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# Induced staggered magnetic fields in antiferromagnets*

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Uniform magnetic fields may couple in higher order to the antiferromagnetic order parameter in a large class of compensated antiferromagnets. We describe this coupling as an induced staggered field. In this paper we give the general rules which determine when the coupling is allowed by the symmetry of the order parameter. We also discuss the dependence of the coupling on field direction. Some implications of these results for the behavior of transition-metal fluorides and for metamagnetic tricritical points are given. In particular we find that there is no induced staggered field in systems such as $FeCl_2$ while a coupling is expected in $FeCO_3$ and $FeF_2$, as well as in the recently discussed case of dysprosium aluminum garnet. In all these systems, however, the staggered field should vanish along certain symmetry directions for which metamagnetic tricritical points should exist.

## I. INTRODUCTION

It was shown in a recent publication[1] that the application of an external magnetic field to the compensated antiferromagnet dysprosium aluminum garnet could lead, depending on the direction of the external field, to an induced staggered magnetic field which coupled directly to the antiferromagnetic order parameter.

This effect is at first surprising since in the simplest models of antiferromagnets (the alternating-spin linear chain, for example) the order parameter is characterized by a wave vector $\vec{q} \neq 0$ and, rigorously, does not couple to a uniform field because there is no way that such a field can distinguish between the antiferromagnetic sublattices. Indeed the concept of a staggered magnetization is so suggestive of a nonzero wave vector that one tends to forget that there are many antiferromagnets for which the magnetic and crystallographic cells are the same and thus for which the wave vector of the order parameter is zero. In these materials, evidently including dysprosium aluminum garnet, uniform fields can distinguish one antiferromagnetic sublattice from another, and can couple to the order parameter.

In this note we discuss the coupling of a uniform magnetic field to the magnetic order parameter in antiferromagnets. We consider first (Sec. II) the restrictions on the form of the coupling due to the symmetry of the order parameter. Next (Sec. III) we discuss as a specific example the case of the transition-metal fluorides. We then (Sec. IV) consider implications of these results for tricritical points in metamagnets.

## II. RESTRICTIONS DUE TO SYMMETRY

The symmetry relevant to the problem of coupling of uniform fields and zero-wave-vector order parameters is that of the point group of the high-symmetry (paramagnetic) crystal structure. We look for combinations of the field and order parameter which are invariants of the point group and which give rise to a part of the magnetic free energy of the form

$$\tilde{G} = -\vec{H}^{\text{ind}} \cdot \vec{\eta} , \tag{1}$$

where $\vec{H}^{\text{ind}}$ is some function of the applied field $\vec{H}$, and $\vec{\eta}$ is the antiferromagnetic order parameter, which in principle can have many components. The form of Eq. (1) is such that $\vec{H}^{\text{ind}}$ has the same effect as a staggered field which alternates in sign among the different antiferromagnetic sublattices. In particular, the sign of $\vec{H}^{\text{ind}}$ can determine the sign of $\vec{\eta}$. Since it is induced by the applied field, $\vec{H}^{\text{ind}}$ may be appropriately called an induced staggered field.

All paramagnetic crystal phases are invariant under the operation of time reversal. By its nature as a magnetic order parameter $\vec{\eta}$ is odd under reversal. In order for $\tilde{G}$ to be invariant then, $\vec{H}^{\text{ind}}$ must be odd under time reversal also. It follows that $\vec{H}^{\text{ind}}$ must be an odd function of $\vec{H}$.

In the general procedure for finding invariants, we identify the point-group representations contained in $\vec{\eta}$ and then find odd-order combinations of the components of $\vec{H}$ which transform according to those representations. The direct products of corresponding $\vec{H}$ and $\vec{\eta}$ representations will then give the required invariants. An illustration of this procedure for the case of the fluorides is given in Appendix I.

