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Magnetic phase diagrams for three coupled magnetic moments

P.E. Brommer *

Van der Waals–Zeeman Institute Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

Abstract

For three arbitrary coupled magnetic moments (sublattices), the stable magnetic structures have been determined. Three classes do exist: three (all), two and no equal coupling parameters, respectively. At any applied field, only one, collinear or non-collinear, structure is stable. Transitions are smooth: the magnetization curves show no jumps.

Keywords: Magnetic phase diagram; Non-collinear structures

In many lanthanide-transition metal (RT) compounds, the magnetic structures can be described, in a two-sublattice model, as a coupling between the total R-moment and the total T-moment [1]. In applied fields, non-collinear structures can be formed [2], corresponding, in the absence of crystalline anisotropy, to a linear dependence, $M = \mu_0 H/|n|$. In this way, for antiferromagnetic coupling, the molecular-field constant $n(<0)$ can be measured directly. The influence of crystalline anisotropy is minimized by performing the experiments on an assembly of small, freely orientable, particles [3]. A two-sublattice model is not always adequate. For instance, in the case of two different R-moments on one sublattice, a three-sublattice model is necessary. Recently, $RMn_{6-x}Cr_xSn_6$ was described [4] as a three-sublattice system, simplified because the Mn-atoms occupy two *equivalent* sublattices (the R-moments form the third one). In Ref. [5], a complete treatment is presented for three *inequivalent* coupled moments, with constant magnitude. Here, the method is explained and the main results are presented.

The (free) energy of a system of magnetic moments $\boldsymbol{M}_1$, $\boldsymbol{M}_2$, $\boldsymbol{M}_3$, placed in an external field $\mu_0\boldsymbol{H}$, coupled either ferro- ($n_{ij} > 0$) or antiferromagnetically ($n_{ij} < 0$) is given by

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

The possible stable structures are found by deriving the equilibrium conditions:

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

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

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

The coefficients a_j and the determinant D of the coefficient matrix of this set of equations should be non-negative, mathematically in order to have an energy minimum, from a physical point of view, because the effective field acting on a moment $\boldsymbol{M}_j$ should be oriented along $\boldsymbol{M}_j$, i.e. equals $a_j\boldsymbol{M}_j$ with positive a_j. For a non-collinear state, D must vanish, since non-zero components perpendicular to $\boldsymbol{H}$ should exist. Moreover, considering a fixed moment $\boldsymbol{M}_1$ and variable moments $\boldsymbol{M}_2$ and $\boldsymbol{M}_3$, we find that the corresponding minor (subdeterminant) $m_{11} = a_2a_3 - n_{23}^2$, should also be non-negative. Using $m_{12} = a_3n_{12} + n_{13}n_{23}$ (cycl), we can derive

$$m_{11}\boldsymbol{M}_2 = (a_3 + n_{23})\mu_0\boldsymbol{H} + m_{12}\boldsymbol{M}_1. \quad (4)$$

An analogous relation holds for $\boldsymbol{M}_3$ (exchange indices 2 and 3). Hence, by addition:

$$m_{11}\boldsymbol{M}_t = (a_2 + a_3 + 2n_{23})\mu_0\boldsymbol{H} + (m_{11} + m_{12} + m_{13})\boldsymbol{M}_1. \quad (5)$$

For vanishing field, these equations reveal that either all minors have non-zero values (collinear structure) or all minors do vanish (non-collinear structure). In the latter case:

$$a_1 = -n_{12}n_{13}/n_{23};\ a_2 = -n_{12}n_{23}/n_{13};\ a_3 = -n_{13}n_{23}/n_{12}. \quad (6)$$

Since a_j should be positive (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).

For non-zero fields, we may exchange, in Eqs. (2a)–(2c), the first colon and the right hand side. Then, analogously, we find that a non-collinear solution may exist provided the sum $m_{11} + m_{12} + m_{13}$ vanishes. Looking in more detail, we find a natural classification of the coupled systems. Suppose, for instance, that m_{11} vanishes. Then,

* Fax: +31-20-525-5788; email: bos@phys.uva.nl.

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all minors should vanish. This can only happen in case all molecular-field constants are equal: $n_{12} = n_{13} = n_{23} = n$, referred to as Class I. Then, for $n < 0$, we find, from any of Eqs. (2a)–2(c), the relation $\boldsymbol{M} = \mu_0 \boldsymbol{H}/|n|$ in a non-collinear structure. Moreover, at a given field, all solutions (possibly non-coplanar) are degenerate.

