+++] \leqslant 0$: only [+ + +], see IIFF.

Type: C.

Case (b): $B_c[-++] \leqslant 0 < B_c[+++]$.

$\mu_0 H$: $0 < [\mathrm{nc}3++] < B_c[+++] < [+++]$;
M_t: $M_3(1 + n_{13}/|n_{12}|) + \mu_0 H/|n_{12}| < M_3 + M_2 + M_1$;

Type: NC.

Case (c): $0 < B_c[-++] < B_c[+++]$; $(M_2 > M_1)$.

$\mu_0 H$: $0 < [-++] < B_c[-++] < [\mathrm{nc}3++] < B_c[+++] < [+++]$;
M_t: $M_3 + M_2 - M_1 < M_3(1 + n_{13}/|n_{12}|) + \mu_0 H/|n_{12}| < M_3 + M_2 + M_1$;

Type: CNC.

Case IIFA: $n_{12} > 0; n_{13} = n_{23} < 0$ (pseudo-two-sublattice system).
case $M_3 > M_1 + M_2$.

Critical fields:

$$|n_{13}|M_c[+++] = |n_{13}|\{M_3 + (M_2 + M_1)\};$$

$$|n_{13}|M_c[--+] = |n_{13}|\{M_3 - (M_2 + M_1)\};$$

$$\mu_0H:\quad 0 < [--+] < |n_{13}|M_c[--+] < [\text{ncF}] < |n_{13}|M_c[+++] < [+++];$$

$$M_t:\quad M_3 - (M_2 + M_1) < M_t = \mu_0H/|n_{13}| < M_3 + (M_2 + M_1);$$

Type: CLC.

case $M_3 = M_1 + M_2$ (compensation: $M_c[--+] = 0$).

The critical field $|n_{13}|M_c[--+]$ vanishes (only [ncF] and [+ + +] remain).

Type: LC.

case $M_3 < M_1 + M_2$.

Critical fields:

$$|n_{13}|M_c[+++] = |n_{13}|\{(M_2 + M_1) + M_3\};$$

$$|n_{13}|M_c[++-] = |n_{13}|\{(M_2 + M_1) - M_3\};$$

$$\mu_0H:\quad 0 < [++-] < |n_{13}|M_c[++-] < [\text{ncF}] < |n_{13}|M_c[+++] < [+++];$$

$$M_t:\quad (M_2 + M_1) - M_3 < M_t = \mu_0H/|n_{13}| < M_3 + (M_2 + M_1);$$

Type: CLC.

Case IIAA>: $n_{13} < n_{12} < 0$, so $|n_{12}| < |n_{13}|$.

case $M_3 \geqslant M_1 + M_2$. Same as IIFA, cases $M_3 > M_1 + M_2$ and $M_3 = M_1 + M_2$.

case $0 \leqslant M_2 - M_1 \leqslant M_3 < M_1 + M_2$.

Critical fields:

$$|n_{13}|M_c[+++] = |n_{13}|(M_2 + M_1) + |n_{13}|M_3\ (> 0);$$

$$|n_{13}|M_c[++-] = |n_{13}|(M_2 + M_1) - |n_{13}|M_3\ (> 0);$$

$$B_c[++-] = |n_{12}|(M_2 + M_1) - |n_{13}|M_3.$$

Case (a): $B_c[++-] \leqslant 0$: same as IIFA, case $M_3 < M_1 + M_2$.

Type: CLC.

Case (b): $0 < B_c[++-]$.

$$\mu_0H:\quad 0 < [\text{nc3}-] < B_c[++-] < [++-] < |n_{13}|M_c[++-] < [\text{ncF}] < |n_{13}|M_c[+++] < [+++];$$

$$M_t: M_t = M_3(|n_{13}|/|n_{12}| - 1) + \mu_0H/|n_{12}| < (M_2 + M_1) - M_3 < M_t = \mu_0H/|n_{13}| < M_3 + (M_2 + M_1);$$

Type: N$^-$CLC.

case $M_3 < M_2 - M_1$ (> 0).

Critical fields, see above, now added (possibly positive):

$$B_c[-+-] = |n_{12}|(M_2 - M_1) - |n_{13}|M_3.$$

Case (a): $B_c[++-] \leqslant 0$: see Case (a) above, same as IIFA, case $M_3 < M_1 + M_2$

Type: CLC.

Case (b): $B_c[-+-] \leqslant 0 < B_c[++-]$: see Case (b) above

Type: N$^-$CLC.

Case (c): $0 < B_c[-+-] < B_c[++-]$.

$$\mu_0H:\quad 0 < [-+-] < B_c[-+-] < [\text{nc3}-] < \{\text{and so on, see Case (b) above}\};$$

$$M_t:\quad (M_2 - M_1) - M_3 < M_t = M_3(|n_{13}|/|n_{12}| - 1) + \mu_0H/|n_{12}| < (\text{etc.})\ \{\text{see Case (b) above}\};$$

Type: CN$^-$CLC.