Although the above method is completely general, it turns out that in the important case where just one component of the order parameter is relevant for the phase-transition behavior, there is a shortcut which makes matters much simplier. In this case the order parameter ($\vec{\eta}$) can at most change sign under the crystal-point-group operations. If we multiply those operations which change the sign of $\vec{\eta}$ by time reversal we have a group of operations, not including time reversal itself, which leave $\vec{\eta}$ unchanged. This group is called the "magnetic point group"[2] of $\vec{\eta}$. It is easy to see that the necessary and sufficient condition that Eq. (1) be invariant under the crystal point group and time reversal is that $\vec{\mathrm{H}}^{\mathrm{ind}}$ be invariant under the magnetic point group. The invariants of the 90 magnetic point groups are given in the literature.[2] It only remains to identify the magnetic point group of the order parameter and look up the allowed odd-order combinations of the applied field.

Of the 90 magnetic point groups, 31 allow a coupling of the order parameter with a uniform field in first order. These groups are at least weakly ferromagnetic and the nature of the coupling has already been discussed.[3,4] The 59 compensated antiferromagnetic groups may be divided as follows (notation is that of Ref. 2):

Coupling in third and higher order (27 groups)[5]: For example, for the group $\underline{4}/mm\underline{m}$, which is that of the $z$ component of the order parameter in the fluorides (see Appendix I), the lowest-order form of $\vec{\mathrm{H}}^{\mathrm{ind}}$ is $H_xH_yH_z$. For other third-order forms see Table IVe of Ref. 2.

Coupling in fifth and higher order—four groups: 422, $4mm$, $\bar{4}2m$, $4/mmm$. The lowest-order form of $\vec{\mathrm{H}}^{\mathrm{ind}}$ is $H_xH_yH_z(H_x^2-H_y^2)$.

Coupling in seventh and higher order—four groups: 622, $6mm$, $\bar{6}m2$, $6/mmm$. The lowest-order form of $\vec{\mathrm{H}}^{\mathrm{ind}}$ is $H_z(6H_x^5H_y-20H_x^3H_y^3+6H_y^5H_x)$.

Coupling in ninth and higher order—three groups: 432, $\bar{4}3m$, $m3m$. The lowest-order form of $\vec{\mathrm{H}}^{\mathrm{ind}}$ is $H_xH_yH_z(H_y^2-H_z^2)(H_z^2-H_x^2)(H_x^2-H_y^2)$.

No coupling in any order—21 groups: those which contain the operation inversion × time reversal.

To the last division we should, of course, add those systems for which the $\vec{q}$ of the order parameter is not zero. In these systems the sign of $\vec{\eta}$ can be changed by a simple lattice translation and it is clear that there are no invariants of the form of Eq. (1). We arrive thus at the following conclusion: For a one-component order parameter an induced staggered field can occur in any antiferromagnet *for which the wave vector of the order parameter is zero and for which the magnetic point group does not contain inversion × time reversal.* If the order parameter belongs to a multidimensional representation of the paramagnetic group, we first select the magnetic subgroups to which individual components belong and construct separate invariants in each of the groups. Any invariant of the paramagnetic group will be contained in a general linear combination of these invariants. To find combinations invariant under the paramagnetic group, we apply the additional operations of the full group to the general expression and particularize coefficients to ensure invariance.

## III. TRANSITION-METAL FLUORIDES

The magnetic properties of the transition-metal fluorides were considered some years ago by Dzyaloshinskii[3] and by Moriya.[4] They showed that these materials, in which there are two translationally inequivalent magnetic-ion sites per unit cell, could exhibit the property of "weak ferromagnetism," provided the components of the oppositely directed spins on the two inequivalent sites lay in the $x$-$y$ plane, as is the case for $\mathrm{NiF_2}$. For the case in which the spins lie along the $z$ axis, however, as in $\mathrm{MnF_2}$, $\mathrm{CoF_2}$, and $\mathrm{FeF_2}$, Dzyaloshinskii predicted, and it was found experimentally, that the antiferromagnetic structure is compensated, with no net magnetic moment.

