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DOI

[10.1016/0304-8853\(95\)00016-X](https://doi.org/10.1016/0304-8853(95)00016-X)

Publication date

1995

Published in

Journal of Magnetism and Magnetic Materials

[Link to publication](#)

Citation for published version (APA):

Zhang, Z. D., Zhao, T., de Chatel, P. F., & de Boer, F. R. (1995). Spin configurations in the absence of a magnetic field in a two-sublattice system. *Journal of Magnetism and Magnetic Materials*, 147, 74-80. [https://doi.org/10.1016/0304-8853\(95\)00016-X](https://doi.org/10.1016/0304-8853(95)00016-X)

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ELSEVIER

Journal of Magnetism and Magnetic Materials 147 (1995) 74-80

Spin configurations in the absence of a magnetic field in a two-sublattice system

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Received 28 October 1994

Abstract

The spin configurations in the absence of an external magnetic field have been systematically investigated for a two-sublattice system. Based on a two-sublattice model, the conditions for the existence of collinear and non-collinear spin structures were derived for either ferromagnetic or ferrimagnetic materials and the phase diagrams of the spin configurations at zero field were drawn, taking into account the second-order anisotropy parameters of both sublattices. The inclusion of higher-order anisotropy is discussed and it is shown that in some cases the same phase diagrams result.

1. Introduction

The attention rare-earth-transition-metal (R-T) intermetallics have received recently [1,2] is primarily due to their application potential as materials for permanent magnets. Some magnetic properties, such as spontaneous magnetization, magnetocrystalline anisotropy and Curie temperature, are required to have high values for this purpose. Apart from the practical benefits, studies of these materials have provided insights leading to a better understanding of the magnetic properties of the 3d and 4f elements. Usually, the magnetic properties of the R-T compounds can be described satisfactorily by a two-sublattice model. This model simply considers the total magnetization as a vector sum of the magnetizations of the rare-earth and the transition-metal sublattices,

which interact via the R-T exchange coupling between the two sublattices.

The R-T spin exchange coupling is of an antiferromagnetic character and leads to a parallel 3d and 4f moment configuration for the light rare-earth metals and to an antiparallel moment configuration for the heavy rare-earths. It is well known that the anisotropies can also affect the relative orientation of the sublattice magnetizations. Usually, the R-T interaction is dominant since it is very strong in comparison with the anisotropies. Nevertheless, they can be comparable in some compounds, and these are the cases of interest in the present paper.

The magnetocrystalline anisotropy is due to the coupling of the magnetic moments of the rare-earth and transition-metal atoms to the crystal lattice. In most compounds, especially at low temperatures, the anisotropy of the R sublattice is stronger than that of the T sublattice. However, in some cases the anisotropy energies of the two sublattices are compa-

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rable in magnitude and may be seen to compete in determining the nature of the net anisotropy of the compound.

The magnetic isotherms of a compound, i.e. its magnetization as a function of the external magnetic field, reveal its intrinsic properties. The effect of the external magnetic field is that it forces the magnetic moments to align in the direction of the field. Depending on an interplay of intrinsic properties, the magnetization processes can be quite different. Some previous workers have studied the magnetization process based on the two-sublattice model. On the one hand, fixed single crystals and aligned-powder samples have been used to investigate the magnetic anisotropies of the compounds [3,4], and on the other, free single crystals and free-powder samples have been studied to determine the strength of the R-T interaction [5,6]. From the application point of view the R-T interaction ought to be strong enough to maintain the existing large magnetocrystalline anisotropy of the R ions to the highest possible temperature. Therefore, in most previous investigations it was assumed that the crystal fields are weaker than the R-T exchange coupling effect [3-6].