Case IIAA<: $n_{12} < n_{13} = n_{23} < 0$, so $|n_{13}| < |n_{12}|$.

Case $M_3 \geqslant M_1 + M_2; M_2 > M_1$ $\{M_2 = M_1$: see below$\}$.

Critical fields (decreasing order).

$$B_c[+++] = |n_{12}|(M_2 + M_1) + |n_{13}|M_3 \quad (> 0);$$

$$B_c[-++] = |n_{12}|(M_2 - M_1) + |n_{13}|M_3 \quad (> 0);$$

$|n_{13}|M_c[-++] = |n_{13}|(M_2 - M_1) + |n_{13}|M_3$

(>0);

$|n_{13}|M_c[+-+] = |n_{13}|M_3 - |n_{13}|(M_2 - M_1)$

(>0);

$B_c[+-+] = |n_{13}|M_3 - |n_{12}|(M_2 - M_1)$;

$B_c[--+] = |n_{13}|M_3 - |n_{12}|(M_2 + M_1)$.

Case (a): $B_c[+-+] \leqslant 0$.

$\mu_0 H$: $0 < [+-+] < |n_{13}|M_c[+-+]$

$< [\text{ncA}] < |n_{13}|M_c[-++] < [-++]$

$< B_c[-++]$

$< [\text{nc3}++] < B_c[+++] < [+++]$;

M_t: $M_3 - (M_2 - M_1) < M_t = \mu_0 H/|n_{13}|$

$< M_3 + (M_2 - M_1) < M_t = M_3(1 - |n_{13}|/|n_{12}|)$

$+\mu_0 H/|n_{12}| < M_3 + M_2 + M_1$;

Type: CLCN$^+$C.

Case (b): $B_c[--+] \leqslant 0 < B_c[+-+]$.

$\mu_0 H$: $0 < [\text{nc3}+-] < B_c[+-+] < [+-+]$

{and so on, see Case (a)};

M_t: $M_3(1 - |n_{13}|/|n_{12}|) + \mu_0 H/|n_{12}|$

$< M_3 - (M_2 - M_1)$ {etc., see Case (a)};

Type: N$^+$CLCN$^+$C; N$^+$ parts extrapolated form one straight line.

Case (c): $0 < B_c[--+] < B_c[+-+]$, so $M_3 > (|n_{12}|/|n_{13}|)(M_2 + M_1) > (M_2 + M_1)$.

$\mu_0 H$: $0 < [--+] < B_c[--+] < [\text{nc3}+-]$

{and so on, see Case (b)};

M_t: $M_3 - (M_2 + M_1) < M_3(1 - |n_{13}|/|n_{12}|)$

$+\mu_0 H/|n_{12}|$

{and so on, see Case (b)};

Type: CN$^+$CLCN$^+$C; N$^+$ parts extrapolated form one straight line.

Note. In case $M_2 = M_1$ the curves are simplified: Case (a) (above) does not occur; in Case (b) the type is reduced to type NC, and in Case (c) to type CNC.

Case $0 < M_2 - M_1 < M_3 < M_1 + M_2$.

Critical fields: see above; now $B_c[--+] < 0$, so Case (c) does not occur only the Cases (a) and (b) are still possible.

Note. In case $M_2 = M_1$ the curves are reduced in the same way as above: only Case (b), curve type NC, remains.

Case $M_3 = M_2 - M_1$ $(M_2 > M_1)$.

Critical fields: see above, down to $|n_{13}|M_c[+-+] = 0$.

Case (a) (above): the curve type is now reduced to type LCN$^+$C also to be regarded as a limit of the case $M_3 < M_2 - M_1$ below.

Case $M_3 < M_2 - M_1$.

Critical fields:

$B_c[+++] = |n_{12}|(M_2 + M_1) + |n_{13}|M_3 \quad (>0)$;

$B_c[-++] = |n_{12}|(M_2 - M_1) + |n_{13}|M_3 \quad (>0)$;

$|n_{13}|M_c[-++] = |n_{13}|(M_2 - M_1) + |n_{13}|M_3$

(>0);

$|n_{13}|M_c[-+-] = |n_{13}|(M_2 - M_1) - |n_{13}|M_3$

(>0);

$\mu_0 H$: $0 < [-+-] < |n_{13}|M_c[-+-] < [\text{ncA}]$

{and so on, see Case (a)};

M_t: $(M_2 - M_1) - M_3 < M_t = \mu_0 H/|n_{13}|$

{and so on, see Case (a)};

Type: CLCN$^+$C.

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