However, it may be seen from Fig. 1 that a uniform magnetic field can distinguish between the two sublattices even in this case. A field along the direction [110], for example, affects spin 1 differently from spin 2, and so should destroy the exact cancellation of moments along $z$. This induced $z$ moment will be up or down according to the sign of the antiferromagnetic order parameter which may thus couple to a field component along $z$. The cou-

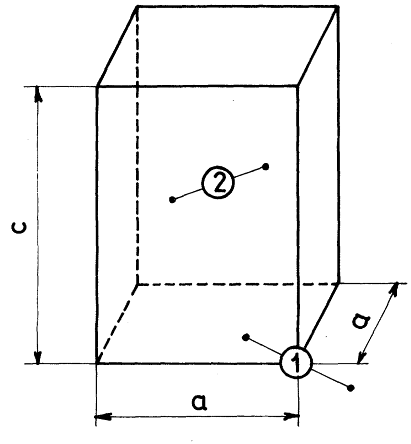

FIG. 1. Tetragonal unit cell for the rutile-structure transition-metal fluorides. Large numbered circles represent the two metal-ion sites $M$. Small circles represent $\mathrm{F^-}$ ions. The $\mathrm{MF_2}$ dumbbells of site 1 are repeated at all the cell corners. The dumbbell axes for the two sites differ by a 90° rotation about the $c$ axis.

pling thus requires field components along each of the $x$, $y$, and $z$ directions, a result which we will now obtain from group theory.

Letting the two spins in the unit cell be $\vec{S}_1$ and $\vec{S}_2$, we introduce $\vec{l} = \vec{S}_1 - \vec{S}_2$, the staggered magnetization. As shown in Appendix I, the components of $\vec{l}$ and of the field $\vec{H}$ form two reducible three-dimensional representations of $D_{4h}$, the crystal point group. Decomposition into irreducible parts shows that $l_z$ transforms as $B_{1g}$, the pair $\{l_y, l_x\}$ transforms as $E_g$, $H_z$ transforms as $A_{2g}$, and $\{H_x, H_y\}$ transforms as $E_g$. An invariant coupling to $\{l_x, l_y\}$ can be formed from the $E$ representation of $\vec{H}$ in first order:

$$G_1 = -\delta(H_x l_y + H_y l_x), \tag{2}$$

where $\delta$ is a constant. These are the coupling terms identified by Dzyaloshinskii and they give rise to the weak ferromagnetism associated with the order parameters $l_x$ and $l_y$.

To couple to $l_z$ requires an odd-order combination of $\vec{H}$ which transforms as $B_{1g}$. The lowest order of such a combination comes from decomposing the product representation $E_g \times E_g \times A_{2g}$ and gives rise to a term of the form (with $\beta$ a constant)

$$G_3 = -\beta H_x H_y H_z l_z \quad , \tag{3}$$

a result which could have been obtained from the magnetic point group of $l_z$. The magnetic free energy for the rutiles can then be written in the form

$$\begin{aligned} G = {} & \tfrac{1}{2} A \vec{l}^{\,2} + \tfrac{1}{2} a l_z^2 - \tfrac{1}{2} b_z H_z^2 - \tfrac{1}{2} b_\perp (H_x^2 + H_y^2) - \delta(H_x l_y + H_y l_x) \\ & - \beta H_x H_y H_z l_z \\ & + (\text{other fourth- and higher-order terms}), \end{aligned} \tag{4}$$

where $A$, $a$, $b_z$, and $b_\perp$ are functions of the temperature. Equation (4) is to be minimized with respect to $\vec{l}$ to obtain the thermodynamic properties of the system.

Although the presence or absence of induced staggered fields may be determined by symmetry considerations alone, the magnitude of the effect requires consideration of physical mechanisms. The physical origin of the parameters $\delta$ and $\beta$ in the free energy is, in the rutile structure, a consequence of the fact that the principle axes of the $g$ tensors of the two magnetic ions in the unit cell are rotated with respect to one another by 90°. Moriya[6] showed that this could explain the parameter $\delta$, and in Ref. 1 it was shown that a nonzero value of $\beta$ could also be produced in this way. A related phenomenon, the presence of nonlinear terms in the susceptibility of antiferromagnets, has been discussed by Dzyaloshinskii[7] and Shtrikman and his co-workers.[5]

A simple example illustrates the $g$-tensor effect mentioned above. We consider two spins $\frac{1}{2}$ coupled antiferromagnetically with an Ising-type interaction in an external field with components both along the $z$ axis and perpendicular to it. The Hamiltonian is

$$\begin{aligned} & H = H_0 + V, \\ & H_0 = J S_1^z S_2^z - g_z \mu_B H_z (S_1^z + S_2^z), \\ & V = \mu_B H_\perp (g_a S_1^x + g_b S_2^x), \quad J > 0 \quad . \end{aligned} \tag{5}$$