Evidently, a true test of the two-sublattice model should go beyond the above limiting cases. In some cases, the R-T interaction may not be strong enough to maintain the completely parallel or antiparallel coupling of the magnetic moments of the two sublattices. In the late 1970s Kudrevatykh et al. [7] as well as Ermolenko [8] considered the effect of the change of the exchange energy in an external field on the magnetization curves of RCo_5 and R_2Co_{17} compounds. Rinaldi and Pareti [9] studied a magnetic two-sublattice system with high competing single-ion anisotropies in the case of the $\text{Pr}_2(\text{Co,Fe})_{17}$ system, in which the Pr and T sublattices have intrinsic magnetocrystalline anisotropies of different signs. Sarkis and Callen [10], in a theoretical paper, showed that in ferrimagnetic materials, if the R-T coupling is not sufficiently strong to keep the R and T moments rigidly antiparallel, one observes a significant reduction in the macroscopic magnetocrystalline anisotropy. Recently, Zhang and co-workers [11-14] developed a calculation procedure to study the magnetization process of the two-sublattice system. A new peak emerging in the second derivative of the magnetization at low fields (in the d^2M/dH^2 versus

H curves) was found to depend on the competition of the weak R-T exchange and the opposite anisotropies [11,12]. For ferromagnetic materials, a decrease of the R-T exchange may result in an increased effective anisotropy [12]. This finding is consistent with the result of Sarkis and Callen [10]. Zhao et al. [15] also studied the effect of the anisotropy of the transition-metal sublattice on the magnetization process of a free-powder ferrimagnetic system.

In the present paper we investigate the two-sublattice model in cases when the relevant interactions are of comparable strength. We find that in the absence of an external field three energy parameters determine the behavior of the model: the R-T exchange energy and the appropriate combination of the anisotropy parameters of the two sublattices. Accordingly, a two-dimensional phase diagram can be constructed with the relative magnitude of the exchange energy with respect to one of the anisotropy energies and the ratio of the two anisotropy energies (cf. Eqs. (3.1) and (4.3) below) as coordinates. The most common cases, when the exchange energy far exceeds the anisotropy energy, which, in turn, is dominated by the R-sublattice contribution, are represented in this coordinate system by the areas in the outside corner of each quadrant. We shall be concerned with the areas close to both axes and the origin.

2. The model

The two-sublattice model to be studied in what follows is defined by the free-energy expression:

$$E = n_{AB} M_A M_B \cos \alpha + \sum_{i=1}^3 K_{iA} \sin^2 \theta_A + \sum_{i=1}^3 K_{iB} \sin^2 \theta_B. \quad (2.1)$$

Here M_A and M_B denote the magnitudes of the magnetization of sublattices A and B, respectively, θ_A and θ_B give their orientations with respect to the c-axis, and α is the angle between them. As the anisotropies within the basal plane are neglected and no external field is included, the two magnetization

vectors always lie in the same plane, which also contains the c -axis, so that the angle between them is $\alpha = \theta_B \pm \theta_A$. If we use a convention allowing for negative values of θ_A and θ_B [15], the latter can be eliminated from Eq. (2.1) without loss of generality and the model can be studied in terms of only two independent variables, θ_A and α :

$$E = n_{AB} M_A M_B \cos \alpha + \sum_{i=1}^3 K_{iA} \sin^2 \theta_A + \sum_{i=1}^3 K_{iB} \sin^2 (\theta_A + \alpha). \quad (2.2)$$

Here we are assuming that M_A and M_B and, consequently, the anisotropy parameters K_{iA} and K_{iB} are constants. The exchange interaction between the two sublattices is described by n_{AB} , the intersublattice molecular-field coefficient. A positive value of n_{AB} favors antiparallel alignment (ferrimagnetism), whereas a negative n_{AB} favors a parallel one (ferromagnetism).

The equilibrium state is found by minimizing Eq. (2.2) with respect to the angles θ_A and α . This involves the non-linear equations

$$\begin{aligned} \frac{\partial E}{\partial \theta_A} &= \sin 2\theta_A \sum_{i=1}^3 i K_{iA} \sin^{2(i-1)} \theta_A \\ &+ \sin 2(\theta_A + \alpha) \sum_{i=1}^3 i K_{iB} \sin^{2(i-1)} (\theta_A + \alpha) \\ &= 0, \end{aligned} \quad (2.3)$$

$$\begin{aligned} \frac{\partial E}{\partial \alpha} &= -n_{AB} M_A M_B \sin \alpha \\ &+ \sin 2(\theta_A + \alpha) \sum_{i=1}^3 i K_{iB} \sin^{2(i-1)} (\theta_A + \alpha) \\ &= 0, \end{aligned} \quad (2.4)$$

and the second derivatives of the free energy with respect to the angles θ_A and α :

