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REGULAR SUPERSTRUCTURES OF MAGNETIC TRANSLATIONAL LATTICES

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In the present paper an attempt is made to discuss the magnetic superstructures from the point of view of spin waves propagation. The classification of superstructures proposed by the author in earlier papers is here developed more precisely, introducing symmetric and non-symmetric superstructures and dividing the former into regular and non-regular orders. The present paper deals with the regular order only. The discussion comprises mainly antiferromagnetic translational lattices, though the general conclusions hold also for ferri- and antiferrimagnetic superstructures.

As the most characteristic property of regular superstructures, the author assumes the presence of alternating parallel ferromagnetic planes. Their existence allows to predict the phenomenon of superstructural anisotropy of spin waves dispersion. The presumable behaviour of spin waves in particular superstructures of cubic lattices is discussed in detail. The theoretical proofs of these hypotheses are announced.

Moreover, a simple geometrical interpretation of Luttinger's method of description of superstructures is given.

I. Introduction

In the present paper, the classification and some general properties of magnetic superstructures are considered from the point of view of the theory of spin waves. The subject covers all magnetic translational lattices consisting of two kinds of spins: $\boldsymbol{A}$ and $\boldsymbol{B}$, each kind forming a ferromagnetic sublattice. These sublattices are labelled A and B respectively. The numbers of the lattice points in A and in B are assumed to be equal; consequently, both interpenetrating sublattices will be identical. Considerations deal chiefly with antiferromagnetic lattices, but the classification and general conclusions hold also for ferri- and antiferrimagnetic ones. The detailed discussion concerns those superstructures, which will here be termed "*regular*".

In the subsequent sections, certain theorems concerning the various categories of superstructures introduced in Chapter II will be given. It should be pointed out that some of these theorems present the character of empirical rules. It is to be hoped that in the future they may be derived theoretically.

II. Classification of superstructures

In previous papers (Cofta 1958 and 1959, Szczeniowski and Cofta 1959) a classification of superstructures which appeared convenient for studying spin waves in ferri- and antiferrimagnetics was proposed. This classification will now be developed more precisely.

1) Symmetric and non-symmetric superstructures

The first necessary step consists in dividing all superstructures considered into two classes, which we shall define as follows. A superstructure will be said to be *symmetric* if each lattice point is a center of symmetry. In other words, we are dealing in this case with such orders of points A and B for which the following rule holds:

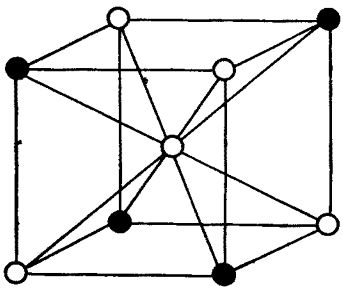

Fig. 1. Example of non-symmetric superstructure in *bcc* lattice

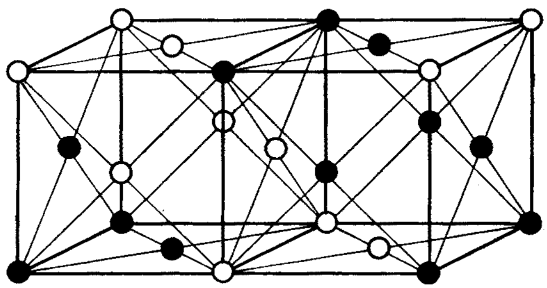

Fig. 2. Example of non-symmetric superstructure in *fcc* lattice

if the lattice point $\boldsymbol{r}$ belongs to A, then $-\boldsymbol{r}$ also belongs to A, and analogically for the sublattice B. This is clearly valid for any origin of the coordinates system (fixed at an arbitrary lattice point).