We assume that the $g$ factors for coupling to the transverse field are different for the two spins, i.e., $g_a$ for spin 1 and $g_b$ for spin 2, because of the different orientations of the principal axes of the $g$ tensors for the two sites. The ground state of $H_0$ is readily seen to be doubly degenerate with $E_{+-} = E_{-+}$, where $E_{mm'}$ is the energy of the state with $S_1^z = m$ and $S_2^z = m'$. Such a degeneracy would be lifted by interaction with a staggered magnetic field along the $z$ axis, i.e., by a field which points up at spin 1 and down at spin 2. The effect of adding the interaction $V$ is also to lift this degeneracy, as a simple second-order perturbation calculation shows. We find

$$E_{-+} - E_{+-} = 2(g_a^2 - g_b^2)(\mu_B H_\perp / J)^2 g_z \mu_B H_z \ , \tag{6}$$

where we have assumed $\mu_B H_z \ll J$. We can construct a spin Hamiltonian for the interaction $V$ if we restrict the action of that Hamiltonian to the two-fold degenerate ground state, neglecting interactions with the states $|{++}\rangle$ and $|{--}\rangle$. The effects of $V$ are then the same as those of

$$V_{\text{eff}} = -g_z H^{\text{ind}} (S_1^z - S_2^z),$$

where the effective staggered field $\vec{H}^{\text{ind}}$ is given by

$$H^{\text{ind}} = (g_a^2 - g_b^2)(\mu_B H_\perp / J)^2 H_z \ .$$

This staggered field is proportional to $|H|^3$, as expected from the group-theoretical argument. A rough order of magnitude for $\vec{H}^{\text{ind}}$ for an antiferromagnet can be obtained for $FeF_2$. If $H_\perp = H_z \sim H/\sqrt{3}$ and $J \sim kT_N$, then $\vec{H}^{\text{ind}} \sim 30$ G when $H = 50$ kG. This induced staggered field should be large enough to produce single antiferromagnetic domain samples of $FeF_2$ and other fluorides.

Similar effects are expected to be found in other substances, and different physical machanisms can then be responsible for the $\delta$ and $\beta$ terms. Thus, Moriya[6] showed that antisymmetric exchange produces the $\delta$ term in $Fe_2O_3$ and $CoCO_3$, among others, and Giordano *et al.*[8] have shown that the geometric arrangement of interacting spins in the presence of an external field could produce an effective staggered field ($\beta$ term) and that such a mechanism is present in DAG. Similarly, higher-order exchange interactions can produce $\delta$-type terms in other materials.

TABLE I. Transformation properties of $\vec{l}$ and $\vec{H}$ under the elements (a) of $D_{4h}^{14}$ for the rutile structure fluorides.[a]

| | $(E\mid 0)$ | $(2_z\mid 0)$ | $(4_z\mid\vec{\tau})$ | $(4_z^3\mid\vec{\tau})$ | $(2_x\mid\vec{\tau})$ | $(2_y\mid\vec{\tau})$ | $(2_{xy}\mid 0)$ | $(2_{x\bar{y}}\mid 0)$ |
|---|---|---|---|---|---|---|---|---|
| $l_x$ | $l_x$ | $-l_x$ | $l_y$ | $-l_y$ | $-l_x$ | $l_x$ | $l_y$ | $-l_y$ |
| $l_y$ | $l_y$ | $-l_y$ | $-l_x$ | $l_x$ | $l_y$ | $-l_y$ | $l_x$ | $-l_x$ |
| $l_z$ | $l_z$ | $l_z$ | $l_z$ | $l_z$ | $l_z$ | $l_z$ | $-l_z$ | $-l_z$ |
| $H_x$ | $H_x$ | $-H_x$ | $-H_y$ | $H_y$ | $H_x$ | $-H_x$ | $H_y$ | $-H_x$ |
| $H_y$ | $H_y$ | $-H_y$ | $H_x$ | $-H_x$ | $-H_y$ | $H_y$ | $H_x$ | $-H_y$ |
| $H_z$ | $H_z$ | $H_z$ | $H_z$ | $H_z$ | $-H_z$ | $-H_z$ | $-H_z$ | $-H_z$ |

[a] $\vec{\tau}=\frac{1}{2}(\vec{a}+\vec{a}+\vec{c})$.