$$\frac{\partial^2 E}{\partial \theta_A^2} = 2 \cos 2\theta_A \sum_{i=1}^3 i K_{iA} \sin^{2(i-1)} \theta_A$$

$$\begin{aligned} &+ \sin^2 2\theta_A \sum_{i=2}^3 i(i-1) K_{iA} \sin^{2(i-2)} \theta_A \\ &+ 2 \cos 2(\theta_A + \alpha) \sum_{i=1}^3 i K_{iB} \sin^{2(i-1)} (\theta_A + \alpha) \\ &+ \sin^2 2(\theta_A + \alpha) \sum_{i=2}^3 i(i-1) K_{iB} \\ &\times \sin^{2(i-2)} (\theta_A + \alpha), \end{aligned} \quad (2.5)$$

$$\begin{aligned} \frac{\partial^2 E}{\partial \alpha^2} &= -n_{AB} M_A M_B \cos \alpha + 2 \cos 2(\theta_A + \alpha) \\ &\times \sum_{i=1}^3 i K_{iB} \sin^{2(i-1)} (\theta_A + \alpha) \\ &+ \sin^2 2(\theta_A + \alpha) \sum_{i=2}^3 i(i-1) K_{iB} \\ &\times \sin^{2(i-2)} (\theta_A + \alpha), \end{aligned} \quad (2.6)$$

$$\begin{aligned} \frac{\partial^2 E}{\partial \theta_A \partial \alpha} &= 2 \cos 2(\theta_A + \alpha) \sum_{i=1}^3 i K_{iB} \\ &\times \sin^{2(i-1)} (\theta_A + \alpha) \\ &+ \sin^2 2(\theta_A + \alpha) \sum_{i=2}^3 i(i-1) K_{iB} \\ &\times \sin^{2(i-2)} (\theta_A + \alpha). \end{aligned} \quad (2.7)$$

All the results and discussion in the present work are derived from this group of equations.

3. Only lowest-order anisotropies considered

In this section we investigate the possible stable configurations of the two-sublattice model in the simplest case, when both sublattice magnetizations are characterized by a single parameter, K_1 , the coefficient of the lowest-order term in the anisotropy energy. The relative magnitude of the exchange and the anisotropy energies can then be simply described in terms of the parameters:

$$x = \frac{n_{AB} M_A M_B}{K_{1A}}, \quad y = \frac{K_{1B}}{K_{1A}}. \quad (3.1)$$

Using this notation, Eqs. (2.3) and (2.4) can be reduced to

$$\sin 2\theta_A + y \sin 2(\theta_A + \alpha) = 0, \quad (3.2)$$

$$-x \sin \alpha + y \sin 2(\theta_A + \alpha) = 0. \quad (3.3)$$

A collinear spin configuration will always satisfy these equations, because for $\alpha = 0, \pi$ and $\theta_A = 0, \pi/2, \pi$ every term in both equations vanishes. To investigate whether such solutions indeed correspond to energy minima one has to evaluate the second derivatives:

$$\frac{\partial^2 E}{\partial \theta_A^2} = 2K_{1A} [\cos 2\theta_A + y \cos 2(\theta_A + \alpha)], \quad (3.4)$$

$$\frac{\partial^2 E}{\partial \theta_A \partial \alpha} = 2K_{1A} y \cos 2(\theta_A + \alpha), \quad (3.5)$$

$$\frac{\partial^2 E}{\partial \alpha^2} = K_{1A} [-x \cos \alpha + 2y \cos 2(\theta_A + \alpha)], \quad (3.6)$$

and consider the expression

$$\begin{aligned} \Delta &= \frac{\partial^2 E}{\partial \theta_A^2} \frac{\partial^2 E}{\partial \alpha^2} - \left(\frac{\partial^2 E}{\partial \theta_A \partial \alpha} \right)^2 \\ &= 2K_{1A}^2 [-x \cos \alpha \cos 2\theta_A \\ &\quad + y(2 \cos 2\theta_A - x \cos \alpha) \cos 2(\theta_A + \alpha)] \end{aligned} \quad (3.7)$$