All superstructures that are not symmetric will be termed *non-symmetric.* From the above definition it follows immediately that, in the case of a non-symmetric superstructure, for each lattice point there exists at least one such relative position $\boldsymbol{r}$, belonging to A, for which $-\boldsymbol{r}$ belongs to B and vice versa. The appearence of such non-symmetric linear chains ... A A B B A A B B ... of nearest or second neighbours distinguishes essentially the non-symmetric superstructures from symmetric ones with respect to the propagation of spin waves in antiferromagnetics, since the ordering ... A A B B A A B B ... of the spins probably constitutes a kind of "obstacle" for spin waves (Cofta 1959). Two examples of unsymmetric superstructures are shown in Figs. 1 and 2. A detailed analysis of unsymmetric orders will be given in a subsequent paper. Let us but state that the existence of unsymmetric linear chains implies directly the impossibility of a translational character of the sublattices A and B, which therefore must be lattices with basis.

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2) Regular and non-regular superstructures

In the case of symmetric superstructures two possibilities exist:
a) both sublattices A and B are translational lattices;
b) the sublattices A and B are not translational lattices (i.e. they are lattices with a basis).

For the first of these two categories, let us retain the name "*regular superstructures*", introduced previously (Cofta 1958 and 1959) in a not entirely exact manner. We shall examine the regular orders in the ensuing Chapters.

The second category of symmetric superstructures will be termed "*non-regular*". In this case, calculation of spin wave problems is more complicated than for regular superstructures. Indeed, the equations for the spin wave amplitudes must be written separately for each translation sublattice. Therefore, if the basis of sublattice A (or B)

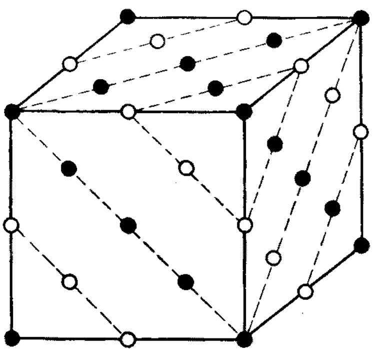

Fig. 3. Example of non-regular superstructure in *fcc* lattice

consists of n points, we have to solve $2n$ simultaneous equations, leading in general to $2n$ branches of the energy spectrum. Instead, in the case of regular superstructures we have $n = 1$ and therefore always only two branches are obtained.

The class of non-regular superstructures does not comprise a large number of examples[1], as compared with the other two classes (i.e. regular and unsymmetric ones). A typical example of non-regular superstructure is that of the hypothetic "order of the second kind" of the *fcc* lattice, considered often in the literature (see e.g. Van Vleck 1951 and Roth 1958). This superstructure is shown in Fig. 3.

III. Characteristic property of regular superstructures

1. Ferromagnetic lattice planes

It is a well known fact that in some cases of superstructures, ferromagnetic sheets occur. Sets of parallel lattice planes, each consisting exclusively of A points or exclusively of B points, exist in the case of non-symmetric as well as in that of

[1] See the note at the last page of the present paper

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regular superstructures (their existence in non-regular superstructures does not seem possible). Among the manifold types of sets of parallel ferromagnetic sheets, the most interesting is the case of alternatingly arranged planes: ... A B A B ... In this case, either of both sublattices consists of identical equidistant lattice planes, and consequently is a translational lattice. This proves that alternating ferromagnetic sheets can occur only in the case of regular orders. We believe that they must occur in each regular superstructure. Hence, we will consider the presence of alternating ferromagnetic sheets as the characteristic property of regular superstructures.

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2. *Three types of regular orders*

The foregoing property enables us to distinguish three different types of regular superstructures. As a criterion of such a classification we shall use the number of different sets of alternating parallel ferromagnetic planes, taking into account only those with the lowest Miller indices (i.e. 0 or 1). These three types are the following:

α) *Monoplanar type.* Only one set of parallel ferromagnetic sheets exists. (As to examples, see Ch. V).

β) *Biplanar type.* Two differently directed, equivalent sets of parallel ferromagnetic sheets exist. (For examples, see Ch. V).