## IV. TRICRITICAL POINTS IN METAMAGNETS

The interest in producing induced staggered fields is particularly great for metamagnets with tricritical points. For these materials the presence of a staggered field carries us away from the tricritical point in a dimension which has not been generally thought to be accessible. A recently discussed case in point is dysprosium aluminum garnet (DAG)[1] for which the existence of a coupling for fields along [111] was argued from a physical mechanism. The precise form of this coupling may now readily be obtained from the rules given in Sec. II. The order parameter in DAG is one dimensional and for the presently accepted structure[9] the magnetic point group is $m3m$. For this group the lowest-order invariant is found in Ref. 2 to be of the Form $H_xH_yH_z$. Thus for DAG tricritical points should occur for fields in symmetry directions such as [100] and [110], and the staggered field portion of the phase diagram should be accessible with field components along [111].[10]

Another metamagnet for which the magnetic and crystallographic cells are the same and for which staggered fields are allowed is $FeCO_3$. Again for this case the order parameter is one dimensional and the form of the induced staggered field is determined by the magnetic point group ($\bar{3}m$). The allowed term is $H_y^3-3H_yH_x^2$. Thus for applied field along $z$ there is no induced staggered field and a tricritical point should occur. The staggered field dimension of the phase diagram can be explored witl a field in the $xy$ plane.

The occurrence of induced staggered fields in metamagnets is far from universal, however. For $FeCl_2$ and $FeBr_2$, for example, the vector of the order parameter is nonzero and thus induced staggered fields cannot be produced in these substances.

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## APPENDIX

The rutile structure belongs to the space group $P4_2/nmn-D_{4h}^{14}$ whose elements are combinations of the primitive translations $\vec{t}=n_1\vec{a}+n_2\vec{a}+n_3\vec{c}$ and the rotational elements

(a) $(E\mid 0)$, $(2_z\mid 0)$, $(4_z\mid\vec{\tau})$, $(\bar{4}_z\mid\vec{\tau})$,
$(2_x\mid\vec{\tau})$, $(2_y\mid\vec{\tau})$, $(2_{xy}\mid 0)$, $(2_{x\bar{y}}\mid 0)$

and

(b) $(i\mid 0)$, $(m_z\mid 0)$, $(\bar{4}_z\mid\vec{\tau})$, $(\bar{4}_z^3\mid\vec{\tau})$,
$(m_x\mid\vec{\tau})$, $(m_y\mid\vec{\tau})$, $(m_{xy}\mid 0)$, $(m_{x\bar{y}}\mid 0)$,

where $(R\mid\vec{\tau})$ denotes a proper or improper rotation $R$, followed by a nonprimative translation $\vec{\tau}=\frac{1}{2}\vec{a}+\frac{1}{2}\vec{a}+\frac{1}{2}\vec{c}$. The elements (b) are those of (a) taken together with inversion, the inversion center being at a metal-ion site. The group $\{(R\mid\vec{\tau}+\vec{t})\}$ is isomorphous with the point group $D_{4h}$ under the correspondence $\{(R\mid\vec{\tau}+\vec{t})\}\to R$.

The transformation properties of $\vec{H}$ are determined entirely by the rotation operations. The transformations of $\vec{l}=\vec{S}_1-\vec{S}_2$, on the other hand, are affected by the fact that the translation $\vec{\tau}$ interchanges sublattices 1 and 2. It may be seen by reference to Fig. 1 that $\vec{l}$ is invariant under inversion, as is $\vec{H}$. Thus we only need consider the transformation under the subset (a) above. These are given in Table I. From the table it is straightforward to construct character tables for $\vec{l}$ and $\vec{H}$, find the irreducible representations of $\vec{l}$, $\vec{H}$ and odd products of $\vec{H}$, and thus find the invariant coupling terms as described in the text.

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