The criterion $\Delta > 0$, which ensures the existence of an extremum, is again satisfied by a collinear spin configuration, provided $y \geq 0$. Indeed, for $\alpha = 0, \pi$, we find

$$\Delta = 2K_{1A}^2 (2y \cos^2 2\theta_A \pm x(1+y) \cos 2\theta_A), \quad (3.8)$$

which can be made positive with $\theta_A = 0$ or $\pi/2$ for any positive value of y . It is easy to verify that the choice of θ_A , which makes Δ positive, leads to $\partial^2 E / \partial \theta_A^2 > 0$ so that the extremum found in this way is a minimum. This result is physically reasonable: as $y > 0$ implies that either both sublattices have easy-axis anisotropy or both of them easy-plane, nothing works against a collinear alignment of M_A and M_B . Accordingly, in the rest of this section we only discuss the case $y < 0$.

Fig. 1. Phase diagrams of the different spin configurations of a two-sublattice system in the absence of an external field ($K_{1A} > 0$). Only the second-order anisotropy constants of the two sublattices are taken into account. The definition of the axes x and y is in Eq. (3.1).

Inspection of Eqs. (3.4) through (3.7) shows that collinear configurations will give energy minima in the following cases:

(1) For ferrimagnetism ($n_{AB} > 0, \alpha = \pi$) in the four regions indicated in Fig. 1 under the conditions:

- I. For $x > 0$ and $0 > y > -1$:
 $x > -2y/(1+y)$ with $\theta_A = 0$;
- II. For $x > 0$ and $y < -1$:
 $x > 2y/(1+y)$ with $\theta_A = \pi/2$;
- III. For $x < 0$ and $y < -1$:
 $x < -2y/(1+y)$ with $\theta_A = 0$;
- IV. For $x < 0$ and $0 > y > -1$:
 $x < 2y/(1+y)$ with $\theta_A = \pi/2$.

(2) For ferromagnetism ($n_{AB} < 0, \alpha = 0$) in the same regions, with the values of θ_A interchanged, i.e. $\theta_A = \pi/2$ in regions I and III, while $\theta_A = 0$ in regions II and IV.

In Fig. 1 these results are summarized and the curves delineating the central part of the phase diagram, where the collinear structure is not stable, are given. In fact, there is some redundancy in the x - y representation, since regions I and II can be mapped on regions III and IV, respectively, by interchanging A and B in Eq. (3.1). It is readily verified that the corresponding transformation, $x' = xy^{-1}$; $y' = y^{-1}$, maps the curves in the diagonally opposite regions onto each other. The duplication of information can

be avoided by giving only two regions, e.g. only $y < -1$, which amounts to the convention that the sublattice with the larger anisotropy constant is denoted by B. We have chosen another convention: the sublattice with easy-axis anisotropy is to be denoted by A ($K_{1A} > 0$), which means that the $x > 0$ half-plane represents ferrimagnetic systems and the $x < 0$ half-plane ferromagnetic ones. This way, the cases (1) and (2) analyzed above can be represented in a single figure.

To find the non-trivial solutions of Eqs. (3.2) and (3.3), we first subtract Eq. (3.3) from Eq. (3.2) to find:

$$\sin 2\theta_A + x \sin \alpha = 0, \quad (3.9)$$

and use this result to eliminate θ_A from Eq. (3.31), which becomes

$$x \sin \alpha = y \sin \alpha \left[2\sqrt{1-x^2 \sin^2 \alpha} \sqrt{1-\sin^2 \alpha} - x(1-2\sin^2 \alpha) \right]. \quad (3.10)$$

Being interested only in non-collinear configurations ($\sin \alpha \neq 0$), we can divide by $\sin \alpha$ to obtain:

$$2y\sqrt{(1-x^2 \sin^2 \alpha)(1-\sin^2 \alpha)} = x[1+y(1-2\sin^2 \alpha)]. \quad (3.11)$$

After some manipulations, one can get an explicit expression for $\sin^2 \alpha$:

$$\sin^2 \alpha = 1 - \frac{[x(1-y)]^2}{4(y^2 - x^2 y)}, \quad (3.12)$$

which has a meaningful solution if

$$0 \leq \sin^2 \alpha \leq 1. \quad (3.13)$$

The first of these inequalities gives the conditions for the non-collinear spin configurations:

$$-\frac{2Y}{1+y} \leq x \leq \frac{2Y}{1+y} \quad \text{if } y < -1, \\ \frac{2Y}{1+y} \leq x \leq \frac{2Y}{1+y} \quad \text{if } 0 > y > -1, \quad (3.14)$$

whereas the second one is always satisfied if $y < 0$, which is the only case of interest here. The conditions derived above are valid for both ferromagnetic and ferrimagnetic cases.