γ) *Multiplanar type.* More than two equivalent sets of alternating ferromagnetic sheets, each having a different direction, exist. This type is realised only in the ortho-

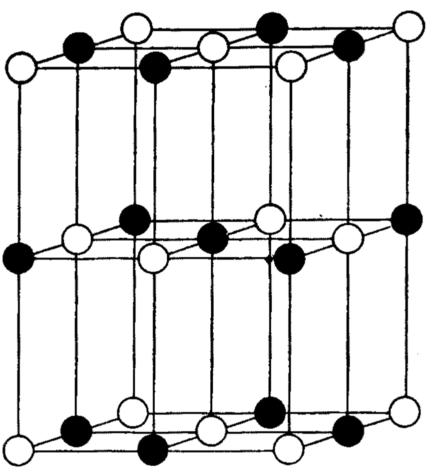

Fig. 4. Natural (multiplanar) superstructure of the simple orthorhombic lattice

rhombic, tetragonal and cubic Bravais systems. It is readily seen that the symmetry degree determines strictly the number of sets in the case of "*multiplanar*" type: there can be only 4 different equivalent sets, namely the 4 sets of planes (111) (Fig. 4). However, it should be mentioned that in the case of body centered lattices there exist

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also 3 sets (100), (010), (001) with lower Miller indices (see Fig. 5), whereas in the case of face centered lattices the multiplanar type cannot be realised.

As can be seen from Fig. 4, a. 5, the "multiplanar" type of regular superstructure is identical with the well-known order in which every nearest neighbour of any given lattice point belongs to the other sublattice. As proposed previously (Cofta 1958), we prefer to use for this case the term "*natural order*".

In the subsequent sections it will be shown that all the natural orders must possess the same physical properties; moreover, they will be seen to present the least interesting cases of regular superstructures.

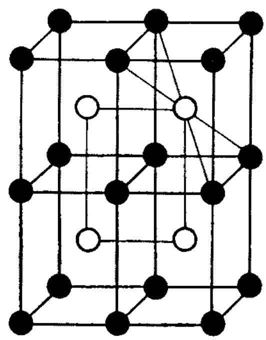

Fig. 5. Natural (multiplanar) superstructure of the body centered orthorhombic lattice

Our observations on the existence and role of sets of alternating ferromagnetic sheets provide a simple way of marking the individual regular superstructures. This consists in giving, in addition to the lattice symbol, the Miller indices of the unique (for uniplanar) or two (for biplanar orders) sets of ferromagnetic sheets. Natural superstructures require no additional marking.

IV. Luttinger's description of superstructures

The results of Luttinger's (1951) considerations relating to the energy of Ising's model provide a very simple method of description of individual superstructures. It is sufficient to find such a vector $\boldsymbol{v}$, appropriate for the given superstructure, for which:

$$\boldsymbol{vr} \text{ is an even number, if } \boldsymbol{r} \text{ belongs to A,}$$

$$\boldsymbol{vr} \text{ is an odd number, if, } \boldsymbol{r} \text{ belongs to B,}$$

or *vice versa*. Then, for each point of A, the number

$$\sigma_{\boldsymbol{r}} = (-1)^{\boldsymbol{vr}} = e^{i\pi \boldsymbol{vr}} \tag{1}$$

is equal to 1, and for each point of B, to -1, or vice versa (Ziman 1952). Let us term $\boldsymbol{v}$ the Luttinger vector. Clearly, $\boldsymbol{v}$ is a lattice vector of the appropriate reciprocal lattice.

Luttinger has calculated the vectors $\boldsymbol{v}$ for the following 5 superstructures (in the notation introduced above): *sc*-(110)-(1$\bar{1}$0), *fcc*-(001), *fcc*-(111) and for both natural superstructures of cubic lattices (*sc* and *bcc*). The Luttinger vectors for other superstructures — if existent — could be found by guessing. When Luttinger vectors are used for calculations by the spin-wave method (Ziman 1952), no information is available on the range of validity of the formulas obtained, since nothing is as yet known on the existence of Luttinger vectors for the various superstructures. It seems that e.g. Ziman's dispersion law for antiferromagnetics is often held to be valid for few orders only (see e.g. Elliot and Lowde 1955).

The situation is entirely clarified on considering the connection between the Luttinger vectors and the alternating ferromagnetic sheets. The connection is a very simple one: the Luttinger vector is a vector perpendicular to the ferromagnetic planes, so that their equation is of the form

$$\boldsymbol{v}\boldsymbol{r} = \mathrm{n} \tag{2}$$

n being an integer. Choosing the origin e.g. at a point of A, we obtain, for n even, the equations of the A planes, and, for n odd, those of the B planes. In other words, the number

$$\sigma = (-1)^n \tag{3}$$

changes its sign on transition from any ferromagnetic plane to the neighbouring one. Now, it is obvious that for the biplanar superstructures we can find two equivalent Luttinger vectors having different directions, and for the natural orders there exist as many as 4 different Luttinger vectors which can be used in the description of such superstructures.