In case m_{11} does not vanish, Eq. (4) shows that $\boldsymbol{M}_2$ (and by analogy $\boldsymbol{M}_3$) is a linear combination of $\boldsymbol{H}$ and $\boldsymbol{M}_1$. Consequently, if a non-collinear structure exists, with non-collinear $\boldsymbol{H}$ and $\boldsymbol{M}_1$, it is a coplanar one. Moreover, inserting $m_{11} + m_{12} + m_{13} = 0$ in Eq. (5), we see that $\boldsymbol{M}_t$ points in the direction of $\boldsymbol{H}$. Furthermore, in case two molecular-field constants are equal (referred to as Class II, with say $n_{13} = n_{23}$), only two distinct types of non-collinear structures may exist: either $\boldsymbol{M}_3$ is oriented along $\boldsymbol{H}$ (parallel or antiparallel), or $\boldsymbol{M}_1$ and $\boldsymbol{M}_2$ behave as one sublattice (oriented either parallel or antiparallel, 'pseudo-two-sublattice model'). In these non-collinear states, the magnetization depends linearly on $\boldsymbol{H}$. The magnetization curve consists of straight lines and constant parts, without any jump (see Fig. 1). Finally, if all molecular-field constants are different (Class III), such non-collinear structures cannot exist: no moment is directed along another moment or along the field. The magnetization curve consists of constant parts (collinear states) connected by possibly strongly curved parts (the non-collinear states). Fig. 2 shows, as an example, the result of a fitting procedure to some preliminary data observed on (Nd,Gd)Ni_2. The Ni sublattice is assumed to have a non-zero (constant) moment. In this way, a way is opened to determine directly the ferromagnetic interaction between a *light* lanthanide and a transition metal.

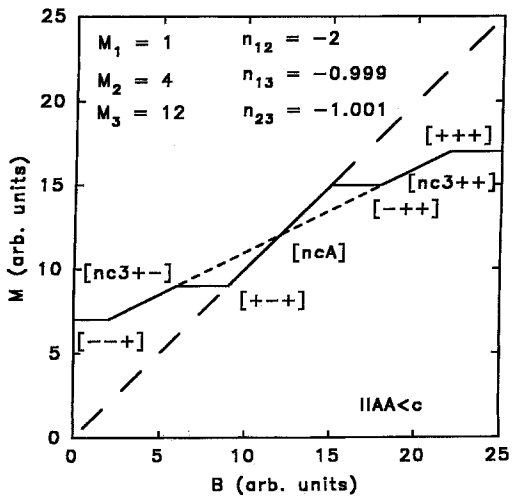

Fig. 1. Class II (the difference between n_{13} and n_{23} must be ignored). See Ref. [5] for more details.

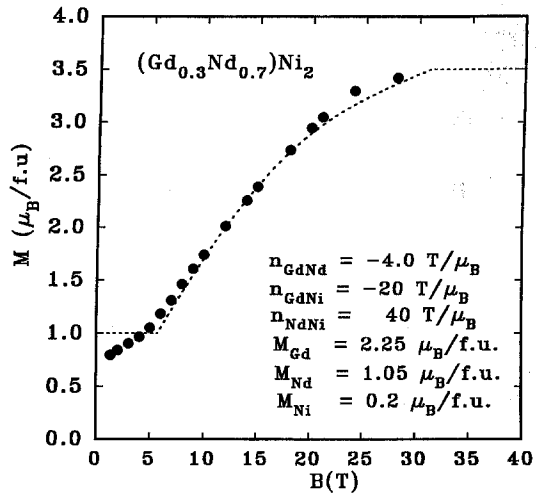

Fig. 2. Class III; application to (Nd,Gd) Ni_2 (see Ref. [5]).

In conclusion, the relationships between the coupling parameters appear to form a natural basis for classification. Class I, 'three equal coupling parameters', is rather simple, but has the remarkable property that 'non-coplanar' structures can occur. Class II, 'only two equal coupling parameters', yields interesting magnetization 'curves', still consisting of a combination of linear parts and constants. Class III, 'three different coupling parameters', allows a non-linear relation between $\boldsymbol{H}$ and $\boldsymbol{M}$.

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