The conditions Eq. (3.14) exactly fill in the gap left by the collinear configurations and the question of the nature of the transition between the two kinds of solutions arises. Substituting the equation of the curves representing the borderlines, $|x| = |2y/(1+y)|$, into Eq. (3.12) one finds that $\sin \alpha = 0$ along these lines; that is, the transition is continuous. Although the lines of $x = 0$ and $y = -1$ are special lines, they do not correspond to any transition since they are not the borders of the different spin configurations. In fact, the canting angle α does change continuously at such lines.

Next, we determine the derivative of the canting angle α with respect to x and y at the borderlines. One may determine this property from Eq. (3.12) as follows:

$$\frac{\partial \alpha}{\partial x} = \frac{\partial(\sin^2 \alpha)}{\partial x} / \sin 2\alpha, \quad (3.15)$$

$$\frac{\partial \alpha}{\partial y} = \frac{\partial(\sin^2 \alpha)}{\partial y} / \sin 2\alpha. \quad (3.16)$$

At the critical lines, we have:

$$\frac{\partial \alpha}{\partial x} \Big|_{x=\pm \frac{2y}{1+y}} = \pm \infty,$$

$$\frac{\partial \alpha}{\partial y} \Big|_{x=\pm \frac{2y}{1+y}} = \pm \infty.$$

It is clear that the transitions between the different spin configurations are of second order. At the line $y = -1$ and $x = 0$, no such singularities are found, we have:

$$\frac{\partial \alpha}{\partial x} \Big|_{y=-1} = \pm \frac{1}{8(1+x^2)},$$

$$\frac{\partial \alpha}{\partial y} \Big|_{y=-1} = \pm \frac{x}{2(1+x^2)};$$

$$\frac{\partial \alpha}{\partial x} \Big|_{x=0} = \pm \frac{y^2 + y + 1}{2y(1-y)^2},$$

$$\frac{\partial \alpha}{\partial y} \Big|_{x=0} = 0.$$

In Fig. 2, the dependences of the canting angle α on x and y are represented.

It is interesting to note that in the ferrimagnetic case ($n_{AB} > 0$) the phase boundaries identified in the present section can also be found by an analysis of the magnetization curves calculated for free-powder samples of such materials [15]. Such magnetization curves are usually, that is, provided the exchange interaction is strong enough, characterized by two critical fields, between which the 'bending process' takes place. Below the lower critical field, the two-sublattice magnetizations maintain their antiparallel orientation. In Ref. [15] an explicit expression is given for the lower critical field, which can be rewritten in terms of x and y , given by Eq. (3.1). The condition that the lower critical field must vanish gives the curves of Fig. 1 and it is indeed in the outside regions that a non-zero critical field can be given. This is exactly the picture emerging in the present analysis: in the inside regions no external field is needed for 'bending', i.e. a stable canted structure exists in the absence of a field.

4. Higher-order anisotropies included

The full analysis given in Section 3 cannot be repeated for the general case without recourse to numerical calculations. However, some formal analogies can be used to trace the conditions for the existence of collinear structures. Indeed, Eqs. (2.3) and (2.4) can be formally rewritten as:

$$\sin 2\theta_A + \eta \sin 2(\theta_A + \alpha) = 0, \quad (4.1)$$

$$-[\sin \alpha + \eta \sin 2(\theta_A + \alpha)] = 0, \quad (4.2)$$

where the parameters ξ and η are defined as

$$\xi = \frac{n_{AB} M_A M_B}{K_A}, \quad \eta = \frac{K_B}{K_A}, \quad (4.3)$$

with

$$K_A = \sum i K_{iA} \sin^{2(i-1)} \theta_A, \quad (4.4)$$

$$K_B = \sum i K_{iB} \sin^{2(i-1)} (\theta_A + \alpha). \quad (4.5)$$

The dependence of ξ and η on the angles θ_A and α makes Eqs. (4.1) and (4.2) incomparably more difficult than the corresponding Eqs. (3.2) and (3.3). However, limiting the considerations to collinear