The geometrical interpretation of Luttinger vectors proposed here yields immediately the important conclusion that Luttinger's method of the description of superstructures is applicable to all regular orders, and to these only. Consequently, the scope of validity of Ziman's (1952) dispersion law for spin waves in antiferromagnetics can be strictly determined: this law is valid exactly for regular superstructures of translational (magnetic) lattices (see also Cofta 1959, page 222).

Instead of Miller indices we may, of course, employ the Luttinger vectors for marking indivdual superstructures.

V. Superstructural anisotropy of spin waves dispersion

1. Isotropic and anisotropic superstuctures

The division of regular superstructures into three types has not only a formal meaning but also corresponds to physical differences in the propagation of spin waves. Let us first consider the monoplanar orders. The very characteristic arrangement of spins undoubtedly privileges the $\boldsymbol{v}$ direction perpendicular to the ferromagnetic

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sheets. This suggests that the same direction will be the preferred one with regard to the propagation of spin waves. For the same reason, it should be expected that in the biplanar superstructures the direction $\boldsymbol{v} = \boldsymbol{v}_1 \times \boldsymbol{v}_2$ along the line of intersection of two nonparallel ferromagnetic planes will be the privileged one for the spin waves.

By the foregoing, the monoplanar as well as the the biplanar orders can be considered to be anisotropic superstructures. In such superstructures, the spin waves should obey an anisotropic dispersion law, independently of the presence of the structural anisotropy. According to an earlier proposal of ours (Szczeniowski and Cofta 1959), the possible effect predicted here will be termed "superstructural anisotropy of spin wave dispersion". A more detailed discussion is inserted in Chapter VI.

In the natural superstructures the situation is quite different. For reasons of lattice symemtry all four $\boldsymbol{v}$ directions of (111) ferromagnetic sheets are equivalent and the same holds for the six directions of intersection of ferromagnetic planes. Therefore no direction is preferred. This conclusion expresses the well-known fact that for natural superstructures the isotropic dispersion law holds (naturally, if there is no structural anisotropy). Hence, the expected behaviour of spin waves in mono- and biplanar orders should be essentially distinct from their behaviour in natural orders.

It is to be noted, however, that the conclusions of the present section are valid only when the exchange integral between two atomic spins depends solely on their distance i.e. when the direct as well as indirect exchange interactions are isotropic. If in positions A and B there are different ions, our suggestions must be slightly corrected. If, however, the chemical lattice is so highly complicated that the indirect exchange coupling is different between different spins of the same sphere of neighbours or, moreover, when semicovalent exchange interactions between coplanar orbitals exist, then our conclusions must be radically revised.

2. *Effects of second interactions*

When the second interactions i.e. interactions between next-nearest neighbours are present, then an important role is played by the substructures of the given lattice. Let us recall that each Bravais lattice of a rectangular system can be divided into 2 or 4 interpenetrating lattices of the same system. These identical component lattices are called substructures or submotifs (Roth 1958). In this way:

1° a simple lattice is composed of 2 face centered ones;
2° a body centered lattice is composed of 2 simple ones;
3° a face centered lattice is composed of 4 simple ones.

The role of substructures is clear with respect to the well-known fact that each substructure consists solely of points which are second neighbours in the whole lattice. A natural suggestion results from this fact. Namely, in the dispersion law the term due to second interactions should show a character corresponding to the type of spin order of the given substructures. Thus, when substructures have mono- or biplanar order, an anisotropic second interactions term may be expected. Instead, when the substructures are ferromagnetic or present the natural order, an isotropic second interactions term may be anticipated (when not taking into account the purely structural effects).

VI. Regular superstructures in cubic antiferromagnetic lattices

We shall now proceed to review the particular regular superstructures which can appear in cubic lattices. The cubic superstructures are the most convenient examples, not only due to their simplicity but also because of the absence for them of structural anisotropy. Although here we confine our considerations to antiferromagnetics, the conclusions will be valid probably also for ferrimagnetic translational lattices.

Let the lattice constant in the structures considered be a. All conclusions of this Chapter hold exactly only if the condition of isotropy of exchange interactions is satisfied.

1. Review of cubic regular superstructures

a) Natural superstructures

First of all we shall settle the matter of natural superstructures. Such orders can appear in *sc* and *bcc* lattices only; in the *fcc* lattice their existence is impossible, since in this structure the nearest neighbours of a given lattice point are also nearest neighbours to one another. In contradiction to the opinions met with sometimes, it should be stated that the natural superstructure is no typical order in antiferromagnetics (as well as in ferrimagnetics with antiparallel spins). It can appear only when negative i.e. antiferromagnetic coupling between nearest neighbours predominates. On the other hand, in most substances with antiparallel spins, strong negative second interactions seem to exist. Only three cases of the natural superstructure in cubic lattices have been revealed hitherto. These are the antiferromagnetic perovskites $LaCrO_3$ (Koehler and Wollan 1957), $LaFeO_3$ (l.c.) and $CaMnO_3$ (Wollan and Koehler 1955) with simple cubic magnetic lattices. All these compounds owe their natural superstructure to the fact that here each two nearest magnetic ions are colinearly separated by an intervening oxygen ion. Let us mention that, outside the cubic system, also only three cases of natural order have been observed, namely in the antiferromagnetics MnF, FeF, and CoF (Erickson 1953) with tetragonal magnetic structure.

b) Superstructure sc-(001)

This monoplanar superstructure, determined unambiguously by our symbol, is shown in Fig. 6. It can be described by aid of the Luttinger vector

$$\boldsymbol{v} = a^{-2}\boldsymbol{c} \tag{4}$$

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Only one case of such superstructure seems to have been discovered hitherto, namely, that of the antiferromagnetic perovskite $LaMnO_3$ (Wollan and Koehler 1955). Unfortunately, this case does not satisfy our restricting condition. Probably we have here coplanar hybridized orbitals in magnetic ions, coupled ferromagnetically by semico-

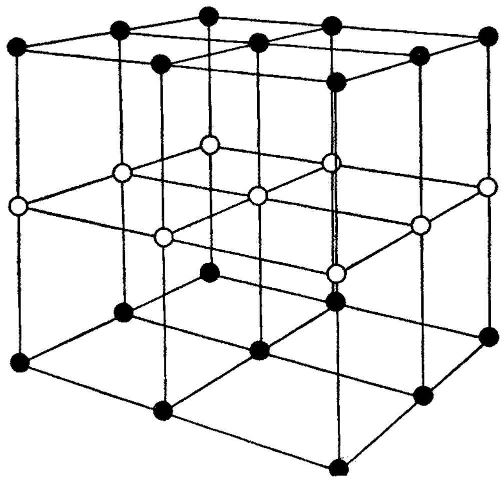

Fig. 6. Superstructure *sc*-(001)

valent exchange within the planes (001) and antiferromagnetically between two neighbouring (001) planes (Shull and Wollan 1956, page 217). In the hypothetical lattice in which our condition would be satisfied, the superstructure *sc*-(001) could presumably exist when the negative second interactions predominate over the positive (i.e. ferromagnetic) nearest interactions.

c) Superstructure *sc*-(110)-($1\bar{1}0$)

This biplanar superstructure is shown in Fig. 7. Here we have the choice between two Luttinger vectors:

$$\boldsymbol{v}_1 = a^{-2}(\boldsymbol{a} + \boldsymbol{b})$$

$$\boldsymbol{v}_2 = a^{-2}(\boldsymbol{a} - \boldsymbol{b}) \tag{5}$$

each of them being suitable for describing this superstructure. The order under consideration has been observed in (0.2 La — 0.8 Ca) MnO (Wollan and Koehler 1955), but it is not sure that our condition of isotropic interactions is fulfilled in the case of this compound. If this condition is satisfied, is seems that the superstructure *sc*-(110)-($\bar{1}\bar{1}0$) can exist when the negative second interactions predominate over the negative nearest ones.