Fig. 2. Dependence of the canting angle α on the values of x and y .

configurations ($\alpha_c = 0$ or π) leaves only one angular variable and one finds directly from Eq. (2.3):

$$\sin 2\theta_A^c \sum i K_i \sin^{2(i-1)} \theta_A^c = 0, \quad (4.6)$$

where

$$K_i = K_{iA} + K_{iB}. \quad (4.7)$$

Eq. (4.6) is formally identical to the one obtained for uniaxial magnets. In that case the K_i are the anisotropy parameters of the magnet, whereas here they stand for the sums of the corresponding anisotropy parameters of the two sublattices.

Asti and Bolzoni [16] have considered the equilibrium states of uniaxial magnets taking into account three anisotropy constants ($i \leq 3$). They have shown, by solving Eq. (4.6), that depending on the relative magnitude of anisotropy constants, easy-axis, easy-plane as well as easy-cone configurations may give the lowest free energy. In Ref. [16], the appropriate magnetic phase diagrams are given in terms of the variables $X = K_2/K_1$ and $Y = K_3/K_1$.

A glance at Eqs. (2.5)–(2.7) suffices to realize that the analysis of Section 3 cannot be repeated easily for the easy-cone case. However, for easy-axis and easy-plane situations again some formal identities are helpful. It can be easily verified that in both cases the analogues of Eqs. (3.12)–(3.14) are obtained, with x and y replaced by ξ and η as defined in Eq. (4.3), where

$$K_{A(B)} = K_{1A(1B)} \quad (4.8)$$

for $\theta_A = 0$ (easy axis), and

$$\mathbf{K}_{A(B)} = \sum_i i\mathbf{K}_{iA(iB)} \quad (4.9)$$

for $\theta_A = \pi/2$ (easy plane). On the basis of this formal analogy, the conclusions drawn in Section 3, including the phase diagram given in Fig. 1, remain valid in terms of the coordinates redefined in Eqs. (4.3), (4.8) and (4.9).

Concerning the easy-cone configurations, we come to the conclusion that a collinear structure is only exceptionally realizable. This is clear from Eqs. (2.3) and (2.4). In the collinear case ($\sin \alpha = 0$), the second term of Eq. (2.4) must vanish, which implies that both terms of Eq. (2.3) must be zero, not only their sum, as required by Eq. (4.6) for this case (easy-cone implies $\sin 2\theta_A^c \neq 0$). Physically, the simplest realization of this condition occurs when both sublattices have the same cone as preferred orientation of the magnetization, a very unusual situation indeed. Another possibility is that an angle θ_A^0 can be found, where both sublattices have an energy minimum (or one of them a maximum) though not necessarily the lowest minimum.

5. Summary

As the most commonly used rare-earth-transition-metal compounds are ferro- or ferrimagnetic, most studies of the two-sublattice model describing such systems are limited to collinear spin configurations. In the present paper we have shown that the model can describe a wealth of other possible spin configurations. The case when a single anisotropy constant is sufficient to characterize each sublattice has been analyzed in detail. We find that, if one of the sublattices has easy-axis anisotropy and the other one easy-plane, in the region of parameter space where the three relevant energies, the exchange energy and the two anisotropy energies, are comparable, the collinear spin structure is not stable. Unlike the collinear structures, which have always easy-axis or easy-plane, anisotropies, the non-collinear, canted structures have their easy magnetization direction around a cone (easy-cone anisotropy). The transition from the collinear to the canted structure is of second order.

A generalization of this analysis to cases where

not only the lowest-order anisotropy energies are of interest is only possible for easy-axis and easy-plane ground states. The reason for this can be seen in the fact that the first two derivatives of the higher-order anisotropies vanish at these two orientations of the magnetization ($\theta_A = 0$ or $\pi/2$). In the more complex and indeed more interesting case of easy-cone anisotropy, collinear structures are the exception and the dependence of the equilibrium orientations can be only determined by numerical methods.

Acknowledgements

The present work has been carried out within the scientific exchange program between China and the Netherlands and has been partly supported by the National Natural Science Foundation of China.

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