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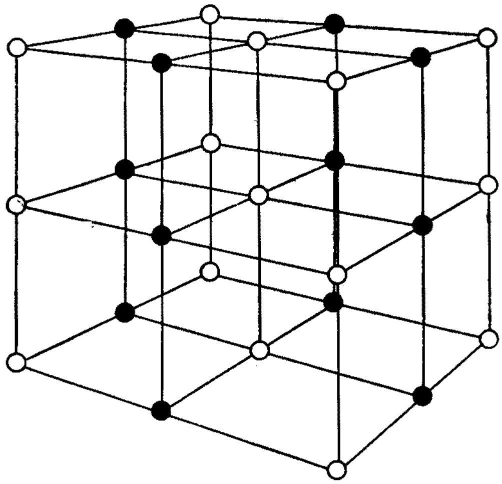

Fig. 7. Superstructure *sc*-(110)-(1$\bar{1}$0)

d) *Superstructure bcc*-(110)

This monoplanar superstructure has not been considered in the literature. It may be seen in Fig. 8. The corresponding Luttinger vector is

$$\boldsymbol{v} = a^{-2}\,(\boldsymbol{a} + \boldsymbol{b}) \tag{6}$$

No realisation of such order in nature is known at present. The possibility of its existence is rather questionable. A more detailed discussion on the problem of its existence will be given in a subsequent paper.

e) *Superstructure fcc*-(001)

This superstructure, shown in Fig. 9, may be described by the Luttinger vector

$$\boldsymbol{v} = 2a^{-2}\,\boldsymbol{c} \tag{7}$$

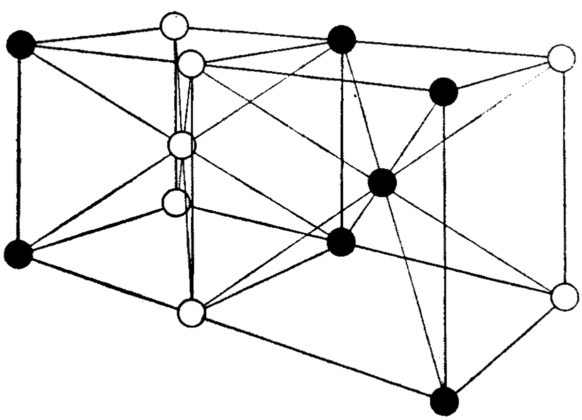

Fig. 8. Superstructure *bcc*-(110)

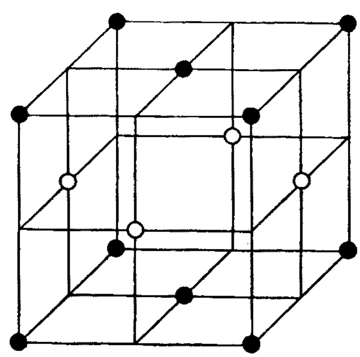

Fig. 9. Superstructure *fcc*-(001)

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It is known in the literature as the "order of the first kind" of *fcc* lattice (Van Vleck 1951, Smart 1952). This superstructure is realised in the antiferromagnetic substance $MnTe_2$ (Corliss et al. 1958). It seems very probable that in this case the nearest interactions are negative and the second ones most probably positive, though other relations may also be possible.

f) Superstructure fcc-(111)

This superstructure, shown in Fig. 10, has often been discussed in the literature. It may be described by the Luttinger vector

$$\boldsymbol{v} = (\boldsymbol{a} + \boldsymbol{b} + \boldsymbol{c})/a^2 \tag{8}$$

Smart (1952) and Labhart (1953) use the term: "order of the second kind" for this superstructure of *fcc* lattice. The superstructure *fc*-(111) has been found to occur in

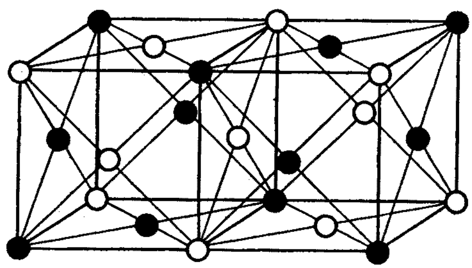

Fig. 10. Superstructure *fcc*-(111)

the well-known case of MnO, in other monoxides of transition metals, such as FeO, CoO, and NiO (Shull et al. 1951) and also in α-MnS (Corliss et al. 1956). It seems certain that such a superstructure is a consequence of considerable preponderance of negative second interactions.

2. *Anisotropic effects to be expected in cubic regular superstructures*

The possible superstructural anisotropic effects, of which a general discussion was given in Chapter V, can be predicted in detail for each of the individual cubic superstructures described in the previous section. The particular hypotheses listed below will be labelled P_1, P_2,... and so on. These hold only if the interactions in all directions are the same at the same distance.

a) Natural superstructures

Here the situation is well-known: the propagation of spin waves is governed by an isotropic dispersion law.

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b) Superstructure sc-(001)

Let us note that, within each ferromagnetic plane, the spins present in this case a quadratic arrangement implying rotational symmetry of the superstructural anisotropy. Moreover, the *fcc* substructures occuring here have the (001) order; therefore, the second interactions privilege the same direction [001] as the nearest ones. Hence, the following expectations on the superstructural dispersion anisotropy may be formulated:

P_1: The nearest interactions term in the dispersion law should present anisotropy with the preferred direction [001]. This anisotropy should present rotational symmetry about [001] i.e. should be uniaxial.

P_2: The second interactions term should present anisotropy with the same preferred direction [001] as the nearest interactions term. Likewise, this anisotropy will be presumably uniaxial (see P_7 below).

c) Superstructure sc-(110)-($1\bar{1}0$)

The privileged direction is here also the [001] one. In planes (001) the spins are arranged antiferromagnetically in quadratic layers and show the natural (two-dimensional) order. Each of two *fcc* substructures presents the (001) order, the same as in the previous case. Hence, we can predict the following behaviour of spin waves:

P_3: The nearest interactions term should present the uniaxial anisotropy with preferred direction [001]

P_4: The second interactions term should present the uniaxial anisotropy in the same direction [001]

d) Superstructure bcc-[110]

This superstructure undoubtedly privileges the direction [110]. The spin in each ferromagnetic plane (110) forms a centered rectangular array, so that the [110] axis is only a two-fold one. Therefore no rotational symmetry can be expected. Each of the two *sc* substructures shows the biplanar order (110)-($1\bar{1}0$) discussed above as case *c*). Hence, the following hypotheses may be made:

P_5: The nearest interactions term should be anisotropic with the most preferred direction [110].

P_6: The second interactions term should show uniaxial anisotropy preferring the direction [001].

e) Superstructure fcc-(001)

The privileged [001] direction is a four-fold axis. Each of the four *sc* substructures is a ferromagnetic one, and therefore privileges no direction whatever. Hence, our hypotheses in this case will be the following:

P_7: The nearest interactions term in the dispersion law should present uniaxial anisotropy with the preferred direction [001]

P_8: The second interactions term should be isotropic.

f) Superstructure fcc-(111)

The favoured [111] direction represents a three-fold axis, suggesting rotational symmetry of anisotropy. Each of the four *sc* substructures exhibits a natural order. Hence, the following hypotheses result:

P_9: The nearest interactions term should show uniaxial anisotropy in the direction [111].

P_{10}: The second interactions term should be isotropic.

The hypotheses P_1, P_7 and P_9 have already been confirmed by our previous approximate calculations (Cofta 1957).

The hypotheses as to spin waves behaviour presented here will be proved in a subsequent paper.

Acknowledgments

The author is indebted to Prof. dr S. Szczeniowski for his discussions and for correcting the present paper.

Notes added in proof.

Note I.

Our statement as to the small number of different non-regular superstructures, expressed in Ch. II, § 2 of the present paper, is not true. Recently, the author has discovered 5 other cases of such type.

Note II.

Quite recently the author has read the paper by Gersch and Koehler, published in the *J. Phys. Chem. Solids*, **5**, 180 (1958). As consistent with the Ising model, accurately the same 7 cubic superstructures presented here, has been obtained by Gersch and Koehler.

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